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Report AFOSR-91-0161

AFOSR-TR. : R 0002

**THE OVERALL RESPONSE OF COMPOSITE MATERIALS
UNDERGOING LARGE DEFORMATIONS**

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October 15, 1991

Final Technical Report for Period 1 February 1991 – 31 August 1992

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REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
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1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE October 15, 1992	3. REPORT TYPE AND DATES COVERED Final Technical Rpt. 2/1/91-8/31/92		
4. TITLE AND SUBTITLE The Overall Response of Composite Materials Undergoing Large Deformations		5. FUNDING NUMBERS G-AFOSR91-0161		
6. AUTHOR(S) Dr. Pedro Ponte-Castaneda				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Univ of Pennsylvania 133 South 36th St., Suite 300 Philadelphia, PA 19104-3246		8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NA Building 410, Bolling AFB DC 20332-6448		10. SPONSORING/MONITORING AGENCY REPORT NUMBER		
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION/AVAILABILITY STATEMENT APPROVED FOR PUBLIC RELEASE; DISTRIBUTION IS UNLIMITED.			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) This research deals with the theoretical prediction of the <i>effective</i> behavior of nonlinear composite materials undergoing large deformations. In particular, applications are envisaged to the high-temperature creeping behavior of metal/metal, metal/ceramic composites, and also of porous materials. Both isotropic and anisotropic configurations are considered including the technologically important case of fiber-reinforced composites, and the fundamentally important case of polycrystalline aggregates. The approach is based on new variational principles developed recently by the author (under AFOSR sponsorship), which allow the estimation of the overall behavior of a given nonlinear composite in terms of the effective properties of a suitably optimized linear comparison composite (with the same microstructure). The key advantage of the method is that it allows <i>direct</i> application of the extensive literature on linear composite materials, in the form of <i>estimates</i> and rigorous <i>bounds</i> , to obtain corresponding results for nonlinear composites. Additionally, the procedure is remarkably simple to implement, and the final results are usually expressed in terms of finite-optimization problems, which can be readily solved with modest computational effort. Recent progress include the application of the method to the determination of extremal yield surfaces for anisotropic rigid/plastic systems, and to the computation of estimates for the effective yield stress of polycrystalline aggregates.				
14. SUBJECT TERMS			15. NUMBER OF PAGES	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT	

Abstract

This research deals with the theoretical prediction of the *effective* behavior of nonlinear composite materials undergoing large deformations. In particular, applications are envisaged to the high-temperature creeping behavior of metal/metal, metal/ceramic composites, and also of porous materials. Both isotropic and anisotropic configurations are considered including the technologically important case of fiber-reinforced composites, and the fundamentally important case of polycrystalline aggregates. The approach is based on new variational principles developed recently by the author (under AFOSR sponsorship), which allow the estimation of the overall behavior of a given nonlinear composite in terms of the effective properties of a suitably optimized linear comparison composite (with the same microstructure). The key advantage of the method is that it allows *direct* application of the extensive literature on linear composite materials, in the form of *estimates* and rigorous *bounds*, to obtain corresponding results for nonlinear composites. Additionally, the procedure is remarkably simple to implement, and the final results are usually expressed in terms of finite-optimization problems, which can be readily solved with modest computational effort. Recent progress include the application of the method to the determination of extremal yield surfaces for anisotropic rigid/plastic systems, and to the computation of estimates for the effective yield stress of polycrystalline aggregates.

Research goals

The central goal of this project is to estimate the *effective* constitutive properties of *nonlinear* composite materials undergoing *large* deformations. Two types of large deformations are of particular interest: large *elastic* deformations, corresponding to materials such as polymeric composites, rubber foams and solid rocket fuel composites; and large *viscous* deformations, corresponding to the high-temperature creeping, or to the dynamic plastic, deformation of metals. The main emphasis for the funding period was to concentrate on the effective behavior of *anisotropic* viscoplastic composites. The effect of anisotropy is considered both globally and locally. Thus, the effect of isotropic inclusions of different shapes on the global anisotropy of the composite is considered, as well as the effect of local anisotropic behavior for the inclusions. Examples of the results obtained are prediction for the effective constitutive relations for fiber-reinforced, metal-matrix composites and estimates for the yield strength of isotropic polycrystalline aggregates of anisotropic single ductile crystals.

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BACKGROUND

There is a fairly recent, but growing literature in the field of composite materials that is concerned with the prediction of the effective behavior of nonlinear composites. To date most of the work on the subject has made use of *ad hoc* models for certain classes of composites, as well as of heavy numerical computations for composites with periodic microstructures. For example, Duva and Hutchinson [11] determined *dilute* estimates for porous creeping composites by making use of the results of Budiansky, Hutchinson and Slutsky [4] for a single void in an infinite matrix of power-law viscous material. Duva [10] proposed a *differential self-consistent* estimate to model the reinforcing effect of rigid particles in a power-law creeping matrix. More recently, Bao, Hutchinson and McMeeking [2] proposed *generalized self-consistent* estimates for the yield stress of two-phase, rigid/perfectly plastic composites. On the other hand, Christman, Needleman and Suresh [5] and Bao, Hutchinson and McMeeking [1] carried out computations on the unit cell of a *periodic* microstructure to model the effect of particle size and shape on the effective behavior of rigidly reinforced power-law, metal-matrix composites.

As far as rigorous methods for estimating the effective behavior of composites with *random* microstructures (real composite materials are hardly periodic), the only method available until recently was the Talbot and Willis [27] extension of the Hashin-Shtrikman variational principles [14] to composites with nonlinear behavior. Ponte Castañeda and Willis [26] applied this procedure to nonlinearly viscous materials, and gave the first rigorous bounds and self-consistent estimates for the effective properties of composites in power-law creep. Later, Ponte Castañeda [17] developed corresponding bounds in the context of finite elasticity. Recently, under the sponsorship of AFOSR, Ponte Castañeda [18] developed a new general method for estimating and bounding the effective behavior of nonlinear composites. The method expresses the effective behavior of nonlinear composites in terms of an optimization problem involving the effective behavior of linear composites materials with similar microstructures. This is very useful because of the large body of results in the form of bounds and estimates of different types that are currently available for linear composites. In particular, the linear Hashin-Shtrikman [14] bounds may be used to generate corresponding nonlinear bounds. The method and some of its applications to specific types of composites are discussed in the following sections.

ACCOMPLISHMENTS

Introduction

In this section, we give a brief account of the theoretical foundations of the new variational method that has been developed by the author to predict the effective behavior of nonlinear composites [18,19,20]. In addition, we give sample applications to isotropic two-phase creeping composites and to two-phase rigid-perfectly plastic laminated composites. In particular, we note that our results for simple two-phase, power-law creep composites compare very favorably with the results of other authors obtained by means of sophisticated large-scale computations for periodic composites. Our results also have the advantage that they can often be given simple analytical forms. For example, we have been able to obtain an improved characterization of porous isotropic and transversely isotropic materials; in fact, because of the bounding properties of our results, we are able to show that the commonly used Gurson [12] model is not appropriate outside the range of dilute porosity and high-triaxiality for which it was originally developed [22]. Similarly, we have been able to obtain yield surfaces for isotropic and transversely isotropic composites with rigid/perfectly plastic phases. These yield surfaces have the features (flat sectors) predicted qualitatively by Hashin [13] and others. Other particular systems are considered in detail in the references.

For the convenience of the reader, the relevant references are included in the Appendix. Reference [20] deals with the development of the new variational principles first studied (under prior AFOSR sponsorship) in [18]. In addition, bounds of the Hashin-Shtrikman type are given for general multiphase ductile composites with local and overall isotropy. The expression for the bounds given in this reference is extremely simple involving finite-dimensional optimization problems. Also in this reference, nonlinear bounds of the Beran type are given for the first time for multiphase incompressible composites with isotropic phases. Applications to viscoplastic composites with isotropic phases, but anisotropic overall behavior is summarized in reference [21]. More detail results are given in [6] for laminated ductile composites, in [7] for elastoplastic fiber-reinforced composites, and in [16] for ductile composites with distributions of aligned rigid spheroidal inclusions. In references [8, 25], effective yield surfaces are determined for rigid-perfectly plastic composites with overall isotropic, laminated and fiber-reinforced microstructures. Finally, reference [23] deals with a mathematical proof of the variational principle in the context of electrostatics, and reference [24] deals with the application of the variational principles to ferroelectric (or ferromagnetic) composites.

The last part of the work, which is not yet completed, includes the extension of the method to composites with anisotropic phases, such as polycrystalline aggregates. For this special class of composite materials, upper bound estimates of the Voigt type, as well as self-consistent estimates, are available through the classical works of Bishop and Hill [3] and Hutchinson [15]. We seek to improve on these results by computing upper bound estimates of the Hashin-Shtrikman type [14]. The results will be reported in [9]

The method

Effective properties

Consider a two-phase heterogeneous material occupying a region in space of unit volume Ω . We characterize the local stress potential $\phi(\boldsymbol{\sigma}, \mathbf{x})$ of the material in terms of the homogeneous phase potentials $\phi^{(r)}(\boldsymbol{\sigma})$ via the relation

$$\phi(\boldsymbol{\sigma}, \mathbf{x}) = \sum_{r=1}^2 \chi^{(r)}(\mathbf{x}) \phi^{(r)}(\boldsymbol{\sigma}), \quad (1)$$

where the $\chi^{(r)}$ ($r = 1, 2$) are the characteristic functions of the two phases. We will assume that the two phases are nonlinear, incompressible and isotropic, so that the potentials $\phi^{(r)}(\boldsymbol{\sigma})$ depend only on the effective stress $\sigma_e = \sqrt{\frac{3}{2} \mathbf{S} \cdot \mathbf{S}}$, where $\mathbf{S}$ is the deviator of $\boldsymbol{\sigma}$. Thus, we write

$$\phi^{(r)}(\boldsymbol{\sigma}) = f^{(r)}(\sigma_e), \quad (2)$$

where the scalar-valued functions $f^{(r)}$ are assumed to satisfy some rather weak hypothesis. For example, $f^{(r)}$ may be chosen to have the power-law form

$$f(\sigma_e) = \frac{1}{n+1} \dot{\epsilon}_o \sigma_o \left(\frac{\sigma_e}{\sigma_o} \right)^{n+1} \quad (3)$$

with n , σ_o and $\dot{\epsilon}_o$ denoting material constants with different values in each phase.

The local constitutive relation for the creeping material is given by

$$\dot{\boldsymbol{\epsilon}} = \frac{\partial \phi}{\partial \boldsymbol{\sigma}}(\boldsymbol{\sigma}, \mathbf{x}), \quad (4)$$

where $\dot{\boldsymbol{\epsilon}}$ is the rate-of-deformation (strain-rate) tensor.

Letting Σ and $\dot{\mathbf{E}}$ denote the average values of the stress and strain-rate fields, respectively, the effective, or overall, behavior of the heterogeneous creeping material is given by the relation

$$\mathbf{E} = \frac{\partial \Phi}{\partial \Sigma}, \quad (5)$$

where the effective energy of the composite Φ follows from the principle of minimum complementary energy, which can be stated in the form

$$\Phi(\Sigma) = \min_{\sigma \in S(\Sigma)} \int_{\Omega} \phi(\sigma, \mathbf{x}) d\mathbf{x}, \quad (6)$$

where $S(\Sigma) = \{\sigma \mid \sigma_{ij,j} = 0 \text{ in } \Omega, \text{ and } \sigma_{ij} n_j = \Sigma_{ij} n_j \text{ on } \partial\Omega\}$ is the set of statically admissible stresses.

In this work, we are interested in predicting the effective behavior of two-phase heterogeneous materials, as described above. We further restrict our consideration to composites with phases in given volume fractions

$$c^{(r)} = \int_{\Omega} \chi^{(r)}(\mathbf{x}) d\mathbf{x}, \quad (7)$$

New variational principles

The new variational principles [18, 19, 20] are based on a representation of the potential for the nonlinear isotropic material in terms of the potentials of a family of linear *comparison* materials. This representation is obtained with the help of the Legendre transformation. Thus, for a homogeneous nonlinear material with "superquadratic" growth in its potential ϕ , and certain additional convexity hypothesis, we have that

$$\phi(\sigma, \mathbf{x}) = \max_{\mu > 0} \{ \phi_o(\sigma, \mathbf{x}) - V(\mu, \mathbf{x}) \}, \quad (8)$$

where

$$V(\mu, \mathbf{x}) = \max_{\sigma} \{ \phi_o(\sigma, \mathbf{x}) - \phi(\sigma, \mathbf{x}) \} \quad (9)$$

and where ϕ_o is the potential of a linear comparison material with shear modulus μ . Note that in the above optimizations, the position vector $\mathbf{x}$ is fixed.

The new variational principle is obtained by averaging the relation (8) and the complementary energy principle (6), and can be expressed in the form

$$\Phi(\Sigma) = \max_{\mu(\mathbf{x}) \geq 0} \left\{ \Phi_o(\Sigma) - \int_{\Omega} V(\mu, \mathbf{x}) d\mathbf{x} \right\}, \quad (10)$$

where

$$\Phi_o(\Sigma) = \min_{\sigma \in S(\Sigma)} \int_{\Omega} \phi_o(\sigma, \mathbf{x}) d\mathbf{x} \quad (11)$$

is the effective potential of a linear heterogeneous comparison material with local potential ϕ_o and arbitrarily variable shear moduli $\mu(\mathbf{x})$.

The variational principle described by (10) and (11) can be given the interpretation of first solving a linear problem for an arbitrarily heterogeneous *linear* material, and then optimizing with respect to the variations in moduli to account for the nonlinearity in the

actual material. This suggests that if the fields happen to be constant over the nonlinear phases in the actual composite, then the variable moduli $\mu(\mathbf{x})$ can be replaced by constant moduli over each phase, *i.e.* by $\mu^{(1)}$ and $\mu^{(2)}$ in phase 1 and phase 2, respectively. More generally, however, we have the following lower bound for Φ , namely,

$$\Phi_-(\Sigma) = \max_{\mu^{(1)}, \mu^{(2)} \geq 0} \{\Phi_o(\Sigma) - c^{(1)}V^{(1)}(\mu^{(1)}) - c^{(2)}V^{(2)}(\mu^{(2)})\}, \quad (12)$$

where $V^{(1)}$ and $V^{(2)}$ correspond to relation (9) evaluated for each of the homogeneous phase potentials $\phi^{(1)}$ and $\phi^{(2)}$, and Φ_o is the effective energy of an isotropic two-phase linear composite with phase moduli $\mu^{(1)}$ and $\mu^{(2)}$ in volume fractions $c^{(1)}$ and $c^{(2)}$, respectively. This latter expression has the advantage over the previous more general relation that it involves only a finite-dimensional optimization problem, and can be further used in conjunction with bounds and estimates for the linear composite to induce corresponding results for the nonlinear composite. We illustrate these possibilities in the next section for a general two-phase incompressible composite.

Sample application to two-phase creeping systems

The estimate (12) of the previous section for the effective energy of the nonlinear incompressible composite can be used in conjunction with the well-known Hashin-Shtrikman bounds for the effective properties of the linear comparison composite to yield estimates for the effective potential of the nonlinear composite. It is shown in reference [20] that the result of this calculation takes on the simple form

$$\Phi_{HS}(\Sigma) = \min_{\omega \geq 0} \{c^{(1)}f^{(1)}(S^{(1)}) + c^{(2)}f^{(2)}(S^{(2)})\}, \quad (13)$$

where $f^{(1)}$ and $f^{(2)}$ characterize the phase behaviors, and $S^{(1)}$ and $S^{(2)}$ are simple expressions of ω involving the phase volume fractions $c^{(1)}$ and $c^{(2)}$, and the overall effective stress applied to the composite Σ_e . For instance, if $\mu^{(1)} \geq \mu^{(2)}$ and the Hashin-Shtrikman lower bound is used for Φ_o , the corresponding expressions for $S^{(1)}$ and $S^{(2)}$ become

$$S^{(1)} = \sqrt{(1 - c^{(2)}\omega)^2 + \frac{2}{3}c^{(2)}\omega^2} \Sigma_e \quad \text{and} \quad S^{(2)} = (1 + c^{(1)}\omega)\Sigma_e, \quad (14)$$

Depending on the relative strengths of the two nonlinear potentials $f^{(1)}$ and $f^{(2)}$, the result of putting (14) into (13) would either lead to a rigorous lower bound for the nonlinear potential Φ , or alternatively, to an upper "estimate" for Φ . Other examples are detailed in the Appendices.

Sample application to two-phase rigid-perfectly plastic laminated composite

The above method [20] for estimating the effective behavior of nonlinear composites has been extended recently to predict the effective *strength* of rigid-perfectly plastic composites [25]. An sample result is given in Figure 1, where the overall yield strength domain of a laminated composite, made up of two phases with simple incompressible, isotropic yield criteria (of the Mises type), is plotted as a function of the two independent stress modes: longitudinal shear (Σ_n) and combinations of axisymmetric and transverse shear (Σ_t) (relative to the perpendicular direction to the layers). We note that the region of stress space described by the new exact estimate is significantly greater than the so-called "quadratic interpolation" of Hill (a quadratic interpolation between the arithmetic mean of the strength of the two phases and the strength of the weaker phase). In addition the new exact estimate has the feature that it possesses a flat sector (see [13]). This is indicative of the severe anisotropies that may arise in highly nonlinear composites.

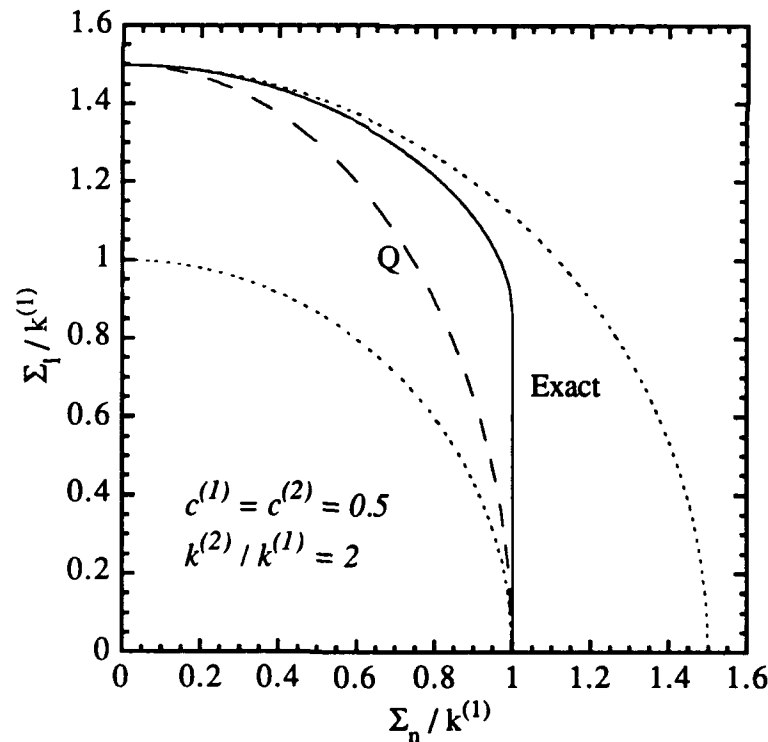


Figure 1. Estimates for the failure strength of a laminated composite, where the phases have a strength ratio $k^{(2)} / k^{(1)} = 2$ and equal volume fractions $c^{(1)} = c^{(2)} = 0.5$. The continuous curve corresponds to the new exact estimates; the long-dash curve to the quadratic interpolation of Hill; and the top and bottom short-dash curves to Bishop-Hill-type of estimates, respectively.

RESEARCH PLANS FOR THE FUTURE

After completing our study of the effective behavior of polycrystalline aggregates, we will begin a new study of damage localizations in composites, which may arise because of the finite kinematics, or alternatively through thermomechanical, or other micro-mechanisms. These are important in the development of realistic models for the behavior of composite materials, as discussed in the recent "Panel Discussion on Homogenization and Damage Localization in Heterogeneous Media" in last year's *Contractors Meeting on Mechanics of Materials* (Dayton, October 1991). Thus, we are in the process of writing a proposal for AFOSR sponsorship of this new, but closely related project.

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Prior grant (AFOSR 89-0288)

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"Estimates for the yield strength of two-phase, perfectly plastic composites" (with G. deBotton).

"Constitutive models for ductile solids reinforced by rigid spheroidal inclusions" (with G. Li and A.S. Douglas).

29th Annual Meeting of the Society of Engineering Science,
University of California, San Diego, September 14-16, 1992.

"The effect of texture development on the localization of porous metals" (with M. Zaidman).

"The elastic plastic behavior of fiber composites" (with G. deBotton)

"Elastic-plastic interfacial crack growth" (with P. A. Mataga)

18th. International Congress of Theoretical and Applied Mechanics,
Technion, Haifa (Israel), August 22-28, 1992.

"The effective behavior of nonlinear composites: A comparison between methods" (with J.R. Willis).

Symposium on Continuum Models of Discrete Systems 7
Paderborn (Germany), June 15-19, 1992.

"The effective properties of nonlinear heterogeneous systems,"

"The effective properties of anisotropic composites,"

"Effective dielectric properties of nonlinear composites" (with G. Li & A.S. Douglas),

"Slow crack growth under remote mixed mode conditions" (with P.A. Mataga & K. Bose),

"Constitutive models for porous metals deformed at high rates" (with M. daSilva & K.T. Ramesh)

28th Annual Meeting of the Society of Engineering Science,
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"On the overall properties of composite materials."

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Washington, July 8-12, 1991.

Invited Seminars

"The effective properties of nonlinear composites."

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"Interfacial crack growth in elastoplastic solids."

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"Bounds and exact estimates for the properties of nonlinear heterogeneous systems."

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Ph.D. Students

Gal de Botton. Ph.D. expected December 1992. Thesis title: "Elastoplastic constitutive relations for nonlinear anisotropic composites". Will start position as Assistant Professor in the Department of Mechanical Engineering of the BenGurion University, Beer-Sheva (Israel) on January 1, 1993.

Appendices

Reference [20]

NEW VARIATIONAL PRINCIPLES IN PLASTICITY AND THEIR APPLICATION TO COMPOSITES MATERIALS

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(Received 15 August 1991; in revised form 27 January 1992)

ABSTRACT

IN THIS PAPER, a variational method for bounding the effective properties of nonlinear composites with isotropic phases, proposed recently by PONTE CASTAÑEDA (*J. Mech. Phys. Solids* 39, 45, 1991), is given full variational principle status. Two dual versions of the new variational principle are presented and their equivalence to each other, and to the classical variational principles, is demonstrated. The variational principles are used to determine bounds and estimates for the effective energy functions of nonlinear composites with prescribed volume fractions in the context of the deformation theory of plasticity. The classical bounds of Voigt and Reuss for completely anisotropic composites are recovered from the new variational principles and are given alternative, simpler forms. Also, use of a novel identity allows the determination of simpler forms for nonlinear Hashin-Shtrikman bounds and estimates for isotropic, particle-reinforced composites, as well as for transversely isotropic, fiber-reinforced composites. Additionally, third-order bounds of the Beran type are determined for the first time for nonlinear composites. The question of the optimality of these bounds is discussed briefly.

1. INTRODUCTION

A VARIATIONAL PROCEDURE for bounding the effective properties of nonlinear composites with isotropic phases has been proposed recently by PONTE CASTAÑEDA (1991a). The procedure allows the estimation of the effective constitutive behavior of a given nonlinear composite in terms of the effective behavior of an appropriately optimized, linear *comparison composite* with the same underlying microstructures as the nonlinear composite. The general nonlinear estimate can be expressed in the form of a lower bound for the effective complementary-energy function of the composite, in such a way that if a lower bound is available for the effective energy of the linear comparison composite, a corresponding lower bound may be deduced for the nonlinear composite. On the other hand, an upper bound for the effective complementary-energy function of the linear comparison composite cannot yield in general an upper bound for the nonlinear composite. In this sense, the procedure is similar to a nonlinear generalization of the Hashin-Shtrikman procedure, developed by TALBOT and WILLIS (1985), following WILLIS (1983), which makes use of a linear *homogeneous comparison material* to obtain bounds and estimates of the Hashin-Shtrikman type for nonlinear composites. The variational procedure of PONTE CASTAÑEDA (1991a), however, allows the use of *any* linear bound, or estimate, for the

comparison composite to yield a corresponding nonlinear bound, or estimate, for the nonlinear composite.

In this paper, we strengthen the variational procedure proposed by PONTE CASTAÑEDA (1991a) to a full variational principle, which under appropriate hypotheses can be shown to be *completely equivalent* to the classical minimum principles of potential and complementary energy. This goal is accomplished *essentially* by allowing a comparison composite to have arbitrarily varying microstructure, instead of being constrained to have the same (piecewise constant) microstructure as the nonlinear composite. Naturally, the variational procedure of PONTE CASTAÑEDA (1991a) results in an *approximation*; but, more generally, the new variational principles allow the imputation of the exact properties of certain classes of special microstructures. For example, DEBORTON and PONTE CASTAÑEDA (1991) have made use of the new variational principles to determine simple expressions for the effective behavior of nonlinear laminates. Here, we will make use of the exact version of the new variational principles to demonstrate the optimality of some of the nonlinear bounds developed by means of the *approximate* version of the variational principle.

The paper is organized as follows. First, the definition of the effective properties of nonlinear composites is reviewed. Then, two dual versions of the new variational principle are presented, with rigorous proofs of their respective equivalence to each other, and to the classical variational principles. In the following section, we apply the *approximate* version of the new variational principle, together with a novel identity introduced by DEBORTON and PONTE CASTAÑEDA (1991), to develop remarkably simple forms for the Voigt and Hashin-Shtrikman lower bounds for the effective complementary-energy function of nonlinear composites with phases in prescribed volume fractions. Results are given for multiple-phase, compressible composites, but a particularly simple form for the effective energy of the nonlinear composite is obtained for the case of two-phase, incompressible composites. This latter result is expressed in terms of a one-dimensional optimization problem [see (4.34)], and generalizes the findings of PONTE CASTAÑEDA (1991b) for two-phase, incompressible composites with a linear and a nonlinear phase. Hashin-Shtrikman bounds are given not only for isotropic nonlinear composites, but also for transversely isotropic, fiber-reinforced materials [see (4.36)]. Additionally, a *third-order* bound of the Beran type is given for isotropic composites [see (4.41)]. To the knowledge of the author, this is the first higher-order bound ever produced in the context of nonlinear composites, and illustrates the versatility of the new variational principles. Finally, in this section, we give a derivation of the Reuss upper bound for the effective complementary-energy function of the nonlinear composite by means of the *exact* version of the new variational principle. This serves to demonstrate that the exact version of the variational principle is able to deliver upper bounds, which cannot be obtained from the approximate version of the variational principle. Although we were unable to determine rigorous upper bounds of the Hashin-Shtrikman and Beran varieties, we produce some arguments justifying the application of the approximate version of the variational procedure in connection with upper bounds for the linear comparison composite to obtain "upper estimates" (not bounds) for the effective properties of certain classes of nonlinear composites. The last subsection deals briefly with the question of attainability of the nonlinear bounds.

In the development of the bounds, we will be guided by the physical requirements of the deformation theory of plasticity, or nonlinear infinitesimal elasticity, although completely analogous results will also hold in the context of nonlinear creep. Additionally, we could follow the arguments presented by DUVA and HUTCHINSON (1984) to make use of the effective complementary-energy function of a given deformation-theory composite to obtain an *approximate* form for the yield function of a flow-theory composite exhibiting the same microstructure (and coinciding plastic response under proportional loading) as the deformation-theory solid. The case of ideal plasticity could also be considered formally as a special case of a pure-power creeping solid in the limit as the appropriately defined power exponent approaches infinity. However, extreme care must be exercised in the interpretation of the concomitant results because of the well-known lack of uniqueness in ideal plasticity, which among other things complicates the definition of effective properties. Because of these difficulties, in this paper, we will exclude from our direct consideration the effective behavior of ideally plastic composites.

2. EFFECTIVE PROPERTIES

We consider a heterogeneous solid occupying a domain of unit volume $\Omega \in R^3$, with boundary $\partial\Omega$. The nonlinear constitutive behavior of the solid is characterized by an energy density function, $w(x, \epsilon)$, depending on the position vector x and the strain field $\epsilon(x)$, in such a way that the stress field $\sigma(x)$ is given by

$$\sigma(x) = \partial_\epsilon w(x, \epsilon). \quad (2.1)$$

Here ∂_ϵ denotes the usual differentiation with respect to ϵ , if w is differentiable. More generally, ∂_ϵ can be given the interpretation of the subdifferential of convex analysis (EKKLAND and TEMAN, 1974; § 1.5).

Following HILL (1963), we define the *effective* constitutive behavior of the heterogeneous solid in terms of the analogous relation

$$\bar{\sigma} = \partial_\epsilon \bar{w}(\bar{\epsilon}), \quad (2.2)$$

where $\bar{\sigma}$ denotes the mean value of the stress field over Ω , and $\bar{w}$ refers to the normalized energy (recall that Ω has unit volume) of the solid when subjected to the uniform boundary condition

$$u = \bar{\epsilon}x, \quad x \in \partial\Omega, \quad (2.3)$$

where u is the displacement field, and $\bar{\epsilon}$ is a constant, symmetric tensor. We note that under this boundary condition, the mean value of the strain over Ω is precisely $\bar{\epsilon}$.

The *effective* energy of the solid, $\bar{w}$, can be obtained directly from the principle of minimum potential energy via the relation

$$\bar{w}(\bar{\epsilon}) = \min_{u \in K(\bar{\epsilon})} W(u), \quad (2.4)$$

where

$$W(\varepsilon) = \int_{\Omega} w(\mathbf{x}, \varepsilon(\mathbf{x})) \, d\mathbf{x} \quad (2.5)$$

is the pertinent energy functional, and

$$K(\bar{\varepsilon}) = \{\varepsilon \mid \varepsilon = \{\nabla u + (\nabla u)^T\} \text{ in } \Omega, \text{ and } u = \bar{\varepsilon} \mathbf{x} \text{ on } \partial\Omega\} \quad (2.6)$$

is the set of kinematically admissible strain fields.

In plasticity, it is usually more convenient to express the constitutive relation of the solid in terms of a complementary-energy density function, $u = w^*$, where w^* is the Legendre transform, or polar function of convex analysis (Ekeland and Temam, 1974; §1.4.1), defined by

$$w^*(\mathbf{x}, \sigma) = \sup_{\varepsilon} \{\varepsilon \cdot \sigma - w(\mathbf{x}, \varepsilon)\}. \quad (2.7)$$

Thus, the constitutive relation of the heterogeneous solid, in its local form, can be written

$$\varepsilon(\mathbf{x}) = \partial_{\varepsilon} u(\mathbf{x}, \sigma). \quad (2.8)$$

Correspondingly, the effective constitutive relation of the solid (in its global form) is given by

$$\bar{\varepsilon} = \partial_{\bar{\varepsilon}} \bar{U}(\bar{\sigma}), \quad (2.9)$$

where $\bar{U}$ corresponds to the normalized complementary energy that would exist in the heterogeneous solid if it was subjected to a uniform constraint on $\partial\Omega$, such that

$$\sigma \mathbf{n} = \bar{\sigma} \mathbf{n}, \quad (2.10)$$

where $\bar{\sigma}$ is a constant, symmetric tensor. Thus, the effective complementary energy of the composite, $\bar{U}$, can be described in terms of the principle of minimum complementary energy via the relation

$$\bar{U}(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} U(\sigma), \quad (2.11)$$

where

$$U(\sigma) = \int_{\Omega} u(\mathbf{x}, \sigma(\mathbf{x})) \, d\mathbf{x} \quad (2.12)$$

is the complementary energy functional, and

$$S(\bar{\sigma}) = \{\sigma \mid \nabla \cdot \sigma = 0 \text{ in } \Omega, \text{ and } \sigma \mathbf{n} = \bar{\sigma} \mathbf{n} \text{ on } \partial\Omega\} \quad (2.13)$$

is the set of statically admissible stress fields.

We will assume that w , and therefore u , are convex functions of ε and σ , respectively, and that they satisfy certain growth conditions to be specified later. Further, by convex duality (Ekeland and Temam, 1974; §1.4.2), we have that

$$w(\mathbf{x}, \varepsilon) = \sup_{\sigma} \{\varepsilon \cdot \sigma - w^*(\mathbf{x}, \sigma)\}. \quad (2.14)$$

Then, the effective energy functions $\bar{W}$ and $\bar{U}$ are convex (see Ponte Castañeda and Willis, 1988), but on the other hand, we have that, in general (Willis, 1989),

$$\bar{W}(\bar{\varepsilon}) \geq \sup_{\sigma} \{\bar{\varepsilon} \cdot \sigma - \bar{U}(\sigma)\}. \quad (2.15)$$

The reason for the inequality is that the two definitions of effective properties (in terms of $\bar{W}$ and $\bar{U}$) correspond to different boundary conditions (uniform traction versus uniform displacement). However, for a composite solid, obtained in the limit as the size of the typical heterogeneity in the solid becomes small compared to the size of the specimen, the two definitions agree, and an equality holds in (2.15).

3 THE NEW VARIATIONAL PRINCIPLES

In the development of the proposed variational principles, we will consider two dual versions of essentially the same result, depending on whether we start from the minimum potential energy formulation (2.4), or from the minimum complementary energy formulation (2.11). In both cases, we will assume that the solid is *locally isotropic*, so that we can write

$$w(\mathbf{x}, \varepsilon) = \phi(\mathbf{x}; \varepsilon, \varepsilon_m), \quad (3.1)$$

where $\phi: \Omega \times R^+ \times R^+ \rightarrow R^+$ is a nonnegative function, that is convex and continuous in its last two arguments, and satisfies the condition that $\phi(\mathbf{x}; 0, 0) = 0$ for all $\mathbf{x}$. Here R^+ is the set of the extended non-negative reals (R is the set of extended reals), and

$$\varepsilon_m = \frac{1}{2} |\text{tr } \varepsilon| \quad \text{and} \quad \varepsilon_e = \sqrt{\frac{2}{3}} \varepsilon \cdot \varepsilon, \quad (3.2)$$

are the *mean* and *effective* (in their plasticity denotations) strains, respectively, where $\varepsilon = \varepsilon - \varepsilon_m \mathbf{I}$ is the deviator strain tensor. We note that the above conditions on ϕ are consistent with the assumed convexity of w , and that the form (3.1) is *not* the most general form for the energy function of a nonlinear isotropic solid (we could also have dependence on the determinant of ε), but this form is still general enough to cover the usual plasticity models of interest here. Also, we emphasize that with the above definitions both ε_m and ε_e are non-negative.

We note here for later reference that under the above hypotheses on w , its polar function can be expressed (see Ekeland and Temam, 1974; §1.4.3 for an analogous result, and also Appendix I for a definition of $\phi^*: \Omega \times R^+ \times R^+ \rightarrow R^+$) in terms of

$$w^*(\mathbf{x}, \sigma) = \phi^*(\mathbf{x}; \frac{1}{2} \sigma_e, \frac{1}{3} \sigma_m), \quad (3.3)$$

where

$$\sigma_m = \frac{1}{3} |\text{tr } \sigma| \quad \text{and} \quad \sigma_e = \sqrt{\frac{2}{3}} \sigma \cdot \sigma \quad (3.4)$$

are respectively the mean and effective stresses, and $\sigma = \sigma - \sigma_m \mathbf{I}$ is the deviator stress tensor. Alternatively, we may represent $u = w^*$ by

$$u(\mathbf{x}, \sigma) = \psi(\mathbf{x}; \sigma_e, \sigma_m), \quad (3.5)$$

where ψ is a non-negative function, convex in its last two arguments, and such that $\psi(\mathbf{x}; 0, 0) = 0$ for all $\mathbf{x}$. These properties follow from simple properties of the polar

function (see EKELAND and TEMAM, 1974; § 1.4). Finally, we have that $w = u^*$ can be expressed in the form

$$u^*(x, \varepsilon) = \psi^*(x; \frac{1}{2}\varepsilon, 3\varepsilon_m). \quad (3.6)$$

3.1. Minimum potential energy formulation

The new variational principle relies on a quadratic change of variables $u = h(\varepsilon)$, with $h: R^+ \rightarrow R^+$ given by $h(\varepsilon) = \varepsilon^2$. We can then define a function $f: \Omega \times R^+ \times R^+ \rightarrow R^+$ such that

$$f(x; u, u_m) = \phi(x; h^{-1}(u), h^{-1}(u_m)). \quad (3.7)$$

By the properties of ϕ , f is a non-negative function with the same dependence on x as ϕ , and such that $f(x; 0, 0) = 0$. We note further that f is continuous, but not necessarily convex (in its last two arguments). In fact, because we are interested in the application of these results to plasticity, we will usually assume that f is concave, or at least that it has concave growth. Thus, for example, for an incompressible power-law material, $w \sim \varepsilon^{(n+1)/2}$ ($1 \leq n < \infty$), and therefore $f \sim u^{(n+1)/2}$, which is a concave function, even though w , itself, is convex.

We can then define (see Appendix I, and VAN TIEL, 1984; § 7.14) the concave polar function $f_*: \Omega \times R^+ \times R^+ \rightarrow R^+$ by

$$f_*(x; p, p_m) = \inf_{u, u_m \geq 0} \{u, p + u_m p_m - f(x; u, u_m)\}, \quad (3.8)$$

and therefore, we have that

$$f(x; u, u_m) \leq \inf_{p, p_m \geq 0} \{u, p + u_m p_m - f_*(x; p, p_m)\}, \quad (3.9)$$

with equality if f is concave.

We proceed by defining the functional F , induced by f , as follows

$$F(u, u_m) = \int_{\Omega} f(x; u(x), u_m(x)) dx, \quad (3.10)$$

and we note that (see EKELAND and TEMAM, 1974; § IX.2.1, 2) that the (concave) polar of the functional F , given by

$$F_*(p, p_m) = \inf_{u, u_m \geq 0} \left\{ \int_{\Omega} [u, p(x) + u_m(x)p_m(x)] dx - F(u, u_m) \right\}, \quad (3.11)$$

can alternatively be represented in terms of f_* via

$$F_*(p, p_m) = \int_{\Omega} f_*(x; p(x), p_m(x)) dx, \quad (3.12)$$

with an analogous representation for the bipolar function F_{**} in terms of f_{**} .

Thus, rewriting the minimum energy principle (2.4) in the form

$$\bar{W}(\varepsilon) = \min_{u, u_m \in K} F(u, u_m), \quad (3.13)$$

where K' is the set of admissible fields $u_r(x)$, $u_m(x)$ induced by K in (2.6), we arrive by means of (3.9) to the inequality

$$\bar{W}(\varepsilon) \leq \bar{W}_+(\varepsilon), \quad (3.14)$$

where

$$\bar{W}_+(\varepsilon) = \min_{u_r, u_m \in K'} \inf_{p_r, p_m \geq 0} L(u_r, u_m; p_r, p_m), \quad (3.15)$$

and L is the functional given by

$$L(u_r, u_m; p_r, p_m) = \int_{\Omega} [u_r(x)p_r(x) + u_m(x)p_m(x)] dx - \int_{\Omega} f_*(x; p_r(x), p_m(x)) dx. \quad (3.16)$$

We emphasize that for concave f , $\bar{W}_+(\varepsilon) = \bar{W}(\varepsilon)$.

Next, we simplify the expression for $\bar{W}_+(\varepsilon)$, by noting that

$$\min_{u_r, u_m \in K'} \inf_{p_r, p_m \geq 0} L(u_r, u_m; p_r, p_m) = \inf_{p_r, p_m \geq 0} \inf_{u_r, u_m \in K'} L(u_r, u_m; p_r, p_m), \quad (3.17)$$

since the order in which infima are evaluated can be interchanged.

It follows that

$$\bar{W}_+(\varepsilon) = \inf_{p_r(x), p_m(x) \geq 0} \left\{ \bar{W}_0(\varepsilon) - \int_{\Omega} f_*(x; p_r(x), p_m(x)) dx \right\}, \quad (3.18)$$

where

$$\bar{W}_0(\varepsilon) = \inf_{u_r, u_m \in K'} \left\{ \int_{\Omega} [u_r(x)p_r(x) + u_m(x)p_m(x)] dx \right\}. \quad (3.19)$$

By introducing the variables $\mu_0 = \frac{1}{2}p_r$ and $\kappa_0 = \frac{1}{2}p_m$, and expressing u_r and u_m in terms of ε and ε_m , respectively, we are able to rewrite the integrand in (3.19) in terms of the quadratic energy function

$$w_0(x, \varepsilon) = \phi_0(x; \varepsilon, \varepsilon_m) = \frac{1}{2}\mu_0(x)\varepsilon^2 + \frac{1}{2}\kappa_0(x)\varepsilon_m^2, \quad (3.20)$$

which corresponds to a linear-elastic, isotropic, heterogeneous solid with non-negative, but otherwise arbitrary, shear modulus $\mu_0(x)$ and bulk modulus $\kappa_0(x)$. With this notation, the new variational principle takes the final form

$$\bar{W}_+(\varepsilon) = \inf_{\mu_0(x), \kappa_0(x) \geq 0} \{ \bar{W}_0(\varepsilon) + V(\mu_0, \kappa_0) \}, \quad (3.21)$$

where

$$\bar{W}_0(\varepsilon) = \min_{u \in K(\varepsilon)} \int_{\Omega} w_0(x, \varepsilon(x)) dx, \quad (3.22)$$

and where V is the functional specified by the function $v(x; \mu_0, \kappa_0) = -\int_{\Omega} f_*(x; \frac{1}{2}\mu_0, \frac{1}{2}\kappa_0)$, such that

$$V(\mu_0, \kappa_0) = \int_{\Omega} v(x; \mu_0(x), \kappa_0(x)) dx. \quad (3.23)$$

We emphasize once again that, under the hypothesis of concave f , which is usually valid in plasticity, the new variational principle and the classical principle of minimum potential energy are *completely equivalent*, and we have equality in (3.14). Thus, under these circumstances, the new variational principle can be given the following interpretation: the effective behavior of the nonlinear heterogeneous solid, as characterized by its effective energy (2.4), can be alternatively determined by means of the effective energy of a linear heterogeneous comparison solid, with arbitrarily variable moduli, $\mu_0(x)$ and $\kappa_0(x)$, whose precise variation is determined by the new variational principle (3.21).

Clearly, the new variational principle is in general at least as difficult to evaluate as the original classical variational principle. The main advantage of the new variational principle, however, is that it allows useful approximations that cannot be obtained directly from the classical variational principle. One possible approximation in (3.21) is to replace the infimum over the set of arbitrarily variable, non-negative moduli by the smaller set of piecewise constant, non-negative moduli. We will consider the application of this approximation in the determination of bounds for the effective properties of nonlinear composites in the following section. This type of approximation was introduced directly by PONTE CASTAÑEDA (1991a). In particular, it was found in that work that when this approximation was used in conjunction with the well-known Hashin-Shtrikman bounds for linear composites, nonlinear bounds resulted that constituted in some cases improvements over the bounds determined earlier by PONTE CASTAÑEDA and WILLIS (1988), using the TALBOT and WILLIS (1985) nonlinear extension of the Hashin-Shtrikman variational principles. Recently, WILLIS (1991) has shown that if a better, optimal choice is made for the homogeneous comparison material in the context of the Talbot-Willis variational principle, the improved results of PONTE CASTAÑEDA (1991a) for the Hashin-Shtrikman bounds can also be obtained from the Talbot-Willis method. Further comparison between the two methods is given in PONTE CASTAÑEDA (1992), where it is shown that the Talbot-Willis variational procedure may be derived directly from the new variational procedure by expressing the effective energy function of the linear comparison composite (in the new variational principle) in terms of the classical linear Hashin-Shtrikman variational principle. Also, WILLIS (1992) has recently proposed a hybrid procedure making use of a *heterogeneous comparison composite* (as in the new variational procedure) instead of the standard *homogeneous comparison material* in the Talbot-Willis method.

Finally, we remark that statement (3.9) can be rewritten in the form

$$w(x; e) \leq \inf_{\mu_0, \kappa_0 \geq 0} \{w_0(x, e) + v(x; \mu_0, \kappa_0)\}, \quad (3.24)$$

(with equality if w is such that f is concave), where $v(x; \mu_0, \kappa_0)$ takes the corresponding form

$$v(x; \mu_0, \kappa_0) = \sup \{u(x, e) - w_0(x, e)\}. \quad (3.25)$$

With this representation of the local energy functions, the new variational principle is seen to be nothing more than the global version of the relation (3.24). Although we have already remarked that the new variational principle (3.21) is only exact for isotropic phases, provided that f is concave, the form (3.25) for $v(x; \mu_0, \kappa_0)$ suggests that the new principle could also be used for composites with anisotropic phases. Indeed this is the case, but in this event, equality could not hold in (3.24), and the new variational principle in the given form, would only yield an upper bound approximation to the effective behavior of the nonlinear heterogeneous solid $\mathcal{H}$; that would be expected to get progressively weaker with the degree of anisotropy in the constituent phases. Improved bounds could be obtained by the introduction of anisotropic comparison materials. This is outside the scope of this paper, and will be pursued elsewhere. By comparison, the Talbot-Willis variational principle can be used to obtain Hashin-Shtrikman bounds for composites with anisotropic phases, although some level of approximation is required for computational convenience (see DENEVEY *et al.*, 1991).

3.2. Minimum complementary energy: formulation

Given the function ψ defined by (3.5), we make use of the same change of variables h as in the previous subsection to define a function $g: \Omega \times R^+ \times R^- \rightarrow R^+$ such that

$$g(x; v, r, u_m) = \psi(x; h^{-1}(v), h^{-1}(u_m)). \quad (3.26)$$

By the properties of ψ , enunciated at the beginning of this section, g is a non-negative function with $g(x; 0, 0) = 0$. Further, g is continuous, but not necessarily convex, in its last two arguments. However, it is shown in Appendix II that if f is concave, then g is convex. As mentioned earlier, this is the usual situation in plasticity. For instance, for the same example of an incompressible power-law material given in the previous subsection, $u \sim \sigma^{n+1}$ ($1 \leq n < \infty$), and $g \sim v^{(n+1)/2}$, which are both convex functions.

We can then define (see Appendix I) the convex polar function $g^*: \Omega \times R^+ \times R^- \rightarrow R^+$ via

$$g^*(x; q, q_m) = \sup_{v, r \geq 0} \{v, q_r + v_m q_m - g(x; v, r, u_m)\}, \quad (3.27)$$

so that

$$g(x; v, r, u_m) \geq \sup_{q, q_m \geq 0} \{v, q_r + v_m q_m - g^*(x; q, q_m)\}, \quad (3.28)$$

with equality if g is convex.

Introducing the functional

$$G(v, v_m) = \int_{\Omega} g^*(x; v(x), v_m(x)) dx, \quad (3.29)$$

and following a procedure analogous to that of the previous subsection, we arrive at the following inequality involving the effective complementary energy of the nonlinear heterogeneous solid as characterized by (2.11), namely,

$$\bar{U}(\bar{\sigma}) \geq \bar{U}_-(\bar{\sigma}), \quad (3.30)$$

where

$$\bar{U}_-(\bar{\sigma}) = \min_{t_r, t_m \in S} \sup_{q_r, q_m \geq 0} M(t_r, t_m, q_r, q_m), \quad (3.31)$$

M is the "saddle" functional given by

$$M(t_r, t_m, q_r, q_m) = \int_0^1 [t_r(x)q_r(x) + t_m(x)q_m(x)] dx - \int_0^1 g^*(x; q_r(x), q_m(x)) dx, \quad (3.32)$$

and S' is the set induced by the above change of variables on the set of admissible stresses S . We emphasize that for concave f (and therefore convex g), $\bar{U}_-(\bar{\sigma}) = \bar{U}(\bar{\sigma})$.

In order to simplify the above expression for $\bar{U}_-$, we are required to interchange the order of the minimum and the supremum. This is allowed by Prop. VI.2.3 of EKELAND and TEMAM (1974), because M is convex in $q_r = \sqrt{t_r}$ and $q_m = \sqrt{t_m}$, and therefore convex in σ (which belongs to the convex set S), and concave (because g^* is convex) in q_r, q_m , and

$$\lim_{t_r, t_m \rightarrow x} M(t_r, t_m, q_r^{(0)}, q_m^{(0)}) = x \quad \text{for } q_r^{(0)}, q_m^{(0)} > 0$$

fixed. Thus, we conclude that

$$\min_{t_r, t_m \in S} \sup_{q_r, q_m \geq 0} M(t_r, t_m, q_r, q_m) = \sup_{q_r, q_m \geq 0} \inf_{t_r, t_m \in S} M(t_r, t_m, q_r, q_m). \quad (3.33)$$

This leads to the following restatement of (3.31)

$$\bar{U}_-(\bar{\sigma}) = \sup_{\mu, \kappa \geq 0} \{ \bar{U}_0(\bar{\sigma}) - V(\mu_0, \kappa_0) \}, \quad (3.34)$$

where

$$\bar{U}_0(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} \int_0^1 u_0(x, \sigma(x)) dx \quad (3.35)$$

is the effective complementary energy of the comparison composite, with $u_0 = u_0^*$ [see (3.20)], and where the functional V is given by relation (3.23), with

$$v(x; \mu_0, \kappa_0) = g^*\left(x; \frac{1}{6\mu_0}, \frac{1}{2\kappa_0}\right). \quad (3.36)$$

This last expression for v can be expressed directly in terms of u and u_0 via

$$v(x; \mu_0, \kappa_0) = \sup_{\sigma} \{ u_0(x, \sigma) - u(x, \sigma) \}. \quad (3.36)$$

In terms of this representation, it is easy to verify that the v functions given by (3.35) and (3.36) are in fact identical (and hence the choice of the same names). Simply note that from (3.36), and the fact that the order of suprema can be interchanged, it

follows that

$$\begin{aligned} v(x; \mu_0, \kappa_0) &= \sup_{\sigma} \left\{ \sup_{\varepsilon} \{ \varepsilon \cdot \sigma - u_0(x, \varepsilon) \} - u(x, \sigma) \right\} \\ &= \sup_{\varepsilon} \left\{ \sup_{\sigma} \{ \varepsilon \cdot \sigma - u(x, \sigma) \} - u_0(x, \varepsilon) \right\} \\ &= \sup_{\varepsilon} \{ w(x, \varepsilon) - u_0(x, \varepsilon) \}, \end{aligned} \quad (3.37)$$

which is in agreement with (3.25). This result can also be obtained by means of result 7) in Appendix II, which shows that

$$\begin{aligned} f_*(p_r, p_m) &= -g^*\left(\frac{1}{9p_r}, \frac{9}{4p_m}\right), \\ u(x, \sigma) &\geq \sup_{\mu_0, \kappa_0 \geq 0} \{ u_0(x, \sigma) - v(x; \mu_0, \kappa_0) \}, \end{aligned} \quad (3.38)$$

Also, note the following counterpart to (3.24), namely,

which is the local version of (3.34).

Finally, we study the relation between the two variational principles $\bar{W}_-$ and $\bar{U}_-$, as given by (3.21) and (3.34). We begin by evaluating the polar of $\bar{W}_-$, yielding

$$\begin{aligned} \bar{W}_*(\bar{\sigma}) &= \sup_{\varepsilon} \{ \bar{\varepsilon} \cdot \bar{\sigma} - \bar{W}_+(\bar{\varepsilon}) \} \\ &= \sup_{\varepsilon} \left\{ \bar{\varepsilon} \cdot \bar{\sigma} - \inf_{\mu, \kappa \geq 0} \{ \bar{W}_0(\bar{\varepsilon}) + V(\mu_0, \kappa_0) \} \right\} \\ &= \sup_{\mu, \kappa \geq 0} \left\{ \sup_{\varepsilon} \{ \bar{\varepsilon} \cdot \bar{\sigma} - \bar{W}_0(\bar{\varepsilon}) \} - V(\mu_0, \kappa_0) \right\} \\ &= \sup_{\mu, \kappa \geq 0} \{ \bar{W}_0^*(\bar{\sigma}) - V(\mu_0, \kappa_0) \}, \end{aligned} \quad (3.39)$$

where once again we have used the fact that the order of the suprema can be interchanged. Next, by specializing the dual version of result (2.15) to the linear comparison composite (i.e. $\bar{W}_0^* \leq \bar{U}_0$), we conclude that

$$\bar{W}_*(\bar{\sigma}) \leq \bar{U}_-(\bar{\sigma}). \quad (3.40)$$

However, for a composite [see the discussion following (2.15)], $\bar{W}_0^* = \bar{U}_0$, and therefore we have $\bar{W}_*(\bar{\sigma}) = \bar{U}_-$ in place of (3.40), which demonstrates that there is no *duality gap* between the minimum potential and minimum complementary energy versions of the new variational principle. Further, if f is concave, then we have equality in (3.14) and (3.30), simultaneously, and therefore we also have equality between $\bar{W}_*$ and $\bar{U}_-$, which is in agreement (as it should be) with the discussion following (2.15). Hence, in applications, one may utilize either one of the two versions of the variational principle, with the choice usually being dictated by convenience.

4. APPLICATION TO BOUNDS

In this section, we are concerned with the determination of bounds for the effective behavior of nonlinear composite materials in the context of plasticity. The approach will be to make use of well-known bounds for linear elastic composites into the new variational principles to induce corresponding bounds for nonlinear plastic composites. Although the procedure can be applied more generally than we will consider in this section, we provide a sampling of the possible applications of the method by including a novel derivation of the classical Voigt/Reuss bounds for generally anisotropic composites, novel forms for the Hashin-Shtrikman bounds for isotropic and transversely isotropic composites, and completely new higher-order bounds of the Beran type also for isotropic composites. We will be interested only in materials with isotropic energy functions satisfying the concavity of f hypothesis, which is adequate in plasticity. This being the case, we have complete equivalence between the new and classical variational principles, and it is then clear from either (3.21) or (3.34) that bounds on the effective energy-density functions of the linear comparison composites can be utilized, at least in principle, to yield corresponding bounds on the effective energy-density functions of nonlinear composites.

In particular, we still study heterogeneous solids with n distinct phases, each of which is homogeneous and isotropic. Also, the volume fractions of each phase will be assumed to be given. According to relation (3.1), we can characterize the energy-density functions of the different phases in terms of functions $\phi^{(r)}: R^+ \times R^+ \rightarrow R^+$, $R^+ = 1, \dots, n$, such that the local energy-density function of the composite is given by

$$\phi(\mathbf{x}, \boldsymbol{\epsilon}, \boldsymbol{\epsilon}_p) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) \phi^{(r)}(\boldsymbol{\epsilon}, \boldsymbol{\epsilon}_p), \quad (4.1)$$

where $\chi^{(r)}(\mathbf{x})$ is the characteristic function of phase r (this function vanishes, unless $\mathbf{x}$ is in phase r , in which case it equals unity). However, because of the usual representation in plasticity of the total strain as the sum of the (linear) elastic strain and the (nonlinear) plastic strain, we find it more convenient to represent the constitutive relation for the composite in terms of the corresponding complementary energy-density functions of each phase, $\psi^{(r)}$, with a representation for the complementary energy-density function of the composite ψ completely analogous to (4.1). Further, in this paper, we will restrict our attention to the class of complementary-energy functions

$$\psi^{(r)}(\boldsymbol{\sigma}, \boldsymbol{\sigma}_p) = \phi^{(r)}(\boldsymbol{\sigma}, \boldsymbol{\sigma}_p) + \frac{1}{2\mu^{(r)}} \boldsymbol{\sigma}_p^2, \quad (4.2)$$

where the functions $\phi^{(r)}$ are convex, non-negative and such that the corresponding f functions are concave, and the $\kappa^{(r)}$ are given non-negative constants. The form (4.2) is quite appropriate in plasticity since the plastic strain is usually taken to be traceless (incompressible), so that the compressible part of the constitutive relation is due only to the elastic strain, and is hence linear (corresponding to a quadratic potential). Thus, $\kappa^{(r)}$ is the (elastic) bulk modulus of phase r , and $\phi^{(r)}$ characterizes the total shear

behavior of phase r (both elastic and plastic contributions). For instance, for a linear elastic material,

$$\phi^{(r)}(\boldsymbol{\sigma}_p) = \frac{1}{6\mu^{(r)}} \boldsymbol{\sigma}_p^2, \quad (4.3)$$

where $\mu^{(r)} \geq 0$ is the elastic shear modulus of the material. More generally, a plastic potential will usually be added to the quadratic potential of (4.3) to account for the plastic strains. In our development in the following sections, we will not specialize our consideration to any specific form for the plastic potential, and our results will hold for arbitrary energy-density functions of the form (4.2), with the given hypotheses on $\phi^{(r)}$ and $\kappa^{(r)}$. We further note that the volume fractions of each phase are given by

$$c^{(r)} = \int_{\Omega} \chi^{(r)}(\mathbf{x}) d\mathbf{x} \quad (4.4)$$

and are such that $\sum_{r=1}^n c^{(r)} = 1$.

In accord with the above-mentioned representation for the local complementary energy-density function $u(\mathbf{x}, \boldsymbol{\sigma}) = \psi(\mathbf{x}; \boldsymbol{\sigma}, \boldsymbol{\sigma}_p)$, we will make use of the complementary energy formulation (3.34) of the new variational principle, although equivalent results would be obtained by means of the dual potential energy formulation (3.21). Thus, we will consider first in Section 4.1 lower bounds for the effective energy function $\bar{U}$, and then continue in Section 4.2 with the harder problem of determining upper bounds for $\bar{U}$. The relationship of the bounds to actual microstructures, and concomitant implications for optimality of the bounds will be addressed briefly in Section 4.3.

4.1. Lower bounds

As indicated in Section 3, lower bounds for the effective complementary energy of the nonlinear composite $\bar{U}$ can be obtained directly from (3.34) by restricting the class of arbitrarily varying (non-negative) comparison moduli to the smaller class of piecewise constant (non-negative) moduli. Thus, for the composites characterized by (4.2), we consider a comparison composite with shear moduli given by

$$\mu_0(\mathbf{x}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) \mu_0^{(r)}, \quad (4.5)$$

where the $\mu_0^{(r)}$ are non-negative constants. We note that we have selected the same microstructural distribution for the comparison composite as for the nonlinear composite. Also, we remark that because the hydrostatic terms in the $\psi^{(r)}$ functions are quadratic, the supremum problem over the comparison bulk moduli in (3.34) is solved exactly by

$$\kappa_0(\mathbf{x}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) \kappa_0^{(r)}, \quad (4.6)$$

where $\kappa_0^{(r)} = \kappa^{(r)}$.

It follows from this discussion that

$$\bar{U}(\bar{\sigma}) \geq \sup_{\mu_0^{(r)} > 0} \left\{ \bar{U}_0(\bar{\sigma}) - \sum_{r=1}^n c^{(r)} v^{(r)}(\mu_0^{(r)}) \right\}, \quad (4.7)$$

where $\bar{U}_0$ corresponds to the effective complementary energy of the linear comparison composite with shear and bulk moduli characterized by (4.5) and (4.6), respectively, and where the functions $v^{(r)}$ take the simplified form

$$v^{(r)}(\mu_0^{(r)}) = \sup_{s \geq 0} \left\{ \frac{1}{6\mu_0^{(r)}} s^2 - \varphi^{(r)}(s) \right\}. \quad (4.8)$$

We note for later reference that

$$\varphi^{(r)}(s) = \sup_{\mu_0^{(r)} > 0} \left\{ \frac{1}{6\mu_0^{(r)}} s^2 - \psi^{(r)}(\mu_0^{(r)}) \right\}. \quad (4.9)$$

Thus, any lower bound for $\bar{U}_0$ can be utilized in (4.7) to induce a lower bound on $\bar{U}$. We note that for nonlinear composites, just as for linear composites, the bounds on the effective energy will depend on the prescribed microstructural information. For instance, the Hashin-Shtrikman bounds for isotropic composites will be tighter than the Voigt-Reuss bounds for generally anisotropic composites. However, it is important to note that the characterization of microstructure is different for linear and nonlinear materials. For example, a linear composite with periodic microstructure and a unit cell possessing icosahedral symmetry has effective behavior with isotropic symmetry. The same is *not* true in general of a periodic nonlinear composite with an icosahedral unit cell. This is because linear behaviors may be characterized in terms of (finite dimensional) tensors, whereas nonlinear behaviors require characterization in terms of general (infinite dimensional) functions. Notwithstanding this difficulty, we observe that the class of isotropic linear *microstructures* includes the class of isotropic nonlinear microstructures. Hence, nonlinear bounds based on linear isotropic microstructures will also hold for nonlinear isotropic microstructures. Analogous results would hold for any other material symmetries for the composite (e.g. transversely isotropic). Thus, a nonlinear lower bound for nonlinear composites with a given symmetry results by replacing $\bar{U}_0$ in (4.7) by a lower bound for $\bar{U}_0$ with the same symmetry.

4.1.1. Voigt bound. We begin by considering the case of a generally anisotropic composite with phase potentials (4.2) in given volume fractions $c^{(r)}$. Thus, in this case, we require a lower bound for the effective complementary-energy function of an anisotropic linear comparison composite with shear and bulk moduli characterized by (4.5) and (4.6), in volume fractions $c^{(r)}$. Such a bound is provided by the so-called Voigt estimate

$$\bar{U}_0^{(V)}(\bar{\sigma}) = \frac{1}{6\mu_0^{(V)}} \bar{\sigma}^2 + \frac{1}{2\kappa_0^{(V)}} \bar{\sigma}_m^2, \quad (4.10)$$

where

$$\mu_0^{(V)} = \sum_{r=1}^n c^{(r)} \mu_0^{(r)} \quad \text{and} \quad \kappa_0^{(V)} = \sum_{r=1}^n c^{(r)} \kappa_0^{(r)}.$$

This result can be readily obtained from the principle of minimum complementary energy (HILL, 1952; PAUL, 1960). Thus, application of this lower bound for the linear comparison composite in (4.7) leads to the following lower bound for the effective energy of the nonlinear composite

$$\bar{U}^{(V)}(\bar{\sigma}) = \sup_{\mu_0^{(r)} > 0} \left\{ \frac{1}{6\mu_0^{(V)}} \bar{\sigma}^2 - \sum_{r=1}^n c^{(r)} v^{(r)}(\mu_0^{(r)}) \right\} + \frac{1}{2\kappa_0^{(V)}} \bar{\sigma}_m^2, \quad (4.11)$$

where $\kappa^{(r)} = \sum_{r=1}^n c^{(r)} \kappa^{(r)}$. The above bound involves $2n$ optimizations (two for each phase), but it can be significantly simplified by means of the identity of Appendix III

$$\frac{1}{\mu_0^{(V)}} = \inf_{\omega^{(r)} > 0} \left\{ \sum_{r=1}^n \frac{c^{(r)}}{\mu_0^{(r)}} (1 - \omega^{(r)})^2 \right\}, \quad (4.12)$$

where the infimum is over the set of variables $\omega^{(r)}$ ($r = 1, \dots, n$), which are subject to a zero-average constraint $\bar{\omega} = \sum_{r=1}^n c^{(r)} \omega^{(r)} = 0$. With this identity, the expression of the bound in (4.11) leads to

$$\bar{U}^{(V)}(\bar{\sigma}) = \sup_{\mu_0^{(r)} > 0} \left\{ \inf_{\omega^{(r)} > 0} \left\{ \sum_{r=1}^n c^{(r)} \left[\frac{1}{6\mu_0^{(r)}} (1 - \omega^{(r)})^2 \bar{\sigma}^2 - \psi^{(r)}(\mu_0^{(r)}) \right] \right\} + \frac{1}{2\kappa_0^{(V)}} \bar{\sigma}_m^2 \right\}. \quad (4.13)$$

Next, recalling that $-\psi^{(r)}$ is concave in $\mu_0^{(r)}$ (since g^* is convex), and noting that the expression inside the square brackets is convex in $\omega^{(r)}$, the Saddle Point Theorem (EKELAND and TEMAM, 1974; § VI.4.2) allows the interchange of the order in which the suprema and infima are evaluated to arrive at

$$\bar{U}^{(V)}(\bar{\sigma}) = \inf_{\mu_0^{(r)} > 0} \left\{ \sum_{r=1}^n c^{(r)} \left[\sup_{\omega^{(r)} > 0} \left\{ \frac{1}{6\mu_0^{(r)}} (1 - \omega^{(r)})^2 \bar{\sigma}^2 - \psi^{(r)}(\mu_0^{(r)}) \right\} \right] + \frac{1}{2\kappa_0^{(V)}} \bar{\sigma}_m^2 \right\}, \quad (4.14)$$

which finally reduces to

$$\bar{U}^{(V)}(\bar{\sigma}) = \inf_{\mu_0^{(r)} > 0} \left\{ \sum_{r=1}^n c^{(r)} \varphi^{(r)}(1 - \omega^{(r)}|\bar{\sigma}) \right\} + \frac{1}{2\kappa_0^{(V)}} \bar{\sigma}_m^2, \quad (4.15)$$

by means of (4.9). To the knowledge of the author, this result is new.

This lower bound for $\bar{U}$, obtained from the linear Voigt bound via the new variational principle, can be shown to be equivalent to the well-known bound, obtained directly from the principle of minimum potential energy (for a nonlinear composite) by assuming a constant strain field over the composite, and given by

$$\bar{U}(\bar{\sigma}) \geq \left(\sum_{i=1}^n c^{(i)} (\varphi^{(i)})^* \right) (\bar{\sigma}_i) + \frac{1}{2\kappa^{(i)}} \bar{\sigma}_i^2. \quad (4.16)$$

For this reason, we refer to the lower bound (4.15) as the nonlinear Voigt bound. We note that, in its standard form, the bound (4.16) involves $n+1$ optimizations (Legendre transforms), whereas the new form (4.15) involves n optimizations with a simple linear constraint. This linear constraint may be easily embedded in the optimization problem to further reduce the number of optimizations to $n-1$. For example, for a two-phase composite, the new form of the Voigt bound reduces to

$$\bar{U}^{(1)}(\bar{\sigma}) = \inf_{\omega} \{ c^{(1)} \varphi^{(1)} (1 - c^{(2)} \omega) \bar{\sigma}_i + c^{(2)} \phi^{(2)} (1 + c^{(1)} \omega) \bar{\sigma}_i \} + \frac{1}{2\kappa^{(1)}} \bar{\sigma}_i^2, \quad (4.17)$$

which involves only one optimization.

4.1.2. Hashin-Shtrikman lower bounds. Next, we consider nonlinear composites with phases having complementary energy-density functions (4.2), in prescribed volume fractions $c^{(i)}$, distributed in such a way that the overall composite is *isotropic*. In this case, the linear comparison composite to be used in conjunction with (4.7) is taken to be isotropic with shear and bulk moduli, given once again by (4.5) and (4.6), respectively, in volume fractions $c^{(i)}$, and with $\kappa_0^{(i)} = \kappa^{(i)}$. The relevant isotropic, linear lower bound, due to HASHIN and SHTRIKMAN (1963), is provided by the relation

$$\bar{U}_0^{HS-1}(\bar{\sigma}) = \frac{1}{6\mu_0^{HS-1}} \bar{\sigma}_i^2 + \frac{1}{2\kappa_0^{HS-1}} \bar{\sigma}_m^2, \quad (4.18)$$

where

$$\mu_0^{HS-1} = \frac{\sum_{i=1}^n c^{(i)} \frac{\mu_0^{(i)}}{\mu_0^{(i)} + \mu_0^{(i)}}}{\sum_{i=1}^n c^{(i)} \frac{1}{\mu_0^{(i)} + \mu_0^{(i)}}} \quad \text{and} \quad \kappa_0^{HS-1} = \frac{\sum_{i=1}^n c^{(i)} \frac{\kappa_0^{(i)}}{3\kappa_0^{(i)} + 4\mu_0^{(i)}}}{\sum_{i=1}^n c^{(i)} \frac{1}{3\kappa_0^{(i)} + 4\mu_0^{(i)}}}, \quad (4.19)$$

with

$$\mu_0^{(i)} = \frac{8\mu_0^{(i)} + 9\kappa_0^{(i)}}{6(2\mu_0^{(i)} + \kappa_0^{(i)})} \mu_0^{(i)}, \quad \mu_0^{(i)} = \max \{ \mu_0^{(i)} \}, \quad \text{and} \quad \kappa_0^{(i)} = \max \{ \kappa_0^{(i)} \}.$$

Thus, the nonlinear bound for the class of isotropic composites with potentials (4.2) in prescribed volume fractions $c^{(i)}$ becomes

$$\bar{U}^{HS-1}(\bar{\sigma}) = \sup_{\omega_0^{(i)} \geq 0} \left\{ \frac{1}{6\mu_0^{HS-1}} \bar{\sigma}_i^2 + \frac{1}{2\kappa_0^{HS-1}} \bar{\sigma}_m^2 - \sum_{i=1}^n c^{(i)} \varphi^{(i)}(\mu_0^{(i)}) \right\}, \quad (4.20)$$

where we have replaced $\kappa_0^{(i)}$ by $\kappa^{(i)}$ in the expression (4.19), for κ_0^{HS-1} . Note that this expression still depends on $\mu_0^{(i)}$. Similarly, we replace $\kappa_0^{(i)}$ by $\kappa^{(i)}$ in the expression (4.19), for μ_0^{HS-1} .

In general, the expression (4.20) for the nonlinear isotropic lower bound is quite complicated. Simplification can be achieved by rewriting the above expression for

μ_0^{HS-1} in the form

$$\frac{1}{\mu_0^{HS-1}} = \left[\sum_{i=1}^n c^{(i)} \left(\frac{1}{\mu_0^{(i)}} + \frac{1}{\mu_0^{(i)}} \right)^{-1} \right]^{-1} - \frac{1}{\mu_0^{(i)}}, \quad (4.21)$$

which in turn can be rewritten in the form

$$\frac{1}{\mu_0^{HS-1}} = \inf_{\omega_0^{(i)} \geq 0} \left\{ \sum_{i=1}^n c^{(i)} \left[\frac{1}{\mu_0^{(i)}} (1 - \omega^{(i)})^2 + \frac{1}{\mu_0^{(i)}} (\omega^{(i)})^2 \right] \right\}, \quad (4.22)$$

by means of the identity of Appendix III.

Similarly, we may write

$$\frac{1}{\kappa_0^{HS-1}} = \inf_{\gamma^{(i)} \geq 0} \left\{ \sum_{i=1}^n c^{(i)} \left[\frac{1}{\kappa^{(i)}} (1 - \gamma^{(i)})^2 + \frac{3}{4\mu_0^{(i)}} (\gamma^{(i)})^2 \right] \right\}, \quad (4.23)$$

where the optimization variables $\gamma^{(i)}$ satisfy the constraint $\bar{\gamma} = \sum_{i=1}^n c^{(i)} \gamma^{(i)} = 0$. Therefore, we have that

$$\begin{aligned} \bar{U}^{HS-1}(\bar{\sigma}) = \sup_{\omega_0^{(i)} \geq 0} \left\{ \inf_{\gamma^{(i)} \geq 0} \left\{ \sum_{i=1}^n c^{(i)} \left[\frac{1}{\mu_0^{(i)}} (1 - \omega^{(i)})^2 + \frac{1}{\mu_0^{(i)}} (\omega^{(i)})^2 \right] \frac{\bar{\sigma}_i^2}{6} + \dots \right. \right. \\ \left. \left. + \left[\frac{1}{\kappa^{(i)}} (1 - \gamma^{(i)})^2 + \frac{3}{4\mu_0^{(i)}} (\gamma^{(i)})^2 \right] \frac{\bar{\sigma}_m^2}{2} - \varphi^{(i)}(\mu_0^{(i)}) \right\} \right\}. \end{aligned} \quad (4.24)$$

By the same arguments following (4.13), allowing the interchange of the suprema and infima, we arrive at

$$\begin{aligned} \bar{U}^{HS-1}(\bar{\sigma}) = \inf_{\omega_0^{(i)} \geq 0} \left\{ \sup_{\gamma^{(i)} \geq 0} \left\{ \sum_{i=1}^n c^{(i)} \left[\frac{1}{\mu_0^{(i)}} (1 - \omega^{(i)})^2 + \frac{1}{\mu_0^{(i)}} (\omega^{(i)})^2 \right] \frac{\bar{\sigma}_i^2}{6} + \dots \right. \right. \\ \left. \left. + \frac{3}{8\mu_0^{(i)}} (\gamma^{(i)})^2 \bar{\sigma}_m^2 - \varphi^{(i)}(\mu_0^{(i)}) + \frac{1}{2\kappa^{(i)}} (1 - \gamma^{(i)})^2 \bar{\sigma}_m^2 \right\} \right\}. \end{aligned} \quad (4.25)$$

Making use of the expression for $\mu_0^{(i)}$, this can in turn be rewritten

$$\begin{aligned} \bar{U}^{HS-1}(\bar{\sigma}) = \min_{\omega_0^{(i)} \geq 0} \left\{ \sup_{\gamma^{(i)} \geq 0} \left\{ \sum_{i=1}^n c^{(i)} \left[\frac{1}{\mu_0^{(i)}} (1 - \omega^{(i)})^2 + \frac{1}{\mu_0^{(i)}} (\omega^{(i)})^2 \right] \frac{\bar{\sigma}_i^2}{6} + \dots \right. \right. \\ \left. \left. + \frac{3}{8\mu_0^{(i)}} (\gamma^{(i)})^2 \bar{\sigma}_m^2 - \varphi^{(i)}(\mu_0^{(i)}) + \frac{1}{2\kappa^{(i)}} (1 - \gamma^{(i)})^2 \bar{\sigma}_m^2 \right\} \right\}, \end{aligned} \quad (4.26)$$

where we have used the fact that

$$\frac{1}{\mu_0^{(i)}} = \frac{6}{\mu_0^{(i)}} \left(\frac{2\mu_0^{(i)} + \kappa_0^{(i+1)}}{\mu_0^{(i)} + 9\kappa_0^{(i+1)}} \right)$$

is a monotonically decreasing function of $\mu_0^{(i)}$.

Finally, by means of (4.9), we arrive at

$$\begin{aligned} \tilde{J}^{(HS-1)}(\bar{\sigma}) = \min_i \left\{ \inf_{\substack{\omega^{(i)}, \bar{\sigma} = 0 \\ \gamma^{(i)} = 0}} \left\{ \sum_{j=1}^n c^{(i)} \varphi^{(i)}(1 - \omega^{(i)}|\bar{\sigma}_j| + c^{(i)}\Delta_0^{(i)} \right. \right. \\ \left. \left. + \left(\sum_{k=1}^n \frac{c^{(k)}}{\kappa^{(k)}} (1 - \gamma^{(k)})^2 \right) \frac{\bar{\sigma}_m^2}{2} \right\} \right\}, \quad (4.27) \end{aligned}$$

where

$$\Delta_0^{(i)}(\bar{\sigma}, \bar{\sigma}_m; \omega^{(i)}, \gamma^{(i)}) = \sup_{\mu_0^{(i)} \geq 0} \left\{ \frac{1}{6\mu_0^{(i)}} s^2 - v^{(i)}(\mu_0^{(i)}) \right\}, \quad (4.28)$$

and

$$\begin{aligned} s = \left[(1 - \omega^{(i)})^2 \bar{\sigma}_j^2 + \frac{6}{c^{(i)}} \left(\frac{2\mu_0^{(i)} + \kappa_0^{(i+1)}}{\mu_0^{(i)} + 9\kappa_0^{(i+1)}} \right) \left(\sum_{k=1}^n c^{(k)} (\omega^{(k)})^2 \right) \bar{\sigma}_j^2 \right. \\ \left. + \frac{9}{4c^{(i)}} \left(\sum_{k=1}^n c^{(k)} (\gamma^{(k)})^2 \right) \bar{\sigma}_m^2 \right]^{1/2}. \quad (4.29) \end{aligned}$$

Note that s depends on $\mu_0^{(i)}$, and therefore the supremum in (4.28) is not trivially evaluated [i.e. (4.9) may not be used]. Thus, the new expression for the nonlinear Hashin-Shtrikman lower bound involves an $2n$ -dimensional constrained optimization as given by (4.27), together with a one-dimensional optimization problem for $\Delta_0^{(i)}$, as given by (4.28). Also, we note that there are n "branches" to the solution (one for each phase), and that the minimum over all the branches yields the desired lower bound.

We remark that the expression for the bound (4.27) simplifies to

$$\begin{aligned} \tilde{J}^{(HS-1)}(\bar{\sigma}) = \min_i \left\{ \inf_{\substack{\omega^{(i)}, \bar{\sigma} = 0 \\ \gamma^{(i)}, \bar{\sigma} = 0}} \left\{ \sum_{j=1}^n c^{(i)} \varphi^{(i)}(s^{(i)}) + c^{(i)} \varphi^{(i)}(s^{(i)}) \right. \right. \\ \left. \left. + \left(\sum_{k=1}^n \frac{c^{(k)}}{\kappa^{(k)}} (1 - \gamma^{(k)})^2 \right) \frac{\bar{\sigma}_m^2}{2} \right\} \right\}, \quad (4.30) \end{aligned}$$

where

$$s^{(i)} = \sqrt{(1 - \omega^{(i)})^2 \bar{\sigma}_j^2 + \frac{2}{3c^{(i)}} \left(\sum_{k=1}^n c^{(k)} (\omega^{(k)})^2 \right) \bar{\sigma}_j^2 + \frac{9}{4c^{(i)}} \left(\sum_{k=1}^n c^{(k)} (\gamma^{(k)})^2 \right) \bar{\sigma}_m^2}, \quad (4.31)$$

if at least one phase is incompressible (so that $\kappa_0^{(i)} \rightarrow \infty$).

If all phases are incompressible, the expression simplifies further to

$$\tilde{J}^{(HS-1)}(\bar{\sigma}) = \min_i \left\{ \inf_{\substack{\omega^{(i)} \\ \gamma^{(i)} = 0}} \left\{ \sum_{j=1}^n c^{(i)} \varphi^{(i)}(s^{(i)}) + c^{(i)} \varphi^{(i)}(s^{(i)}) \right\} \right\}, \quad (4.32)$$

where the $s^{(i)}$ are the same as above, and

$$s^{(i)} = \sqrt{(1 - \omega^{(i)})^2 \bar{\sigma}_j^2 + \frac{2}{3c^{(i)}} \left(\sum_{k=1}^n c^{(k)} (\omega^{(k)})^2 \right) \bar{\sigma}_j^2}. \quad (4.33)$$

If there are only two phases, the constraints can be accounted for by letting $\omega^{(1)} = c^{(2)}\omega$ and $\omega^{(2)} = -c^{(1)}\omega$ (with similar forms for $\gamma^{(1)}$ and $\gamma^{(2)}$). Then, when both phases are incompressible, we arrive at the simple expression

$$\tilde{J}^{(HS-1)}(\bar{\sigma}) = \min \left\{ \inf_{\omega} \left\{ c^{(1)} \varphi^{(1)} \left[(1 - c^{(2)}\omega) \bar{\sigma}_j + c^{(2)} \varphi^{(2)} \left(\sqrt{(1 + c^{(1)}\omega)^2 + 3c^{(1)}\omega^2 \bar{\sigma}_j^2} \right) \right] \right\} \right. \\ \left. \inf_{\omega} \left\{ c^{(1)} \varphi^{(1)} \left[(1 - c^{(2)}\omega) \sqrt{(1 - c^{(2)}\omega)^2 + 3c^{(2)}\omega^2 \bar{\sigma}_j^2} + c^{(2)} \varphi^{(2)} \left[(1 + c^{(1)}\omega) \bar{\sigma}_j \right] \right] \right\} \right\} \quad (4.34)$$

This expression generalizes results given by PONTE CASTAÑEDA (1991b) for a dual-phase incompressible composite with a linear and a nonlinear phase. Another simple result is provided by the porous material, when one phase, say phase 2, is void, and the other is incompressible. Then, the lower bound (4.30) reduces to (since $\omega^{(2)} = \gamma^{(2)} = 1$)

$$\tilde{J}^{(HS-1)}(\bar{\sigma}) = c^{(1)} \varphi^{(1)} \left(\frac{1}{c^{(1)}} \sqrt{(1 + 3c^{(2)}) \bar{\sigma}_j^2 + 3c^{(2)} \bar{\sigma}_m^2} \right), \quad (4.35)$$

first given by PONTE CASTAÑEDA (1991a) (see also PONTE CASTAÑEDA, 1991c). WILLIS (1991) has also shown explicitly that this particular result can be obtained from the Talbot-Willis variational principle by proper choice of the *homogeneous* comparison material. A weaker version of this result, also making use of the Talbot-Willis variational principle, was provided by PONTE CASTAÑEDA and WILLIS (1988). There are other simple cases in the context of isotropic nonlinear composites, but in the interest of time, they will be considered elsewhere.

Finally, we remark that the new variational principle can also be used in connection with bounds for linear anisotropic composites with *special* symmetries to obtain corresponding bounds for nonlinear anisotropic composites with given symmetries (the results given in Section 4.1.1 hold for *generally* anisotropic composites). For instance, the case of fiber-reinforced materials is considered by DeBORTON and PONTE

CASTAÑEDA (1992). These authors make use of the corresponding linear bounds of HILL (1964) and HASHIN (1965) (see also WALPOLE, 1969). The resulting nonlinear bounds for the fiber-reinforced composite are more complicated than the bounds for the isotropic composite (since they have transversely isotropic symmetry), but have essentially the same form. For illustrative purposes, we include here the result for an n -phase, fiber-reinforced composite with isotropic, incompressible phases is prescribed volume fractions $c^{(i)}$, which is given by

$$\bar{U}^{(M+1)}(\bar{\sigma}) = \min_{\substack{\omega^{(i)} \geq 0 \\ \sum_{i=1}^n \omega^{(i)} = 1}} \left\{ \inf_{\substack{\bar{\sigma}_n, \bar{\sigma}_p, \bar{\sigma}_s \\ \bar{\sigma}_n + \bar{\sigma}_p + \bar{\sigma}_s = \bar{\sigma}}} \left\{ \sum_{i=1}^n c^{(i)} \varphi^{(i)}(\bar{\sigma}^{(i)}) + c^{(n)} \varphi^{(n)}(\bar{\sigma}^{(n)}) \right\} \right\}, \quad (4.36)$$

where

$$\bar{\sigma}^{(i)} = \sqrt{(1 - \omega^{(i)})^2 (\bar{\epsilon}_n^2 + \bar{\epsilon}_p^2) + (1 - \omega^{(i)})^2 (\bar{\sigma}_n - \bar{\sigma}_p)^2}, \quad (4.37)$$

$$\bar{\sigma}^{(n)} = \sqrt{\left[(1 - \omega^{(n)})^2 + \frac{1}{c^{(n)}} \sum_{i=1}^n c^{(i)} (\omega^{(i)})^2 \right] (\bar{\epsilon}_n^2 + \bar{\epsilon}_p^2) + (1 - \omega^{(n)})^2 (\bar{\sigma}_n - \bar{\sigma}_p)^2},$$

with

$$\bar{\sigma}_n = \bar{\sigma}_i, n, n_i, \quad \bar{\sigma}_p = \frac{1}{2} \bar{\sigma}_i (\delta_{ij} - n_i n_j),$$

$$\bar{\epsilon}_n = \sqrt{3(\bar{\sigma}_i \bar{\sigma}_j \delta_{ij} n_k - \bar{\sigma}_n^2)}, \quad \text{and} \quad \bar{\epsilon}_p = \sqrt{\bar{\sigma}_n^2 - \bar{\epsilon}_n^2} - (\bar{\sigma}_n - \bar{\sigma}_p)^2,$$

denoting the transversely isotropic invariants of the mean stress $\bar{\sigma}$ implied by the fiber orientation vector $\mathbf{n}$. Results in a different, perhaps more complicated, form for this case have also been given by TALBOT and WILLIS (1991), using the Talbot-Willis variational method.

4.1.3. Higher-order lower bounds. So far, we have given examples of bounds that are either well-known, or can be obtained, at least in principle, by other procedures. Thus, the Voigt nonlinear bound can be obtained more directly from the classical principle of minimum complementary energy, and the nonlinear Hashin-Shtrikman lower bounds can also be obtained by means of the Talbot-Willis nonlinear extension of the Hashin-Shtrikman variational principle. However, we emphasize that the simple forms (4.15) and (4.17) for the Voigt bound, and (4.27)–(4.32) for the Hashin-Shtrikman bounds are new. In this subsection, we discuss the determination of nonlinear bounds that have not been possible by any other method hitherto.

Although many different higher-order bounds are available for linear-elastic composites, we will restrict our consideration to the third-order bounds of BERAN and MOLYNEUX (1965), and MCCOY (1970). The reason is that while higher-order bounds may have quite complex forms involving many additional geometric parameters (other than the volume fractions), MILTON (1982) has given a particularly simple form for the third-order bounds of dual-phase isotropic composites, involving only two additional geometric parameters. Milton's form for the bounds $\mu_0^{(M+1)}$ and $\kappa_0^{(M+1)}$ is expressed by

$$\frac{1}{\mu_0^{(M+1)}} = \left[\sum_{i=1}^2 c^{(i)} \left(\frac{1}{\mu_0^{(i)}} + \frac{1}{\mu_0^{(M+1)}} \right)^{-1} \right]^{-1} - \frac{1}{\mu_0^{(M+1)}}, \quad (4.38)$$

which is identical in form to (4.21), but with $\mu_0^{(i)} = \Theta$, where Θ is given by a rather complicated expression [see (3.29) in MILTON (1982)] involving $\mu_0^{(i)}$ and $\kappa_0^{(i)}$, as well as the geometric parameters $\eta^{(i)}$ and $\zeta^{(i)}$, and by

$$\frac{1}{\kappa_0^{(M+1)}} = \left[\sum_{i=1}^2 c^{(i)} \left(\frac{1}{\kappa_0^{(i)}} + \frac{1}{\kappa_0^{(M+1)}} \right)^{-1} \right]^{-1} - \frac{1}{\kappa_0^{(M+1)}}, \quad (4.39)$$

where $\kappa_0^{(i)} = \frac{4}{3} \sum_{j=1}^2 \zeta^{(j)} \kappa_0^{(j)}$. Here, the geometric parameters $\eta^{(i)}$ and $\zeta^{(i)}$ lie in the interval $[0, 1]$, and $\eta^{(2)} = 1 - \eta^{(1)}$ and $\zeta^{(2)} = 1 - \zeta^{(1)}$.

Substitution of these expressions for the comparison moduli into (4.7) yields an isotropic lower bound $\bar{U}^{(M+1)}$ for the effective energy of the nonlinear composite involving the phase potentials $\varphi^{(1)}$ and $\varphi^{(2)}$ of the two phases, the corresponding volume fractions $c^{(1)}$ and $c^{(2)}$, and the two additional geometric parameters $\eta^{(1)}$ and $\zeta^{(1)}$. However, due to the complicated form of $\mu_0^{(M+1)}$ in (4.38), no further simplification of the nonlinear bound is available. Fortunately, for the case of two incompressible phases, the expression for $\mu_0^{(M+1)}$ simplifies to

$$\mu_0^{(M+1)} = \frac{3}{2} \sum_{i=1}^2 \eta^{(i)} \mu_0^{(i)}. \quad (4.40)$$

which depends only on the geometric parameter $\eta^{(1)}$. Then, use of a procedure very similar to the one following (4.21) for the Hashin-Shtrikman lower bound leads to the nonlinear third-order lower bound

$$\bar{U}^{(M+1)}(\bar{\sigma}) = \inf_{\omega^{(i)}} \left\{ c^{(1)} \varphi^{(1)} \left(\sqrt{(1 - c^{(1)} \omega)^2 + \frac{3}{2} c^{(2)} \eta^{(1)} \omega^2 (1 - \eta^{(1)})^2} \bar{\sigma}_i} \right) + \dots \right. \\ \left. + c^{(2)} \varphi^{(2)} \left(\sqrt{(1 - c^{(1)} \omega)^2 + \frac{3}{2} c^{(1)} \eta^{(1)} \omega^2 (1 + \eta^{(1)})^2} \bar{\sigma}_i} \right) \right\}, \quad (4.41)$$

where $\eta^{(2)} = 1 - \eta^{(1)}$. We remark that the corresponding nonlinear Hashin-Shtrikman lower bound (4.34) follows immediately from this result by choosing either $\eta^{(1)} = 0$, or $\eta^{(1)} = 1$, whichever yields the lowest value (note that the infimum problem over η becomes trivial in either case). This is completely analogous to the corresponding result for linear two-phase incompressible composites (MILTON, 1982). Finally, we note that a superficially simpler result is obtained by making use of the potential energy formulation instead of the complementary energy formulation, as we have done above, since the infimum over η does not appear in the corresponding form for $\bar{U}^{(M+1)}$. However, such an expression will in general be harder to evaluate, because in plasticity the form of the energy function of the phases is generally more complicated than the corresponding form of the complementary energy functions, used in (4.41).

4.2. Upper bounds and estimates

It is clear that the approximate version of the new variational principle (4.7) will not be helpful in the determination of upper bounds for $\bar{U}$. Therefore, we must

resort to the exact version of the new variational principle (3.34), which makes the determination of upper bounds for $\bar{U}$ an intrinsically harder problem than the corresponding lower bound problem. In this subsection, we will attempt to go as far as we can with the problem of rigorous upper bounds, and then revert to the simpler problem of finding estimates for the upper bound, or "upper estimates". This is accomplished essentially by making use of the approximation (4.7) in spite of the difficulty just mentioned. In Section 4.3, we will provide some justification for making this approximation.

4.2.1. Reuss bound. We begin by considering the generally anisotropic composite made up of phases with energy functions (4.2) in prescribed volume fractions $c^{(n)}$, without any further restriction of the microstructure. Then, the effective complementary energy function $\bar{U}_0$ of the linear comparison composite with arbitrarily inhomogeneous moduli $\mu_0(\mathbf{x})$ and $\kappa_0(\mathbf{x})$ in prescribed volume fractions $c^{(n)}$ can be shown to be bounded above by

$$\bar{U}_0^{(R)}(\bar{\sigma}) = \frac{1}{6\mu_0^{(R)}} \bar{\sigma}_i^2 + \frac{1}{2\kappa_0^{(R)}} \bar{\sigma}_m^2, \quad (4.42)$$

where

$$\mu_0^{(R)} = \left(\int_{\Omega} \frac{1}{\mu_0(\mathbf{x})} dx \right)^{-1} \quad \text{and} \quad \kappa_0^{(R)} = \left(\int_{\Omega} \frac{1}{\kappa_0(\mathbf{x})} dx \right)^{-1}. \quad (4.43)$$

Expressions (4.43) are the continuous versions of the standard (discrete) Reuss estimates.

Correspondingly, from (3.34), the effective complementary energy function of the nonlinear composite $\bar{U}$ is bounded above by

$$\bar{U}^{(R)}(\bar{\sigma}) = \sup_{\mu_0(\mathbf{x}) \geq 0} \left\{ \frac{1}{6\mu_0} \bar{\sigma}_i^2 - \int_{\Omega} v(\mathbf{x}; \mu_0(\mathbf{x})) dx \right\} + \frac{1}{2\kappa^{(R)}} \bar{\sigma}_m^2, \quad (4.44)$$

where

$$v(\mathbf{x}; \mu_0) = \sup_{s \geq 0} \left\{ \frac{1}{6\mu_0} s^2 - \varphi(\mathbf{x}; s) \right\}, \quad (4.45)$$

and where

$$\kappa^{(R)} = \left(\sum_{n=1}^N \frac{c^{(n)}}{\kappa^{(n)}} \right)^{-1},$$

since the hydrostatic component of the stress-strain relation of the nonlinear composite is taken to be linear [see (4.2)].

Additionally, we have that

$$\varphi(\mathbf{x}; s) = \sup_{\mu_0 \geq 0} \left\{ \frac{1}{6\mu_0} s^2 - v(\mathbf{x}; \mu_0) \right\}, \quad (4.46)$$

which allows further simplification of (4.44), since

$$\begin{aligned} \bar{U}^{(R)}(\bar{\sigma}) &= \sup_{\mu_0(\mathbf{x}) \geq 0} \left\{ \int_{\Omega} \left[\frac{1}{6\mu_0(\mathbf{x})} \bar{\sigma}_i^2 - v(\mathbf{x}; \mu_0(\mathbf{x})) \right] dx + \frac{1}{2\kappa^{(R)}} \bar{\sigma}_m^2 \right\} \\ &= \int_{\Omega} \sup_{\mu_0(\mathbf{x}) \geq 0} \left\{ \frac{1}{6\mu_0(\mathbf{x})} \bar{\sigma}_i^2 - v(\mathbf{x}; \mu_0(\mathbf{x})) \right\} dx + \frac{1}{2\kappa^{(R)}} \bar{\sigma}_m^2 \\ &= \int_{\Omega} \varphi(\mathbf{x}; \bar{\sigma}_i) dx + \frac{1}{2\kappa^{(R)}} \bar{\sigma}_m^2, \end{aligned} \quad (4.47)$$

and therefore

$$\bar{U}^{(R)}(\bar{\sigma}) = \sum_{n=1}^N c^{(n)} \varphi^{(n)}(\bar{\sigma}_i) + \frac{1}{2\kappa^{(R)}} \bar{\sigma}_m^2. \quad (4.48)$$

This last result is the Reuss estimate for the nonlinear composite, which could have been obtained from the principle of minimum complementary energy, applied directly to the nonlinear composite. The present, less direct, derivation of this result via the new variational principle serves to demonstrate that the new method can at least recover the classical bounds exactly. This derivation serves further to illustrate how the new variational principle may be used in other contexts for which upper bounds may not be available via the classical principles.

Interestingly, we remark that the above result for the nonlinear Reuss bound (4.48) could have also been obtained by means of the approximation (4.7), following a procedure analogous to that used in the derivation of the nonlinear Voigt bound (4.15). This suggests that the approximation introduced in (4.7) is exact in this case. We will give some insight to the reasons behind this phenomenon in Section 4.3.

4.2.2. Hashin-Shtrikman upper estimates. We consider next the upper bound problem for the class of nonlinear composites with phases with potentials (4.2) in fixed volume fractions (4.4), distributed in such a way that the effective behavior of the composite is isotropic. In this case, however, direct application of the exact version of the new variational principle (3.34) leads to a problem that is much harder than the one solved above for the Reuss bound for anisotropic microstructures. For this reason, we will not pursue this approach here, and resort instead to the use of the approximation introduced in (4.7). Then, upper bounds for the effective energy $\bar{U}_0$ of the class of isotropic linear comparison composites with prescribed volume fractions led only to estimates for the upper bound (not rigorous upper bounds) of the class of isotropic nonlinear composites with prescribed volume fractions. We will refer to these estimates as "upper estimates" since they may be close to the upper bound. (In fact, the corresponding computation for the Reuss upper bound suggests that in some cases these estimates may yield rigorous bounds.) Further, in practical applications, we are rarely interested in precise bounds; instead, estimates are often preferred.

The computation of the "upper estimates" follows a procedure completely analogous to the computation of the lower bounds. Thus, the effective energy function of the linear comparison composite $\bar{U}_0$ is bounded above by

$$\bar{U}_0^{(MS+1)}(\bar{\sigma}) = \frac{1}{6\mu_0^{(MS+1)}} \bar{\sigma}^2 + \frac{1}{2\kappa_0^{(MS+1)}} \bar{\sigma}_m^2, \quad (4.49)$$

where $\mu_0^{(MS+1)}$ and $\kappa_0^{(MS+1)}$ are given by expressions similar to (4.19), except that $\mu_0^{(n)}$, $\mu_0^{(n+1)}$ and $\kappa_0^{(n+1)}$ must be replaced by

$$\mu_0^{(n+1)} = \frac{8\mu_0^{(n)} + 9\kappa_0^{(n-1)}}{6(2\mu_0^{(n)} + \kappa_0^{(n-1)})}, \quad \mu_0^{(n)} = \min\{\mu_0^{(n)}\} \quad \text{and} \quad \kappa_0^{(n-1)} = \min\{\kappa_0^{(n)}\}.$$

Then, application of this upper bound for $\bar{U}_0$ in (4.7) leads to the "upper estimate" $\bar{U}_0^{(MS+1)}$ for $\bar{U}$. The final result for $\bar{U}_0^{(MS+1)}$ is

$$\begin{aligned} \bar{U}_0^{(MS+1)}(\bar{\sigma}) = \max_i \left\{ \inf_{\omega^{(i)}, \bar{\sigma}_m} \left\{ \sum_{j=1}^n c^{(i)} \varphi^{(i)}(1 - \omega^{(i)}) \bar{\sigma}_m + c^{(ii)} \Delta_0^{(i)} \right. \right. \\ \left. \left. + \left(\sum_{j=1}^n \frac{c^{(i)}}{\kappa_0^{(i)}} (1 - \omega^{(i)})^2 \frac{\bar{\sigma}_m^2}{2} \right) \right\} \right\}, \quad (4.50) \end{aligned}$$

where $\Delta_0^{(i)}$ is as given by (4.28) and (4.29), but with $\kappa_0^{(n-1)}$ replaced by $\kappa_0^{(n)}$. Note that, apart from this modification, the expression (4.50) for $\bar{U}_0^{(MS+1)}$ differs from the expression (4.27) for $\bar{U}_0^{(MS)}$ only in that the outermost minimum over all phases has been replaced by a maximum.

When all the phases are incompressible (so that $\kappa_0^{(n-1)} \rightarrow \infty$), the above result reduces to

$$\bar{U}_0^{(MS+1)}(\bar{\sigma}) = \max_i \left\{ \inf_{\omega^{(i)}, \bar{\sigma}_m} \left\{ \sum_{j=1}^n c^{(i)} \varphi^{(i)}(s^{(i)}) + c^{(ii)} \varphi^{(ii)}(s^{(ii)}) \right\} \right\}, \quad (4.51)$$

where $s^{(i)}$ and $s^{(ii)}$ are given by (4.31), and (4.33), respectively. Note that the only difference in this case between the lower bound and the upper estimate is that the outermost minimum in (4.32) is replaced by a maximum in (4.51). Thus, from a computational point of view, the lower bound and upper estimates should be computed together since they involve the same "branches" which must be minimized in one case, and maximized in the other. A special case of the upper estimate (4.51) has been discussed previously by PONTE CASTAÑEDA (1991b) for two-phase composites with a linear and a nonlinear phase. A special case is the rigidly reinforced material, which has

$$\bar{U}_0^{(MS+1)}(\bar{\sigma}) = c^{(1)} \varphi^{(1)} \left(\frac{\bar{\sigma}_m}{\sqrt{1 + \frac{1}{3} c^{(1)}}} \right), \quad (4.52)$$

where phase 2 has been selected as the reinforcement phase.

Finally, an upper estimate for the incompressible fiber-reinforced composite with transversely isotropic symmetry is given by the same relation (4.36) for the lower bound, together with (4.37), except that the minimum over all phases is replaced by a maximum.

4.2.3. Higher-order upper estimates. Higher-order "upper estimates" for two-phase nonlinear composites are obtained by making use in (4.7) of the lower bounds of Milton for a linear comparison composite with the same distribution of phases as the nonlinear composite. These bounds for the shear modulus $\mu_0^{(M)}$ and bulk modulus $\kappa_0^{(M)}$ admit the same forms (4.38) and (4.39) as the corresponding upper bounds $\mu_0^{(M-1)}$ and $\kappa_0^{(M-1)}$, with the difference that $\mu_0^{(n)}$ must be replaced by $\mu_0^{(n-1)} = \Xi/6$, where Ξ is given by expression (3.29) in MILTON (1982), and $\kappa_0^{(n-1)}$ is to be replaced by

$$\kappa_0^{(n-1)} = \frac{4}{3} \left(\sum_{i=1}^n \frac{\eta^{(i)}}{\kappa_0^{(i)}} \right)^{-1}.$$

These results depend also only on the two geometric parameters $\eta^{(i)}$ and $\zeta^{(i)}$, in addition to the phase volume fractions $c^{(i)}$ and $c^{(2)}$.

As was the case for the lower bound $\bar{U}_0^{(M-1)}$, the expression for the corresponding upper estimate $\bar{U}_0^{(M)}$ is very complicated, except for the case where the two phases are incompressible. In this case, the expression for $\mu_0^{(M)}$ reduces to

$$\mu_0^{(M)} = \frac{3}{2} \left(\sum_{i=1}^n \frac{\eta^{(i)}}{\mu_0^{(i)}} \right)^{-1}, \quad (4.53)$$

which depends also only on $\eta^{(i)}$, and we obtain the upper estimate

$$\begin{aligned} \bar{U}_0^{(M)}(\bar{\sigma}) = \inf_{\omega} \left\{ c^{(1)} \varphi^{(1)} \left(\sqrt{1 - c^{(2)} \omega} \right) + \frac{3}{2} c^{(2)} \eta^{(2)} \omega^2 \bar{\sigma}_m + \dots \right. \\ \left. + c^{(2)} \varphi^{(2)} \left(\sqrt{1 + c^{(1)} \omega} \right) + \frac{3}{2} c^{(1)} \eta^{(1)} \omega^2 \bar{\sigma}_m \right\}, \quad (4.54) \end{aligned}$$

which involves only one optimization. The corresponding nonlinear Hashin-Shtrikman upper estimate is obtained by choosing either $\eta^{(1)} = 0$, or $\eta^{(1)} = 1$, whichever yields the higher value.

4.3. Attainability of the bounds

In this section, we address briefly the question of *optimality* of the nonlinear Voigt-Reuss and Hashin-Shtrikman bounds. By this we mean whether specific microstructures exist, within a given class of composites, which have effective energy functions $\bar{U}$ attaining the bounds (for the given class of composites). Further, in the case of anisotropic composites, microstructures saturating the bounds must exist for every possible loading configuration, in order for the bound to be optimal. If the bound is saturated only for special loading configurations, the bound is said to be *sharp* (but not optimal). We remark that, even in the context of linear elastic composites, there are still many open questions in connection with the optimality of bounds, and most of the results that are known to date are for two-phase systems. For this reason, we will limit our attention in this section to two-phase nonlinear composites. We further

note that in the context of linear elasticity, the knowledge of optimal bounds for the effective energy functions of a given class of composites does not suffice in general to characterize the set of all possible elasticity tensors within the class of composites, also known as the G -closure. However, in some special cases, with sufficient symmetry, optimal bounds for the effective energy function do suffice to characterize the G -closure.

By construction, the nonlinear bounds can only be optimal (sharp), if the corresponding linear bounds are optimal (sharp). The microstructures that are commonly used in the context of linear elastic composites to prove optimality are *sequentially laminated materials* (see KOHN and MURTON, 1988). These are hierarchical microstructures obtained by layering in a different orientation the result of a previous lamination procedure with one of the original (or new) phases. Thus, a rank I laminate (Fig. 1a) with orientation n_1 is produced by layering the two phases in prescribed proportions, with a certain layer thickness $\delta_1 \ll 1$. A rank II laminate (Fig. 1b) is obtained by layering the rank I laminate with one of the initial phases (or with a third phase) in a different direction n_2 , in given proportions, with layer thickness $\delta_2 \ll 1$, such that $\delta_1, \delta_2 \ll 1$. Higher rank laminates may be obtained by iterating this procedure. The main advantage of this class of microstructures is that their effective properties can be computed exactly, because the fields are piecewise constant within each elemental layer in the composite.

For the same reason, sequentially laminated microstructures would also appear to be useful in the context of nonlinear composites. Further, the new variational principle seems to be ideally suited for these microstructures because the fields are piecewise constant within each layer, and therefore a piecewise constant choice for the comparison moduli (within that layer) would lead to exact results. Thus, we can see that the effective energy $\bar{U}$ of a two-phase, nonlinear, rank I laminate (Fig. 1a) can be computed exactly via the new variational principle in terms of a two-phase, linear, rank I laminate. However, for a *two-phase, nonlinear, rank II laminate* (Fig. 1b), the effective energy of a *three-phase, linear, rank II laminate* is required in order to compute the effective energy of the nonlinear laminate exactly in terms of the new variational principle. (This is because the fields are different constants within different layers in the laminate, even if they are made of the same nonlinear material.) This observation has the implication that nonlinear bounds that are obtained from optimal linear bounds involving sequentially laminated microstructures of a rank higher than one are probably not optimal. This is because the microstructures that attain the corresponding linear bounds, from which the nonlinear bounds are derived, cannot be optimal in general for the nonlinear materials. Only in the (unusual) situation where the optimal (sharp) microgeometry is a rank I laminate can we be sure of the corresponding optimality (sharpness) of the nonlinear bound. Additionally, in the special case where the "matrix" phase is linear, the nonlinear lower bound (on $\bar{U}$) is also optimal, even when the optimal microstructures are laminates of rank higher than one (see PONTE CASTAÑEDA, 1991b). This is because the optimal choice for the moduli of the comparison composite within linear phases in the nonlinear composite are the same as the actual moduli of the linear phases. In the following subsections we will address the optimality of the nonlinear Voigt/Reuss bounds, and Hashin-Shtrikman bounds by means of the above-mentioned criteria.

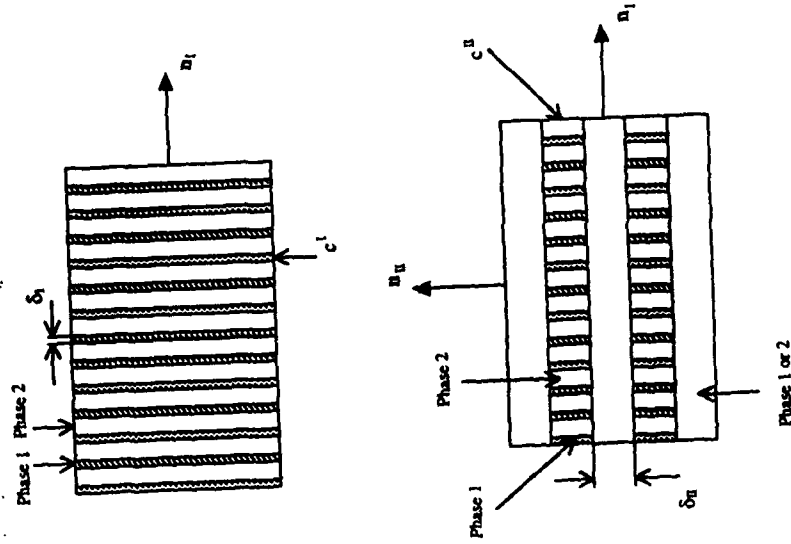


FIG. 1. (a) A rank I laminate and (b) a rank II laminate.

4.3.1. *Voigt/Reuss bounds.* For linear-elastic, two-phase, generally anisotropic composites with prescribed volume fractions, the Voigt/Reuss bounds on the effective energy function $\bar{U}$ are known to be *non-optimal*. GIBIANSKI and CHERKAEV (1987) (see also AVELLANEDA, 1987) have determined optimal bounds for the effective energy function $\bar{U}$ for this class of composites. KOHN and LIPTON (1988) have found optimal bounds for the subclass of incompressible two-phase composites, and determined additionally that the Voigt (but not the Reuss) bound is actually optimal in this special case. (Although the new variational principle could in principle be used in conjunction with these optimal bounds to obtain corresponding nonlinear bounds, these optimal linear bounds have rather complicated forms, and that is why the simpler Voigt/Reuss bounds were used here instead.) Thus, the nonlinear Voigt/Reuss bounds are not

optimal in general. However, the linear Voigt bound for the two-phase, incompressible composite does involve rank I laminate optimal microstructure, and by the criteria established in the previous paragraph, the nonlinear Voigt bound is also optimal for the class of two-phase, incompressible composites (but not more generally). Although the nonlinear Reuss bound is not optimal, it is sharp under special loading configurations [see KOHN and LIPSON (1988) for the corresponding result for linear elastic composites]. We add that simple expressions for the effective properties of nonlinear, rank I laminates have been determined by DEBORTON and PONTE CASTAÑEDA (1991). Their results are in agreement with the conclusions of this subsection.

4.3.2. Hashin-Shtrikman bounds and estimates. For linear-elastic, two-phase, isotropic composites with prescribed volume fractions, the Hashin-Shtrikman bounds on the effective energy function $\bar{U}$ are optimal. This was demonstrated by FRANCFORT and MURAT (1986). However, the corresponding optimal microstructures involve laminates of a rank higher than one, and therefore these microstructures do not serve to attain the nonlinear Hashin-Shtrikman lower bound on $\bar{U}$ (as explained above). An exception is provided by the two-phase, nonlinear composite with one linear phase: in this case, the lower bound can be shown to be optimal (see PONTE CASTAÑEDA, 1991b). Finally, we note that in general the nonlinear "upper estimate" for $\bar{U}$ is probably as far below the optimal upper bound, as the lower bound given above is below the optimal lower bound (and hence the "upper estimate" cannot be a rigorous upper bound). Thus, if we ignore the rigorous interpretation of the Hashin-Shtrikman estimates as bounds, and think of them instead as approximate estimates of the strongest and weakest composites in the given class of composites, then in practice both of them should probably be held on an equal footing, and it is only on a rigorous sense that the lower bound is superior to the upper estimate.

5. CONCLUDING REMARKS

In this paper, we have introduced dual versions of a new variational principle, based on the principles of minimum potential and complementary energy, allowing the estimation of the effective behavior of *nonlinear* composites with isotropic phases in prescribed volume fractions. The estimation of the effective behavior of the nonlinear composites is in terms of the effective energy functions of suitably optimized linear comparison composites. These new variational principles generalize a procedure introduced by PONTE CASTAÑEDA (1991a) for bounding the effective properties of nonlinear composites.

The variational principles have been used to derive bounds and estimates for the effective properties of certain classes of nonlinear composites. Thus, upper and lower bounds of the Voigt and Reuss type have been derived for the effective complementary-energy function $\bar{U}$ of generally anisotropic composites, with multiple phases in prescribed volume fractions. Although these bounds are classical, a novel form has been proposed for the Voigt bound. The nonlinear Voigt/Reuss bounds were found to be in general *not* optimal. Additionally, nonlinear lower bounds of the Hashin-Shtrikman type have been given for the effective complementary-energy function $\bar{U}$ of the classes

of isotropic composites, and transversely isotropic, fiber-reinforced composites, respectively. Although bounds of this type are not new, and can be derived alternatively from the Talbot-Willis variational procedure, the form of the bounds given here is both new and is perhaps the simplest available thus far for general multiple-phase composites. We note in this context that most of the explicit results to date have dealt exclusively with porous and rigidly reinforced materials. The Hashin-Shtrikman lower bounds are probably *not* optimal in general. We have also proposed that estimates obtained by making use of the Hashin-Shtrikman upper bounds for the linear comparison composite into the approximate version of the new variational principle could be interpreted as "upper estimates" (not rigorous upper bounds) for the effective energy function of nonlinear composites. Finally, third-order nonlinear bounds of the Beran type have also been determined for the first time for the class of isotropic, two-phase composites with prescribed volume fractions and given Milton geometric parameters. When both phases are incompressible, remarkably simple forms result for the lower bound and the upper estimate. Clearly, many possibilities exist for further investigation. Central, and probably most difficult, among them is the determination of rigorous upper bounds of the Hashin-Shtrikman and higher-order types (for the effective complementary-energy function of nonlinear composites). Also, extensions of the new variational principles are needed to deal more precisely with nonlinear composites with anisotropic phases, such as are present in ductile polycrystalline media.

ACKNOWLEDGEMENTS

This research was supported by the Air Force Office of Scientific Research (Grant No. 91-0161). Additional support in the form of computing equipment by the Research Foundation of the University of Pennsylvania is acknowledged. Thanks are due to Professor J. R. Willis of the University of Bath for useful discussions.

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APPENDIX I

The non-negative functions ϕ and f introduced in Section 3 are defined on $\Omega \times R^+ \times R^+$. On the other hand, the usual definition of the polar function refers to functions defined on multiple copies of R (the extended set of reals). In this appendix, we show how to define polars for functions of the type of ϕ and f , defined on multiple copies of R^+ . For simplicity, we carry out the development for a one-dimensional function (and drop the dependence on x); similar developments hold for multiple-dimensional functions. Also, we consider only the case of the convex polar function; analogous results hold for the concave polar function. Thus, we consider a function $\tau: R^+ \rightarrow R^+$ satisfying the condition $\tau(0) = 0$, and define its extension to R by, say

$$\tilde{\tau}(x) = \tau(|x|). \quad (1.1)$$

where $\tilde{\tau}: R \rightarrow R^+$ is also seen to be a non-negative function satisfying $\tilde{\tau}(0) = 0$.

The convex polar function of $\tilde{\tau}$ is then defined in the usual way by

$$\tau^*(y) = \sup_x \{xy - \tilde{\tau}(x)\}. \quad (1.2)$$

Now, for $y \geq 0$, we have that

$$\begin{aligned} \tau^*(y) &= \sup_{x \geq 0} \{xy - \tilde{\tau}(x)\} \\ &= \sup_{x \geq 0} \{xy - \tau(x)\}, \end{aligned} \quad (1.3)$$

where we have also used the fact that $\tilde{\tau}$ is the non-negative and $\tilde{\tau}(0) = 0$. Note further that τ^*

is also non-negative, and such that $\tau^*(0) = 0$. Therefore, it makes sense to define $\tau^*: R^+ \rightarrow R^+$ via

$$\tau^*(y) = \sup_{x \geq 0} \{xy - \tau(x)\}. \quad (1.4)$$

Finally, it follows from the corresponding duality relation for $\tilde{\tau}$ that

$$\tau(x) \geq \sup_{y \geq 0} \{xy - \tau^*(y)\}, \quad (1.5)$$

with equality if τ is convex.

APPENDIX II

The main result proved in this appendix is that *convexity* of f implies *convexity* of g . The demonstration of this result has two parts. We begin by recalling some result from Section 3. Thus, relation (3.3) can be written (dropping the dependence on x)

$$\psi(\sigma, \sigma_m) = \sup_{\epsilon, \epsilon_m \geq 0} \{3\sigma_m \epsilon + 3\sigma_m \epsilon_m - \phi(\epsilon, \epsilon_m)\}, \quad (11.1)$$

and rewritten

$$g(\epsilon, \epsilon_m) = \sup_{\sigma, \sigma_m \geq 0} \{3\sqrt{u_m \epsilon} + 3\sqrt{u_m \epsilon_m} - f(u, u_m)\}, \quad (11.2)$$

in terms of f and g . Similarly, relation (3.6) can be written

$$\phi(\epsilon, \epsilon_m) = \sup_{\sigma, \sigma_m \geq 0} \{3\sigma_m \epsilon + 3\sigma_m \epsilon_m - \psi(\sigma, \sigma_m)\}, \quad (11.3)$$

which in turn yields

$$f(u, u_m) = \sup_{\epsilon, \epsilon_m \geq 0} \{3\sqrt{u_m \epsilon} + 3\sqrt{u_m \epsilon_m} - g(\epsilon, \epsilon_m)\}. \quad (11.4)$$

The first part of the demonstration follows from

$$\begin{aligned} f_0(p, p_m) &= \inf_{\epsilon, \epsilon_m \geq 0} \{u_m p + u_m p_m - f(u, u_m)\} \\ &= \inf_{\epsilon, \epsilon_m \geq 0} \{u_m p + u_m p_m - \sup_{\epsilon, \epsilon_m \geq 0} \{3\sqrt{u_m \epsilon} + 3\sqrt{u_m \epsilon_m} - g(\epsilon, \epsilon_m)\}\} \\ &= \inf_{\epsilon, \epsilon_m \geq 0} \left\{ \inf_{\epsilon, \epsilon_m \geq 0} \{u_m p + u_m p_m - 3\sqrt{u_m \epsilon} - 3\sqrt{u_m \epsilon_m}\} + g(\epsilon, \epsilon_m) \right\}, \end{aligned} \quad (11.5)$$

where we have used (11.4), the fact that $-\sup\{h\} = \inf\{-h\}$, and interchanged the resulting infimum over the ϵ variables with the infimum over the u variables. But the innermost infimum is trivial to compute, and there results

$$\begin{aligned} f_0(p, p_m) &= \inf_{\epsilon, \epsilon_m \geq 0} \left\{ -\frac{1}{9} \frac{p}{p_m} - \frac{9}{4} \frac{p_m}{p_m} + g(\epsilon, \epsilon_m) \right\} \\ &= -\sup_{\epsilon, \epsilon_m \geq 0} \left\{ \frac{1}{9} \frac{p}{p_m} + \frac{9}{4} \frac{p_m}{p_m} - g(\epsilon, \epsilon_m) \right\}, \end{aligned} \quad (11.6)$$

which leads to the important identity

$$f_0(p, p_m) = -g^*\left(\frac{1}{9p}, \frac{9}{4p_m}\right). \quad (11.7)$$

The second and final part of the demonstration makes use of (11.2) and of the fact that f is concave (so that $f = f_{**}$) to obtain

$$\begin{aligned}
g(t, r_m) &= \sup_{u_m, p_m} \left\{ 3\sqrt{u_m r_m} + 3\sqrt{u_m r_m} - \inf_{p_m, p_m} \{u_m p_m + u_m p_m - f_0(p_m, p_m)\} \right\} \\
&= \sup_{p_m, p_m} \left\{ \sup_{u_m, p_m} \{3\sqrt{u_m r_m} + 3\sqrt{u_m r_m} - u_m p_m - u_m p_m\} + f_0(p_m, p_m) \right\}, \quad (II.8)
\end{aligned}$$

where we have interchanged the suprema. The innermost supremum is the negative of the innermost infimum in (II.5), and therefore making use of (II.7), we deduce that

$$\begin{aligned}
g(t, r_m) &= \sup_{p_m, p_m} \left\{ \frac{1}{9} \frac{r_m}{p_m} + \frac{1}{4} \frac{r_m}{p_m} + f_0(p_m, p_m) \right\} \\
&= \sup_{p_m, p_m} \left\{ \frac{1}{9} \frac{r_m}{p_m} + \frac{1}{4} \frac{r_m}{p_m} - g^*(\frac{1}{9} \frac{r_m}{p_m}, \frac{1}{4} \frac{r_m}{p_m}) \right\} \\
&= g^{**}(t, r_m). \quad (II.9)
\end{aligned}$$

This final result proves that g is convex, and hence completes the proof.

APPENDIX III

In this appendix we give a simple proof of the identity (4.12) used throughout the body of the paper. We begin by introducing a Lagrange multiplier to account for the zero-average constraint on the optimization variables, $\omega^{(n)}$. Thus,

$$\inf_{\omega^{(n)}} \left\{ \sum_{i=1}^N \frac{c^{(n)}}{\mu^{(n)}} (1 - \omega^{(n)})^2 \right\} = \inf_{\omega^{(n)}} \left\{ \sum_{i=1}^N \frac{c^{(n)}}{\mu^{(n)}} (1 - \omega^{(n)})^2 + \sup_i \left\{ \lambda \sum_{i=1}^N c^{(n)} \omega^{(n)} \right\} \right\}, \quad (III.1)$$

and, by means of the Saddle Point theorem, we have

$$\inf_{\omega^{(n)}} \left\{ \sum_{i=1}^N \frac{c^{(n)}}{\mu^{(n)}} (1 - \omega^{(n)})^2 \right\} = \sup_i \left\{ \inf_{\omega^{(n)}} \left\{ \sum_{i=1}^N \frac{c^{(n)}}{\mu^{(n)}} (1 - \omega^{(n)})^2 + \lambda \sum_{i=1}^N c^{(n)} \omega^{(n)} \right\} \right\}, \quad (III.2)$$

The infimum over the $\omega^{(n)}$ is satisfied by

$$\omega^{(n)} = 1 - \frac{\lambda}{2} \mu^{(n)}. \quad (III.3)$$

which finally leads to

$$\begin{aligned}
\inf_{\omega^{(n)}} \left\{ \sum_{i=1}^N \frac{c^{(n)}}{\mu^{(n)}} (1 - \omega^{(n)})^2 \right\} &= \sup_i \left\{ \sum_{i=1}^N c^{(n)} \left(1 - \frac{\lambda}{2} \mu^{(n)} \right)^2 \right\} \\
&= \sup_i \left\{ \lambda - \frac{\lambda^2}{4} \sum_{i=1}^N c^{(n)} \mu^{(n)} \right\} \\
&= \left(\sum_{i=1}^N c^{(n)} \mu^{(n)} \right)^{-1}. \quad (III.4)
\end{aligned}$$

Reference [21]

EFFECTIVE ANISOTROPIC PROPERTIES OF CREEPING COMPOSITES

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ABSTRACT

A recently developed variational principle for estimating the effective properties of nonlinear composites in terms of the corresponding properties of linear composites with the same microstructural distributions of phases is applied to two model anisotropic composite materials. The model materials considered are laminated materials and fibre-reinforced materials, and they correspond in the dilute limit of the inclusion phase to materials reinforced by aligned platelets and fibers, respectively. For simplicity, the power exponent of one of the phases will be taken to be unity corresponding to linear behavior, while the other phase will be assumed to satisfy a pure-power creeping law. Both phases will be assumed to be isotropic and incompressible.

INTRODUCTION

A new procedure for estimating the effective properties of composite materials with phases exhibiting nonlinear constitutive behavior has been proposed recently by Ponte Castañeda [6]. The straightforward and versatile procedure expresses the effective properties of the nonlinear composite in terms of the effective properties of a family of linear composites with the same distribution of phases as the nonlinear composite. Thus, bounds and estimates for the effective properties of linear composites can be translated directly into bounds and estimates for the corresponding nonlinear composite. Appropriate references for the linear theory of composites are provided by the works of Christensen [1]. The new procedure was applied in the above reference [6] to composite materials containing a nonlinear isotropic matrix either weakened or reinforced by isotropic distributions of voids or rigid particles, respectively. The case of a ductile matrix reinforced by incompressible elastic particles was studied in [7]. The results of these studies were given in the form of estimates and rigorous bounds for the effective properties of such materials. The Hashin-Shtrikman bounds obtained via the new method directly from the Hashin-Shtrikman [3] bounds for the linear comparison material were found to be an improvement over the corresponding bounds obtained in [8] for the same class of nonlinear materials using the nonlinear extension of the Hashin-Shtrikman variational principle [3] proposed by Talbot and Willis [9]. Recently, however, Willis [11] has shown that the bounds obtained via the new method can also be obtained by the Talbot-Willis method with a better choice of their comparison material. More generally, the new procedure can make use of other bounds and estimates for the linear comparison material to yield corresponding bounds and estimates for nonlinear materials. In fact, the new procedure can be shown [2] to yield exact results for nonlinear composites with special microstructures.

In this paper, we apply the new procedure to composite materials containing a creeping phase of the power-law type and a linearly creeping phase. Although the phases will be assumed to be isotropic, the composite itself will be assumed to be *anisotropic* by identifying

preferred orientations in the microstructural distribution of the phases. Two examples will be considered, both of which have transversely isotropic overall symmetries: laminated materials and fibre-reinforced materials. The first class of materials can be given the interpretation of materials reinforced by aligned platelets for dilute concentrations of the linear phase; whereas, the second class of materials can be given the interpretation of materials reinforced by short fibers for dilute concentrations of the linear phase. For simplicity, we assume that the phases are incompressible and perfectly bonded to each other. In the next three sections, we will give, respectively, a brief definition of effective properties, an introduction to the new variational principle of [6], and application of the principle to the two classes of materials discussed above. More detailed studies including specific results for the nonlinear laminates and fiber-reinforced materials can be found in [2] and [5], respectively.

EFFECTIVE PROPERTIES

Consider a two-phase composite occupying a region of unit volume Ω , such that the local stress potential $U(\boldsymbol{\sigma}, \mathbf{x})$ is expressed in terms of the homogeneous phase potentials $U^{(r)}(\boldsymbol{\sigma})$ via

$$U(\boldsymbol{\sigma}, \mathbf{x}) = \sum_{r=1}^2 \chi^{(r)}(\mathbf{x}) U^{(r)}(\boldsymbol{\sigma}), \quad (1)$$

where $\chi^{(r)}$ is the characteristic function of phase r . The phases are assumed to be incompressible and isotropic, so that the potentials $U^{(r)}(\boldsymbol{\sigma})$ can be assumed to depend only on the effective stress $\sigma_e = \sqrt{\frac{3}{2} \mathbf{S} \cdot \mathbf{S}}$, where $\mathbf{S}$ is the deviator of $\boldsymbol{\sigma}$. Thus, we write $U^{(r)}(\boldsymbol{\sigma}) = f^{(r)}(\sigma_e)$, where the $f^{(r)}$ are scalar-valued functions. Then, the local constitutive relation for the creeping material is given by

$$\mathbf{D} = \frac{\partial U}{\partial \boldsymbol{\sigma}}(\boldsymbol{\sigma}, \mathbf{x}), \quad (2)$$

where $\mathbf{D}$ is the rate-of-deformation (strain-rate) tensor.

To define the effective properties of the heterogeneous material we introduce, following Hill [4], a uniform constraint boundary condition

$$\boldsymbol{\sigma} \mathbf{n} = \bar{\boldsymbol{\sigma}} \mathbf{n}, \quad \mathbf{x} \in \partial\Omega, \quad (3)$$

where $\partial\Omega$ denotes the boundary of the composite, $\mathbf{n}$ is its unit outward normal, and $\bar{\boldsymbol{\sigma}}$ is a given constant symmetric tensor. It follows that the average stress is precisely $\bar{\boldsymbol{\sigma}}$, and we define the average strain-rate $\bar{\mathbf{D}}$ in a similar manner.

Then, the effective behavior of the composite, or the relation between the average stress and the average strain-rate follows from the principle of minimum complementary energy, which can be stated in the form

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma} \in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U(\boldsymbol{\sigma}, \mathbf{x}) dV, \quad (4)$$

where $S(\bar{\boldsymbol{\sigma}}) = \{\boldsymbol{\sigma} | \sigma_{ij,j} = 0 \text{ in } \Omega, \text{ and } \sigma_{ij} n_j = \bar{\sigma}_{ij} n_j \text{ on } \partial\Omega\}$ is the set of statically admissible stresses, and where we have assumed convexity of the nonlinear potential $U(\boldsymbol{\sigma}, \mathbf{x})$. Thus, assuming that $\tilde{U}(\bar{\boldsymbol{\sigma}})$ is differentiable, we have that

$$\bar{\mathbf{D}} = \frac{\partial \tilde{U}}{\partial \bar{\boldsymbol{\sigma}}}(\bar{\boldsymbol{\sigma}}, \mathbf{x}). \quad (5)$$

The task will be to determine bounds and estimates for $\tilde{U}(\bar{\boldsymbol{\sigma}})$, which is known to be convex.

NONLINEAR VARIATIONAL PRINCIPLES

A new variational principle for determining bounds and estimates for the effective properties of nonlinear composites in terms of the effective properties of linear composites was proposed by Ponte Castañeda [6]. In this section, we specialize this result for the case where both phases are incompressible, and phase 2 is linear so that

$$U^{(2)}(\sigma) = \frac{1}{6\mu^{(2)}} \sigma^2.$$

The new variational principle is based on a representation of the potential of the nonlinear material in terms of the potentials of a family of linear *comparison* materials. Thus, for a homogeneous nonlinear material with "stronger than quadratic" growth in its potential, $U(\sigma)$, and certain additional convexity hypothesis, we have that

$$U(\sigma) = \max_{\mu > 0} \{U_o(\sigma) - V(\mu)\}, \quad (6)$$

where

$$V(\mu) = \max_{\sigma} \{U_o(\sigma) - U(\sigma)\} \quad (7)$$

and where $U_o(\sigma)$ is the potential of a linear comparison material with shear modulus μ .

The new variational principle is obtained by making use of relation (6) applied to the nonlinear phase 1 in the complementary energy principle (4) to obtain the following relation for the nonlinear composite

$$\tilde{U}(\bar{\sigma}) = \max_{\mu^{(1)}(\mathbf{x})} \left\{ \tilde{U}_o(\bar{\sigma}) - \int_{\Omega^{(1)}} V^{(1)}(\mu^{(1)}) dV \right\}, \quad (8)$$

where

$$\tilde{U}_o(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} \int_{\Omega} U_o(\sigma, \mathbf{x}) dV, \quad (9)$$

is the effective potential of a linear comparison material with local potential $U_o(\sigma, \mathbf{x})$ and shear moduli $\mu^{(1)}$ and $\mu^{(2)}$ in phases 1 and 2, respectively. Further, we note that, in general, the comparison moduli $\mu^{(1)}$ are functions of $\mathbf{x}$.

The variational principle described by (8) roughly corresponds to solving a linear problem for a heterogeneous material with arbitrary moduli variation within the nonlinear phase, and then optimizing with respect to the variations in moduli within the nonlinear phase. Thus, the nonlinear material can be thought of as a "linear" material with variable moduli that are determined by prescription (8) in such a way that its properties agree at each $\mathbf{x}$ with those of the nonlinear material. This suggests that if the fields happen to be constant over the nonlinear phase, then the variable moduli $\mu^{(1)}(\mathbf{x})$ can be replaced by constant moduli $\mu^{(1)}$. More generally, however, we have the following lower bound for $\tilde{U}(\bar{\sigma})$, namely,

$$\tilde{U}_-(\bar{\sigma}) = \max_{\mu^{(1)} > 0} \{ \tilde{U}_o(\bar{\sigma}) - c^{(1)} V^{(1)}(\mu^{(1)}) \}, \quad (10)$$

where $c^{(1)}$ is the volume fraction of phase 1.

APPLICATION TO LAMINATES

In this section, we summarize from reference [2] the results of applying the new variational principle to a laminated material. For this geometry, it is well known that the stress and strain fields take on different constant values within each phase. This suggests that taking $\mu^{(1)}$ to be constant within the nonlinear phase will lead to an exact result as given by (8) or (10) (in this case $\tilde{U}$ and $\tilde{U}_-$ are identical). The key ingredient in this development is the exact result for the effective potential of the linear laminate. This result can be expressed in the form

$$\tilde{U}(\bar{\sigma}) = \frac{1}{6\hat{\mu}} \tau_n^2 + \frac{1}{6\bar{\mu}} \left[\tau_p^2 + (\sigma_n - \sigma_p)^2 \right], \quad (11)$$

where τ_n , τ_p , σ_n and σ_p are the transversely isotropic invariants of the stress tensor corresponding respectively to the out-of-plane tensile, in-plane hydrostatic, out-of-plane shear and in-plane shear stresses. Also, $\bar{\mu}$ and $\hat{\mu}$ stand, respectively, for the Voigt and Reuss estimates of the shear modulus.

Application of this result into the new variational principle can then be shown to lead to the following simple expression for the effective energy of the laminate

$$\tilde{U}(\bar{\sigma}) = \min_{\omega} \left\{ c^{(1)} f^{(1)}(s^{(1)}) + c^{(2)} f^{(2)}(s^{(2)}) \right\}, \quad (12)$$

where $s^{(1)}$ and $s^{(2)}$ are functions of ω given by the relations $s^{(1)} = \sqrt{(1 + c^{(2)}\omega)^2 (\sigma_e^2 - \tau_n^2) + \tau_n^2}$

and $s^{(2)} = \sqrt{(1 - c^{(1)}\omega)^2 (\sigma_e^2 - \tau_n^2) + \tau_n^2}$.

Analogous forms can be derived for the fibre-reinforced material, except that for this geometry, the effective energy cannot be given explicitly by (8), but only characterized in the form of bounds by means of (10). Detailed results of these calculations are given in [5], where they are also compared to the results of Talbot and Willis [10] for the same microstructure using the Talbot-Willis variational principle. Finally, both types of results can be averaged over all possible orientations to yield results depicting approximately the effect of inclusion shape on the effective properties of isotropic composites.

ACKNOWLEDGEMENTS

This work was supported by the Air Force Office of Scientific Research under grant 91-0161.

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Reference [6]

ON THE DUCTILITY OF LAMINATED MATERIALS

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(Received 18 November 1991; in revised form 2 March 1992)

Abstract—A laminated material is one of the few composite systems for which the effective constitutive behavior can be determined exactly. This is well known for laminated composites with linearly elastic phases in prescribed volume fractions. For these composites, explicit expressions for the effective moduli have been available for at least 30 years. However, it appears that corresponding expressions for the effective energy functions of laminated composites with phases exhibiting nonlinear constitutive behavior are currently unavailable. In this paper, we make straightforward use of a new variational procedure, recently developed by one of the authors, to obtain simple expressions for the effective energy functions of laminated composites with isotropic ductile phases in prescribed volume fractions. The same expressions are given an alternative derivation, starting directly from the classical variational principles. Explicit results are then computed for ductile/brittle systems, such as aluminum/alumina laminates, and also for laminated composites made up of two perfectly plastic phases with different yield stresses. The results—which are representative of other anisotropic geometries, such as fiber-reinforced solids—exhibit a strong coupling between different loading modes that is enhanced by material nonlinearity.

1. INTRODUCTION

This work is concerned with the determination of the effective constitutive behavior of laminated composite materials with plastically deforming phases in prescribed volume fractions. We will deal with the *exact* effective behavior of such materials, and therefore, we will exclude from our consideration the so-called approximate theories of laminated plates (Christensen, 1979). In the context of linear elasticity, laminated composites have been studied extensively in the literature. Postma (1955) and White and Angona (1955) concerned themselves with the study of two-phase, periodic laminates in connection with wave propagation in stratified media. Backus (1962) extended their results to multi-phase, nonperiodic composites, again in the wave propagation context. The extension to anisotropic layers was considered by Walpole (1969) for aligned, transversely isotropic phases, and by Chou *et al.* (1972) and Pagano (1974) for more general anisotropy of the phases. Recently, Norris (1991) has developed alternative expressions for the effective moduli tensor of laminated composites with generally anisotropic phases, exploiting the interior and exterior projection tensors of Hill (1972, 1983). Other related works include iterative formulae developed by Francfort and Murat (1986) in the context of linear elasticity allowing simple expressions for the effective moduli of multi-sandwich structures (laminates embedded within laminates of different orientations). These microstructures are of theoretical value in the demonstration of the optimality of bounds for the effective properties of composite materials with more general microstructures [see Kohn (1987) and Lipton (1991b)]. In spite of the large level of activity for linear laminates, briefly summarized above, the theory of nonlinear laminated composites does not seem to have been developed very much. To the knowledge of the authors, the only work thus far in this direction is a generalization of the Francfort–Murat formula for simple laminated materials with one nonlinear phase and one *linear* phase due to Kohn (1990) and Milton (1990). Nonlinear results do exist, however, within the context of the approximate laminated plate theories.

The justification of the study of nonlinear laminated composites may be partially understood in terms of the following considerations. First, it is a configuration of practical importance: for instance, the use of linear laminated theories in geophysical applications is well known, but it is also known that the properties of the materials composing the surface of the Earth may exhibit nonlinear constitutive behavior, particularly, deep within the surface, where the materials are subject to large compressive stresses. Second, we will

find that the laminated microstructure is illustrative of the significant coupling that may arise in nonlinear, anisotropic materials between different loading modes. This nonlinear coupling is also observed in other more complex microstructures, such as fiber-reinforced composites. Third, the laminated microstructure corresponds to the simplest possible type of anisotropic composite with nonlinear phases, in the sense that *exact* results may be obtained for its effective properties, as will be shown herein. In this connection, the present work should be considered in the light of a research program attempting to characterize the effective properties of nonlinear composites in general. Thus far, methods have been developed for understanding the effective behavior of nonlinear composites by Talbot and Willis (1985), and by Ponte Castañeda (1991a, 1992). These methods, although different in essence, can be shown to yield exactly the same results in some cases, but the new method is more general than the first in that it can be used to obtain estimates *other than* Hashin-Shtrikman bounds and self-consistent estimates [see Willis (1991) and Ponte Castañeda (1992)]. Bounds and estimates for the effective properties of nonlinear composites with isotropic overall symmetries have been obtained by Ponte Castañeda and Willis (1988), and Willis (1989) making use of the Talbot-Willis method, and by Ponte Castañeda (1991a, b) making use of the new method. Results for fiber-reinforced composites have also been developed very recently by Talbot and Willis (1991), and by Ponte Castañeda (1992) and deBotton and Ponte Castañeda (1992).

The rest of the paper is structured as follows. In Section 2 the definition of effective properties is reviewed, and their variational characterization is given in terms of both the classical and new variational principles of Ponte Castañeda (1991a, 1992). In Section 3 general nonlinear laminated composites are considered, and general formulae are derived in Sections 4 and 5 for the effective properties of incompressible and compressible laminates, respectively. Additionally, in Section 6, more specific results are given for two-phase laminates. In particular, the cases of ductile materials reinforced by linearly elastic layers, and of laminates with two perfectly plastic phases are considered. Finally, some additional relevant results are given in four appendices; in particular, in Appendix IV, an alternative derivation is given of the results of Sections 4 and 5 using the classical variational principle.

2. EFFECTIVE PROPERTIES AND THEIR VARIATIONAL CHARACTERIZATION

In this section, we are interested in the characterization of the effective, or overall, constitutive behavior of composites materials with plastically deforming phases. For our purposes, a *composite* is a heterogeneous material with two distinct length scales: one macroscopic, L , describing the gross size of the specimen and the scale of variation of the applied loading conditions, and a microscopic scale, l , characterizing the size of the typical inhomogeneity, such that $l \ll L$. More precise definitions can be found in the review article by Kohn (1987).

For simplicity, the constitutive behavior of the phases will be characterized by the deformation theory of plasticity, or equivalently by nonlinear infinitesimal elasticity. However, with minor changes in notation, the results of the analyses of this paper will also be relevant to the high-temperature creeping behaviour of composite laminates. Additionally, the results can be used in an *approximate* fashion to suggest yield functions for laminated composites in the context of the incremental theory of plasticity, as suggested by Duva and Hutchinson (1984), and other investigators.

In the following description of effective properties, the composite is assumed to occupy a domain of unit volume Ω , with boundary $\partial\Omega$. Then, the nonlinear plastic behavior of the composite is characterized by means of a complementary-energy density function, $U(\mathbf{x}, \boldsymbol{\sigma})$, depending on the position vector $\mathbf{x}$ and the stress field $\boldsymbol{\sigma}(\mathbf{x})$, in such a way that the strain field $\boldsymbol{\varepsilon}(\mathbf{x})$ is given by

$$\boldsymbol{\varepsilon}(\mathbf{x}) = \frac{\partial U(\mathbf{x}, \boldsymbol{\sigma})}{\partial \boldsymbol{\sigma}}. \quad (1)$$

Following Hill (1963), we define the *effective* constitutive behavior of the heterogeneous solid in terms of the analogous relation

$$\bar{\varepsilon} = \frac{\partial \tilde{U}}{\partial \bar{\sigma}}, \quad (2)$$

where $\bar{\varepsilon}$ denotes the mean value of the strain field over Ω , and $\tilde{U}$ refers to the normalized (recall that Ω has unit volume) complementary-energy function of the solid when subjected to the *uniform constraint* boundary condition

$$\sigma \mathbf{n} = \bar{\sigma} \mathbf{n}, \quad \mathbf{x} \in \partial\Omega, \quad (3)$$

where $\mathbf{n}$ is the outward unit normal to $\partial\Omega$, and $\bar{\sigma}$ is a constant, symmetric tensor. We recall that under this type of boundary condition, the mean value of the stress over Ω is precisely $\bar{\sigma}$.

The *effective* complementary energy function of the composite, $\tilde{U}$, can be obtained directly in terms of the principle of minimum complementary energy by means of

$$\tilde{U}(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} \int_{\Omega} U(\mathbf{x}, \sigma) dv, \quad (4)$$

where

$$S(\bar{\sigma}) = \{\sigma \mid \nabla \cdot \sigma = 0 \text{ in } \Omega, \text{ and } \sigma \mathbf{n} = \bar{\sigma} \mathbf{n} \text{ on } \partial\Omega\} \quad (5)$$

is the set of *statically* admissible stress fields. Note that the first set of conditions in (5) are the equilibrium equations, and that the minimizing conditions (Euler-Lagrange equations) of (4) are the compatibility equations. Further, composite materials typically exhibit sharp interfaces across which the material properties are discontinuous, although the phases are *assumed* to be perfectly bonded. Therefore, across such interfaces, the equilibrium equations must be reinterpreted in terms of continuity of the traction stresses, and correspondingly the compatibility equations must be replaced by continuity of the tangential components of the strain tensor.

We note that, given relation (2) in terms of $\tilde{U}$, the problem of characterizing the effective behavior of the composite reduces to that of determining $\tilde{U}$. However, while in principle $\tilde{U}$ can be computed from (4); in practice, this variational principle is not very useful for two reasons. First, usually the microstructure of a typical composite is not completely specified; and second, the problem described by (4) is a nonlinear one on account of the nonlinear behavior of the constituent phases. For the problem of interest in this paper, the first issue is not a concern because the phase volume fractions suffice to characterize the microstructure of a laminated composite material. However, the second issue presents real difficulties. For this reason, we describe next a new variational principle, introduced recently by Ponte Castañeda (1991a), which deals precisely with the problem of constitutive nonlinearity. This is accomplished by expressing the effective energy function of the *nonlinear* composite in terms of a variational statement involving the effective energy functions of the class of *linear* comparison composites. Thus, the new variational principle allows the extension of well-known results for linear composites to corresponding results for nonlinear ones. In this paper, we will make use of this variational principle, and of well-known results for the effective properties of linearly elastic laminates, to determine the effective constitutive behavior of ductile laminates. Before proceeding with this task, we briefly review the new variational principle.

The new variational principle for the effective energy of the composite $\tilde{U}$ is obtained by means of the Legendre transformation, applied to a modified set of variables. We will assume that the heterogeneous solid is locally *isotropic*, such that

$$U(\mathbf{x}, \sigma) = \psi(\mathbf{x}; \tau_e, \sigma_m), \quad (6)$$

where ψ is a non-negative function, satisfying the condition that $\psi(\mathbf{x}; 0, 0) = 0$ for all $\mathbf{x}$. Additionally, ψ is convex in the variables [cf. (A2)]

$$\sigma_m = \frac{1}{3} \text{tr } \sigma \quad \text{and} \quad \tau_e = \sqrt{\frac{1}{2} \sigma' \cdot \sigma'}, \quad (7)$$

denoting the *mean* and *effective* (in their plasticity usages) stresses, respectively, where $\sigma' = \sigma - \sigma_m \mathbf{I}$ is the stress deviator tensor. We note that form (6) is *not* the most general form for the energy function of a nonlinear isotropic solid (we could also have dependence on the determinant of σ), but this form is still general enough to cover the usual plasticity models of interest here. Further, we will assume that the growth in U as the magnitude of the stress tensor becomes large is stronger than quadratic. This is of course consistent with the ductile behavior of the material.

The new variational principle is obtained then in terms of the following expression for the energy-density function of the heterogeneous solid, namely,

$$U(\mathbf{x}, \sigma) = \max_{\mu_0, \kappa_0 \geq 0} \{U_0(\mathbf{x}, \sigma) - V(\mathbf{x}; \mu_0, \kappa_0)\}, \quad (8)$$

where U_0 is the energy-density function of a linearly elastic comparison solid with shear modulus μ_0 , and bulk modulus κ_0 , such that

$$U_0(\mathbf{x}, \sigma) = \frac{1}{2\mu_0} \tau_e^2 + \frac{1}{2\kappa_0} \sigma_m^2, \quad (9)$$

and where

$$V(\mathbf{x}; \mu_0, \kappa_0) = \max_{\sigma} \{U_0(\mathbf{x}, \sigma) - U(\mathbf{x}, \sigma)\}. \quad (10)$$

(Note that the maximum in the above function is usually bounded, because of the stronger than quadratic assumption on U .) These expressions are obtained by means of the changes of variables, $v_e = \tau_e^2$ and $v_m = \sigma_m^2$, which lead to the definition of a nonnegative function f , such that

$$f(\mathbf{x}; v_e, v_m) = \psi(\mathbf{x}; \tau_e, \sigma_m). \quad (11)$$

Then, expression (10) is nothing more than the Legendre dual of f ; in fact, we have that $V(\mathbf{x}; \mu_0, \kappa_0) = f^*(\mathbf{x}; p_e, p_m)$, with $p_e = 1/(2\mu_0)$ and $p_m = 1/(2\kappa_0)$. Here, f^* is the Legendre transform of f , given by

$$f^*(\mathbf{x}; p_e, p_m) = \max_{v_e, v_m \geq 0} \{p_e v_e + p_m v_m - f(\mathbf{x}; v_e, v_m)\}, \quad (12)$$

and (8) is a statement of Legendre duality for convex f (i.e. $f^{**} = f$, but written in terms of U and V). For details, we refer the reader to Ponte Castañeda (1992).

The new variational principle is then obtained essentially by inserting expression (8) for U into the principle of minimum complementary energy (4), and interchanging the order of the minimum over the set of admissible stresses with the maximum over the comparison moduli. The result may be expressed in the form (Ponte Castañeda, 1992)

$$\bar{U}(\bar{\sigma}) = \max_{\mu_0(\mathbf{x}), \kappa_0(\mathbf{x}) \geq 0} \left\{ \bar{U}_0(\bar{\sigma}) - \int_{\Omega} V(\mathbf{x}; \mu_0(\mathbf{x}), \kappa_0(\mathbf{x})) d\mathbf{x} \right\}, \quad (13)$$

where

$$\bar{U}_0(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} \int_{\Omega} U_0(\mathbf{x}, \sigma) d\mathbf{x} \quad (14)$$

is the effective energy of the linear comparison composite. We emphasize that expression

(13) is a variational principle in its own right since it involves an infinite-dimensional optimization over the set of nonnegative functions $\mu_0(\mathbf{x})$ and $\kappa_0(\mathbf{x})$. Thus, even if we had an explicit expression for the effective energy function of the linear comparison composite $\tilde{U}_0$ [not an easy calculation in general for arbitrary $\mu_0(\mathbf{x})$ and $\kappa_0(\mathbf{x})$], the above variational principle would still be difficult to implement. However, we will see that for a laminated composite, the above problem simplifies dramatically. Similarly, it was shown by Ponte Castañeda (1991a, 1992) that the above variational principle can also be utilized in an approximate fashion to compute bounds for the effective properties of nonlinear composites with more general microstructures. Additionally, in the same references, dual versions of (13) are also given in terms of the minimum potential energy of the composite; however, in this paper we prefer to use the above formulation due to the fact that it is easier to express the stress/strain relation for a ductile material in terms of the complementary energy-density function U than in terms of its Legendre counterpart, the energy-density function $W = U^*$.

3. APPLICATION OF THE NEW VARIATIONAL PRINCIPLE TO A NONLINEAR LAMINATED COMPOSITE

In this section, we specialize the general formulation of the previous section to the case of laminated composites. Such materials consist of n homogeneous, isotropic phases occupying nonintersecting layered regions $\Omega^{(r)}$ ($r = 1, 2, \dots, n$), with union Ω and with normal $\mathbf{n}$. The complementary energy-density function for the laminated material is then expressible in the form

$$U(\boldsymbol{\sigma}, \mathbf{x}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x} \cdot \mathbf{n}) U^{(r)}(\boldsymbol{\sigma}), \quad (15)$$

where $\chi^{(r)}(\mathbf{x} \cdot \mathbf{n})$ (equal to 1 for $\mathbf{x}$ in phase r , and 0 otherwise) is the characteristic function of phase r , and $U^{(r)}(\boldsymbol{\sigma}) = \psi^{(r)}(\tau_e, \sigma_m)$ is the corresponding homogeneous, isotropic energy-density function. Also the volume fraction $c^{(r)}$ of each phase is determined by the corresponding characteristic functions $\chi^{(r)}$ via the relation

$$c^{(r)} = \int_{\Omega} \chi^{(r)}(\mathbf{x} \cdot \mathbf{n}) d\mathbf{x}. \quad (16)$$

We remark that a laminated composite with *perfectly bonded*, isotropic phases possesses *transversely isotropic* symmetry (with transverse direction $\mathbf{n}$). In some sense, it represents the simplest composite material with transverse isotropy; other examples of practical importance include fiber-reinforced materials with isotropic constituent phases. These will be considered elsewhere. Because of the particular type of anisotropy involved in laminated composites, we have included in Appendix A a brief summary, largely after Walpole (1981), of the appropriate invariants and other useful definitions for transversely isotropic materials.

The computation of the effective energy-density function of a laminated composite is made easy by the following property of laminated composites. If the thickness of the typical layer is small compared to the size of the laminate (i.e. if the laminate—linear or nonlinear—is a *composite* in the sense defined in Section 2), then, away from a boundary-layer region close to the boundary of the composite, the fields are constant within each layer (a different constant in each layer). Therefore, the problem of determining the effective energy function of a laminated composite reduces to that of determining the constant fields within each phase of the composite by imposition of the appropriate jump conditions (continuity of traction stresses and tangential strains) across the interfaces between the different layers, as well as the averaging conditions stated in Section 2. Thus, the problem of determining the effective energy function of a laminated composite, unlike the corresponding problem for a general composite, simplifies to an algebraic one. Although, in principle, the resulting problem can always be solved; in practice, it may be difficult to obtain explicit results

because the jump conditions take the form of complicated sets of nonlinear algebraic equations. However, if the composite is made up of linear phases (with quadratic energy functions in each phase), the jump conditions are also linear and they can be solved in closed form, as discussed in Section 1.

The results for the effective energy functions of linear laminated composites are given in the next two sections; in the balance of this section, we make use of the new variational principle (13) to determine an *exact* expression for the effective energy function of a nonlinear laminated composite $\tilde{U}$, written in terms of the effective energy functions $\tilde{U}_0$ of the class of linearly elastic comparison laminates. This is accomplished by noting that, for a laminate, the minimizing comparison moduli functions $\mu_0(\mathbf{x})$ and $\kappa_0(\mathbf{x})$ in (13) must be constant within each phase. Therefore, it suffices to optimize with respect to the set of *constant* [over each phase r ($r = 1, \dots, n$)] comparison moduli, $\mu_0^{(r)}$ and $\kappa_0^{(r)}$. Thus, we have that

$$\tilde{U}(\bar{\sigma}) = \max_{\mu_0^{(r)}, \kappa_0^{(r)} > 0} \left\{ \tilde{U}_0(\bar{\sigma}) - \sum_{s=1}^n c^{(s)} V^{(s)}(\mu_0^{(s)}, \kappa_0^{(s)}) \right\}, \quad (17)$$

where, from (10),

$$V^{(r)}(\mu_0^{(r)}, \kappa_0^{(r)}) = \max_{\tau_c, \sigma_m} \left\{ \frac{1}{2\mu_0^{(r)}} \tau_c^2 + \frac{1}{2\kappa_0^{(r)}} \sigma_m^2 - \psi^{(r)}(\tau_c, \sigma_m) \right\}, \quad (18)$$

and where $\tilde{U}_0$ is the effective energy-density function of a linearly elastic laminated material made up of n phases in volume fractions $c^{(r)}$, with shear and bulk moduli, $\mu_0^{(r)}$ and $\kappa_0^{(r)}$, respectively.

On the face of it, expressions (17) with (18) for the effective energy function of a nonlinear laminated composite do not appear to offer much of an analytical advantage over the standard procedure of determining the stress fields within each phase (by solving the appropriate nonlinear jump conditions) and putting them directly in the complementary energy principle (4). This is due to the large number of optimizations involved in expressions (17) and (18) (i.e. a total of $4n$ optimizations for an n -phase laminate). However, we shall see in the next two sections that application of the particular form for the effective energy function of a linearly elastic laminate in (17) leads to a simpler optimization problem for the effective energy function of the nonlinear laminate. Further, we observe that, from a computational point of view, it is generally easier to minimize (or maximize) functions than it is to solve nonlinear sets of equations, and therefore, the methods developed in this paper are computationally superior to the standard procedure of solving systems of nonlinear equations (arising from the jump conditions). In Section 4, we begin by considering the simpler case of a laminated composite with incompressible, isotropic phases, and in Section 5, we tackle the more complicated problem of a general laminated composite with compressible, isotropic phases.

4. THE INCOMPRESSIBLE LAMINATED COMPOSITE

In this section, we deal with the special case of laminated composites with *incompressible*, isotropic phases. In this case, the energy-density functions of each phase take the simpler form $U^{(r)}(\sigma) = \psi^{(r)}(\tau_c)$. Then, relations (17) and (18), expressing the effective energy function $\tilde{U}$ of the nonlinear laminate, reduce to

$$\tilde{U}(\bar{\sigma}) = \max_{\mu_0^{(r)} > 0} \left\{ \tilde{U}_0(\bar{\sigma}) - \sum_{s=1}^n c^{(s)} V^{(s)}(\mu_0^{(s)}) \right\}, \quad \text{and} \quad V^{(r)}(\mu_0^{(r)}) = \max_{\tau_c} \left\{ \frac{1}{2\mu_0^{(r)}} \tau_c^2 - \psi^{(r)}(\tau_c) \right\}, \quad (19, 20)$$

where $\tilde{U}_0$ now refers to the effective energy-density function of a linearly elastic laminated

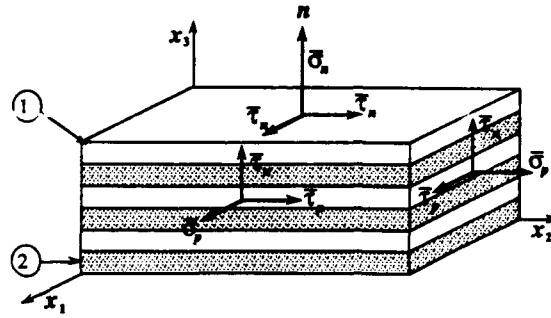


Fig. 1. A two-phase laminated material.

material composed of n incompressible phases with shear moduli $\mu_0^{(r)}$ in prescribed volume fractions $c^{(r)}$.

The effective energy-density function of the linear (incompressible) comparison laminate $\tilde{U}_0$ may be computed from the general results of Walpole (1969), specialized to the case of incompressible, isotropic phases. We obtain

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\bar{\mu}_0} (\bar{\tau}_p^2 + \bar{\tau}_d^2) + \left(\frac{1}{2\bar{\mu}_0} \right) \bar{\tau}_n^2, \quad (21)$$

where the overbars on the moduli denote volume averages (e.g. $\bar{\mu}_0 = \sum_{i=1}^n c^{(i)} \mu_0^{(i)}$), and where $\bar{\tau}_n$, $\bar{\tau}_p$ and $\bar{\tau}_d$ are the three transversely isotropic invariants of the applied stress tensor $\bar{\sigma}$ (which is trace-free) corresponding to the three independent modes for an incompressible, transversely isotropic, linear material (see Appendix A and Fig. 1). They are the transverse shear stress [(A5)₃], the longitudinal shear stress [(A5)₄] and the deviatoric stress [(A5)_{1,2} and (A12)], respectively. We note that the three independent modes for a general incompressible, transversely isotropic material reduce to two independent modes for an incompressible laminated composite (since the transverse and deviatoric modes have the same effective response). Note further that, because of the identity $\bar{\tau}_c^2 = \bar{\tau}_p^2 + \bar{\tau}_d^2 + \bar{\tau}_n^2$ from the section on incompressible materials in Appendix A [(A6)₂], we are able to rewrite the first term in brackets in (21) in the form $(\bar{\tau}_c^2 - \bar{\tau}_n^2)$.

With expression (21) for $\tilde{U}_0$, we can now return to the computation of $\tilde{U}$, implied by (19). In this connection, we find that the following identity, proved in Appendix B, is useful in reducing the number of optimizations, namely,

$$\frac{1}{\bar{\mu}_0} = \min_{\omega^{(r)}, \bar{\omega}=0} \left\{ \sum_{i=1}^n \frac{c^{(i)}}{\mu_0^{(i)}} (1 - \omega^{(i)})^2 \right\}, \quad (22)$$

where the (constant) optimization variables $\omega^{(r)}$ ($r = 1, \dots, n$) are required to satisfy the constraint $\bar{\omega} = 0$. Then, substituting (21), together with (22), into (19) leads to the result

$$\tilde{U}(\bar{\sigma}) = \max_{\mu_0^{(r)} > 0} \min_{\omega^{(r)}, \bar{\omega}=0} \left\{ \sum_{i=1}^n c^{(i)} \left[\frac{1}{2\mu_0^{(i)}} (\tau^{(i)})^2 - V^{(i)}(\mu_0^{(i)}) \right] \right\}, \quad (23)$$

where

$$\tau^{(i)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_d^2)(1 - \omega^{(i)})^2 + \bar{\tau}_n^2}. \quad (24)$$

We note that, by definition, the functions $-V^{(r)}(\mu_0^{(r)}) = -(f^{(r)})^*(1/(2\mu_0^{(r)}))$ are concave in $1/(\mu_0^{(r)})$, and similarly the variables $\tau^{(r)}$ are convex in $\omega^{(r)}$. Therefore, by the Saddle Point Theorem (Rockafellar, 1970, Corollary 37.3.1), we are allowed to interchange the order of the maximum and the minimum in (23). Further, it follows from (8) [assuming convexity of $f^{(r)}$, where $f^{(r)}(v_c) = \psi^{(r)}(\tau_c)$ with $v_c = \tau_c^2$; see (11)] that

$$\psi^{(s)}(\tau) = \max_{\mu_0^{(s)} > 0} \left\{ \frac{1}{2\mu_0^{(s)}} (\tau)^2 - V^{(s)}(\mu_0^{(s)}) \right\}. \quad (25)$$

Therefore, we conclude from (23) that

$$\tilde{U}(\bar{\sigma}) = \min_{\omega^{(r)}, \bar{\omega}=0} \left\{ \sum_{s=1}^n c^{(s)} \psi^{(s)}(\tau^{(s)}) \right\}, \quad (26)$$

where the variables $\tau^{(s)}$ are given by relations (24). Evidently, this form is much simpler than the original form given by (19) and (20); it involves an n -dimensional constrained optimization in place of the $2n$ -dimensional optimization problem implied by the original form. However, the linear constraint $\bar{\omega} = 0$ for the n optimization variables $\omega^{(r)}$ ($r = 1, \dots, n$) can be embedded into the optimization problem (26) by letting the n th variable $\omega^{(n)}$ be expressed in terms of the other $n-1$ variables $\omega^{(s)}$ ($s = 1, \dots, n-1$) via

$$\omega^{(n)} = -\frac{1}{c^{(n)}} \sum_{s=1}^{n-1} c^{(s)} \omega^{(s)}. \quad (27)$$

With this modification, the problem (26) reduces to an $(n-1)$ -dimensional optimization problem over the unconstrained variables $\omega^{(s)}$ ($s = 1, \dots, n-1$). For instance, for the case of a two-phase laminated composite, the problem (26) reduces to the one-dimensional optimization problem

$$\tilde{U}(\bar{\sigma}) = \min_{\omega} \{ c^{(1)} \psi^{(1)}[\sqrt{(1-c^{(2)}\omega)^2(\bar{\tau}_e^2 - \bar{\tau}_n^2) + \bar{\tau}_n^2}] + c^{(2)} \psi^{(2)}[\sqrt{(1+c^{(1)}\omega)^2(\bar{\tau}_e^2 - \bar{\tau}_n^2) + \bar{\tau}_n^2}] \}, \quad (28)$$

which is expressed in terms of one (unconstrained) optimization variable ω . Here, we have made the following identifications, $\omega^{(1)} = c^{(2)}\omega$ and $\omega^{(2)} = -c^{(1)}\omega$.

Finally, we remark that simple expressions for the effective stress/strain relations of the nonlinear transversely isotropic laminated composite may be obtained by means of the results of Appendix C. These relations may be written in terms of the incompressible, transversely isotropic invariants of the average strain tensor $\bar{\epsilon}$, namely, the transverse shear strain $\bar{\gamma}_p$, the deviatoric shear strain $\bar{\gamma}_d$, and the longitudinal shear strain $\bar{\gamma}_n$. These strain invariants are defined in Appendix A, and are completely analogous to the corresponding (incompressible) transversely isotropic invariants of the average stress. Thus, with the help of relations (C7), we may write

$$\begin{aligned} \bar{\gamma}_p &= \left[\sum_{r=1}^n c^{(r)} (1 - \hat{\omega}^{(r)})^2 \frac{1}{\hat{\tau}^{(r)}} \frac{d\psi^{(r)}}{d\tau^{(r)}}(\hat{\tau}^{(r)}) \right] \frac{\bar{\tau}_p}{2}, \\ \bar{\gamma}_d &= \left[\sum_{r=1}^n c^{(r)} (1 - \hat{\omega}^{(r)})^2 \frac{1}{\hat{\tau}^{(r)}} \frac{d\psi^{(r)}}{d\tau^{(r)}}(\hat{\tau}^{(r)}) \right] \frac{\bar{\tau}_d}{2}, \\ \bar{\gamma}_n &= \left[\sum_{r=1}^n c^{(r)} \frac{1}{\hat{\tau}^{(r)}} \frac{d\psi^{(r)}}{d\tau^{(r)}}(\hat{\tau}^{(r)}) \right] \frac{\bar{\tau}_n}{2}, \end{aligned} \quad (29)$$

where $\hat{\tau}^{(r)} = \tau^{(r)}(\hat{\omega}^{(r)})$, and where the variables $\hat{\omega}^{(r)}$ are the optimized values of the $\omega^{(r)}$ from (26). We note that for the nonlinear laminated composite, there is full coupling between all the distortional (shear) modes. This is different from the situation for the corresponding linear laminated composite [see (21)], where all three modes are uncoupled. As we will see in the ensuing discussions, this inter-mode coupling is one of the intrinsic features of laminated (and other anisotropic) nonlinear composites.

5. THE COMPRESSIBLE LAMINATED COMPOSITE

With the insight gained in the previous section, we attempt in the present section to obtain corresponding results for n -phase laminated composites with nonlinear, isotropic, compressible phases. In this case, we can apply the results (17) and (18) from Section 3 directly; we only require an expression for the effective energy function $\tilde{U}_0$ of the linearly elastic laminate with isotropic, compressible phases in prescribed volume fractions. This energy function may be computed directly from the results of Walpole (1969) for the transversely isotropic moduli of linearly elastic laminated composites. The final result may be written in the form

$$\tilde{U}_0(\bar{\sigma}) = \tilde{U}_1(\bar{\sigma}) + \tilde{U}_2(\bar{\sigma}), \quad \text{where} \quad \tilde{U}_1(\bar{\sigma}) = \frac{1}{2\bar{\mu}_0} \bar{\tau}_p^2 + \overline{\left(\frac{1}{2\mu_0}\right)} \bar{\tau}_n^2, \quad (30, 31)$$

$$\tilde{U}_2(\bar{\sigma}) = \frac{1}{2\bar{\eta}_0} \bar{\sigma}_p^2 - \frac{1}{\bar{\eta}_0} \overline{\left(\frac{3\kappa_0 - 2\mu_0}{3\kappa_0 + 4\mu_0}\right)} \bar{\sigma}_p \bar{\sigma}_n + \frac{1}{2} \left[\overline{\left(\frac{3}{3\kappa_0 + 4\mu_0}\right)} + \frac{1}{\bar{\eta}_0} \overline{\left(\frac{3\kappa_0 - 2\mu_0}{3\kappa_0 + 4\mu_0}\right)}^2 \right] \bar{\sigma}_n^2, \quad (32)$$

with $\eta_0 = 9\kappa_0\mu_0/(3\kappa_0 + 4\mu_0)$, and where $\bar{\sigma}_p$, $\bar{\sigma}_n$, $\bar{\tau}_p$ and $\bar{\tau}_n$ are the four transversely isotropic invariants (up to quadratic in order) of the applied stress $\bar{\sigma}$. They denote, respectively, the in-plane hydrostatic stress, the normal tensile stress, the transverse shear stress and the longitudinal shear stress (see Appendix A and Fig. 1). The reason behind the above splitting of $\tilde{U}_0$ lies in the similarity between the first part of (30), as given by (31), for the distortional (shear) modes of the compressible laminate and relation (21) for the incompressible composite (with $\bar{\tau}_p^2 + \bar{\tau}_d^2$ replaced by $\bar{\tau}_p^2$). Thus, it follows immediately that

$$\tilde{U}_1(\bar{\sigma}) = \min_{\omega_e^{(s)}, \bar{\omega}_e = 0} \left\{ \sum_{s=1}^n \frac{c^{(s)}}{2\mu_0^{(s)}} [(1 - \omega_e^{(s)})^2 \bar{\tau}_p^2 + \bar{\tau}_n^2] \right\}, \quad (33)$$

where the $\omega_e^{(s)}$ are the corresponding optimization variables, and they are subject to the constraint $\bar{\omega}_e = 0$. The second part is more complicated, but it can be shown by straightforward computation that, if $\bar{\sigma}_p \neq 0$, $\tilde{U}_2(\bar{\sigma})$ may be represented in the form

$$\tilde{U}_2(\bar{\sigma}) = \min_{\omega_m^{(s)}, \bar{\omega}_m = 0} \left\{ \sum_{s=1}^n \frac{c^{(s)}}{6\mu_0^{(s)}} [\bar{\sigma}_n - (1 - \omega_m^{(s)}) \bar{\sigma}_p]^2 + \sum_{s=1}^n \frac{c^{(s)}}{2\kappa_0^{(s)}} [\frac{1}{3} \bar{\sigma}_n + \frac{2}{3} (1 - \omega_m^{(s)}) \bar{\sigma}_p]^2 \right\}, \quad (34)$$

where the optimization variables $\omega_m^{(s)}$ are also subject to the constraint $\bar{\omega}_m = 0$.

By putting together relations (33) and (34), we arrive at the following expression for the linear comparison laminate

$$\tilde{U}_0(\bar{\sigma}) = \min_{\substack{\omega_e^{(s)}, \bar{\omega}_e = 0 \\ \omega_m^{(s)}, \bar{\omega}_m = 0}} \left\{ \sum_{s=1}^n \frac{c^{(s)}}{2\mu_0^{(s)}} (\tau_e^{(s)})^2 + \sum_{s=1}^n \frac{c^{(s)}}{2\kappa_0^{(s)}} (\sigma_m^{(s)})^2 \right\}, \quad (35)$$

where

$$\tau_e^{(s)} = \sqrt{(1 - \omega_e^{(s)})^2 \bar{\tau}_p^2 + \bar{\tau}_n^2 + \frac{1}{3} [\bar{\sigma}_n - (1 - \omega_m^{(s)}) \bar{\sigma}_p]^2}, \quad \text{and} \quad \sigma_m^{(s)} = \frac{1}{3} \bar{\sigma}_n + \frac{2}{3} (1 - \omega_m^{(s)}) \bar{\sigma}_p. \quad (36)$$

We note that this result is reminiscent of the type of result that one would expect to arise directly from the principle of minimum complementary energy. That this result is indeed directly obtainable from the principle of minimum complementary energy is demonstrated in Appendix D.

Then, following a procedure similar to the one followed in the development of expression (26) for the effective energy function of the nonlinear, incompressible laminated composite, but making use of (17) and (18), we arrive at the following expression for the effective energy function of the nonlinear laminated composite

$$\tilde{U}(\bar{\sigma}) = \min_{\substack{\omega_e^{(r)}, \omega_m^{(r)} \\ \dot{\omega}_e = \dot{\omega}_m = 0}} \left\{ \sum_{s=1}^n c^{(s)} \psi^{(s)}(\tau_e^{(s)}, \sigma_m^{(s)}) \right\}, \quad (37)$$

where $\tau_e^{(s)}$ and $\sigma_m^{(s)}$ are given by (36). Here, we have made use of the Saddle Point theorem allowing the interchange in the order of the minimum over the $\omega_e^{(r)}, \omega_m^{(r)}$ variables with the maximum over the comparison moduli $\mu_0^{(r)}$ and $\kappa_0^{(r)}$. We note that form (37) for the effective energy function $\tilde{U}$ of the nonlinear laminated composite is a direct generalization of form (35) for the effective energy function $\tilde{U}_0$ of the linear comparison laminate. We also note that while the distortional and dilatational modes are not coupled in the linear laminated composite [although the dilatational modes are coupled among themselves, as (32) shows], all four modes are strongly coupled for the nonlinear laminated composite.

Having obtained the simple form (37) for the effective energy function of a nonlinear laminate by means of the new variational principle, it can be demonstrated that the same result may be obtained directly from the principle of minimum complementary energy. This alternative derivation of (37) is given in Appendix D. We note in this connection that while the derivation of Appendix D may be physically more appealing than the above derivation, in this paper we have chosen to emphasize the derivation based on the new variational principles for the following reasons. The derivation based on the principle of minimum complementary energy depends on the fact that the fields are constant within different phases in the laminated composite; however, for a more general microstructure, such as a fiber-reinforced composite, the fields are no longer constant within the phases, and the minimum complementary energy approach would not work. On the other hand, deBotton and Ponte Castañeda (1992) have made use of the new variational principles to obtain expressions analogous to expressions (36) and (37) for the effective energy functions of nonlinear fiber-reinforced composites. Thus, the approach based on the new variational principles is more general and that is the reason for emphasizing the new approach even in the simple case of a laminated composite, where the new approach is not strictly required. In Appendix D, we also show that an alternative form of (37) is possible, which is not subject to the $\bar{\sigma}_p \neq 0$ restriction, although we note that the above form is still valid in the limit as $\bar{\sigma}_p \rightarrow 0$ (it is just not valid in a pointwise sense at $\bar{\sigma}_p = 0$, because the optimizing variables $\omega_m^{(r)}$ become unbounded in that limit).

The new representation for the effective energy function of a nonlinear laminated composite $\tilde{U}$ can be seen to involve only a $2n$ -dimensional optimization problem with two linear constraints. This is major reduction in order compared with the original expressions (17) and (18) involving a $4n$ -dimensional optimization problem. However, as noted in the previous section, further reductions are possible [to a $2(n-1)$ -dimensional optimization problem] by embedding the linear constraints directly into the optimization problem (37). For example, for the case of a two-phase composite, we obtain a result involving only a two-dimensional optimization problem prescribed in terms of the variables ω_e, ω_m via

$$\tilde{U}(\bar{\sigma}) = \min_{\omega_e, \omega_m} \{c^{(1)} \psi^{(1)}(\tau_e^{(1)}, \sigma_m^{(1)}) + c^{(2)} \psi^{(2)}(\tau_e^{(2)}, \sigma_m^{(2)})\}, \quad (38)$$

where $\tau_e^{(1)}, \sigma_m^{(1)}$ and $\tau_e^{(2)}, \sigma_m^{(2)}$ are given by relations (36) with $\omega_e^{(1)} = c^{(2)}\omega_e$, $\omega_e^{(2)} = -c^{(1)}\omega_e$, $\omega_m^{(1)} = c^{(2)}\omega_m$ and $\omega_m^{(2)} = -c^{(1)}\omega_m$.

Finally, we remark that simple expressions for the effective stress/strain relations of the transversely isotropic laminated composite may be obtained by means of the results of Appendix C. These may be written in terms of the transversely isotropic invariants of the average strain tensor $\bar{\epsilon}$, the in-plane hydrostatic strain $\bar{\epsilon}_p$, the normal tensile strain $\bar{\epsilon}_n$, the transverse shear strain $\bar{\gamma}_p$, and the longitudinal shear strain $\bar{\gamma}_n$. These are defined in Appendix A, and are completely analogous to the corresponding transversely isotropic invariants of the average stress. Thus, with the help of relations (C8), we may write

$$\begin{aligned} \bar{\epsilon}_p &= \frac{1}{6} \sum_{r=1}^n c^{(r)} (1 - \dot{\omega}_m^{(r)}) \left[2 \frac{\partial \psi^{(r)}}{\partial \sigma_m^{(r)}} (\tau_e^{(r)}, \sigma_m^{(r)}) + [(1 - \dot{\omega}_m^{(r)}) \bar{\sigma}_p - \bar{\sigma}_n] \frac{1}{\tau_e^{(r)}} \frac{\partial \psi^{(r)}}{\partial \tau_e^{(r)}} (\tau_e^{(r)}, \sigma_m^{(r)}) \right], \\ \bar{\epsilon}_n &= \frac{1}{3} \sum_{r=1}^n c^{(r)} \left[\frac{\partial \psi^{(r)}}{\partial \sigma_m^{(r)}} (\tau_e^{(r)}, \sigma_m^{(r)}) - [(1 - \dot{\omega}_m^{(r)}) \bar{\sigma}_p - \bar{\sigma}_n] \frac{1}{\tau_e^{(r)}} \frac{\partial \psi^{(r)}}{\partial \tau_e^{(r)}} (\tau_e^{(r)}, \sigma_m^{(r)}) \right], \end{aligned}$$

$$\begin{aligned}\bar{\gamma}_p &= \left[\sum_{r=1}^n c^{(r)} (1 - \hat{\omega}_e^{(r)})^2 \frac{1}{\hat{\tau}_e^{(r)}} \frac{\partial \psi^{(r)}}{\partial \tau_e^{(r)}} (\hat{\tau}_e^{(r)}, \hat{\sigma}_m^{(r)}) \right] \frac{\bar{\tau}_p}{2}, \\ \bar{\gamma}_n &= \left[\sum_{r=1}^n c^{(r)} \frac{1}{\hat{\tau}_e^{(r)}} \frac{\partial \psi^{(r)}}{\partial \tau_e^{(r)}} (\hat{\tau}_e^{(r)}, \hat{\sigma}_m^{(r)}) \right] \frac{\bar{\tau}_n}{2},\end{aligned}\quad (39)$$

where $\hat{\tau}_e^{(r)} = \tau_e^{(r)}(\hat{\omega}_e^{(r)}, \hat{\omega}_m^{(r)})$, $\hat{\sigma}_m^{(r)} = \sigma_m^{(r)}(\hat{\omega}_e^{(r)}, \hat{\omega}_m^{(r)})$, and where $\hat{\omega}_e^{(r)}, \hat{\omega}_m^{(r)}$ are the optimized values of $\omega_e^{(r)}, \omega_m^{(r)}$ from (37). We note that in this form, the coupling between the distortional and dilatational modes in the nonlinear material become evident since $\bar{\gamma}_p, \bar{\gamma}_n$ depend on $\bar{\sigma}_n, \bar{\sigma}_p$, and, conversely, $\bar{\epsilon}_n, \bar{\epsilon}_p$ depend on $\bar{\tau}_n, \bar{\tau}_p$.

6. APPLICATION TO LAMINATED COMPOSITES WITH POWER-LAW CONSTITUTIVE BEHAVIOR

In this section, we specialize the results of Sections 4 and 5 for three classes of laminated composites. The first subsection deals with the case of an incompressible laminated material made up of layers of a phase with "linear plus power-hardening" constitutive behavior, reinforced with stiffer layers of a linear-elastic material. In the study of these incompressible laminates, we will emphasize the coupling between different distortional loading modes arising as a consequence of nonlinearity and anisotropy in the laminates. The second subsection is dedicated to the study of a compressible, aluminum/alumina laminate, and the understanding of the dilatational modes is emphasized in this case. The third subsection deals with an incompressible laminated composite made up of two rigid/perfectly plastic phases with different yield stresses; it is interesting to note that, in this special case, completely explicit results are obtained for the effective yield function of the laminate.

6.1. Incompressible laminated composites

In this subsection, we consider an incompressible, two-phase laminated composite characterized by the following constitutive laws for the two isotropic phases. Phase 1 is governed by "linear plus power-hardening" constitutive behavior described by the energy-density function

$$\psi^{(1)}(\tau_e) = \int_0^{\sqrt{3}\tau_e} F^{(1)}(s) ds, \quad \text{where} \quad F^{(1)}(s) = \varepsilon_0 \left\{ \frac{s}{\sigma_0} + \left[\left(\frac{s}{\sigma_0} \right)^n - \left(\frac{\sigma_y}{\sigma_0} \right)^n \right] H(s - \sigma_y) \right\}.$$

(40, 41)

Here H is the unit step function (equal to 0 when $s \leq \sigma_y$ and to 1 otherwise), and ε_0, σ_0 are strain, stress normalization factors such that $\sigma_0/\varepsilon_0 = 3\mu^{(1)}$, with $\mu^{(1)}$ denoting the shear modulus of phase 1. Then, the function $F^{(1)}$ represents the uniaxial stress/strain relation of phase 1 under simple tension loading conditions. Thus, the behavior of phase 1 is linear when the uniaxial stress is lower than the yield stress, σ_y , and is linear plus power-hardening for stresses larger than σ_y . The factor $\sqrt{3}$ in (40) is needed in order to fit the isotropic stress invariant τ_e to the uniaxial case. Phase 2 is linear and governed by the quadratic energy-density function

$$\psi^{(2)}(\tau_e) = \frac{1}{2\mu^{(2)}} \tau_e^2, \quad (42)$$

where $\mu^{(2)}$ is the shear modulus of the phase.

With the above constitutive behavior for the two phases (1 and 2), which are prescribed in volume fractions $(1 - c^{(2)})$ and $c^{(2)}$, respectively, the effective energy-density function of the incompressible laminated composite may be expressed in dimensionless form via the relation

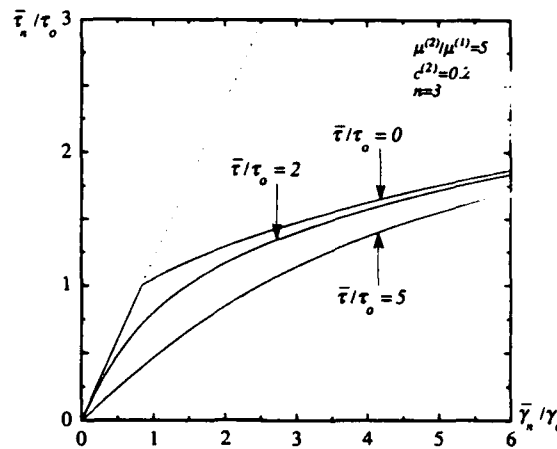


Fig. 2. The relations between the longitudinal shear stress $\bar{\tau}_n$ and strain $\bar{\gamma}_n$ of the incompressible, nonlinear laminate (continuous lines), and the reference linear laminate (short-dashed lines) for three different values of the other stress mode $\bar{\tau}$; $\bar{\tau}/\tau_0 = 0$, $\bar{\tau}/\tau_0 = 2$ and $\bar{\tau}/\tau_0 = 5$.

$$\frac{\bar{U}(\bar{\sigma})}{\tau_0 \gamma_0} = G \left\{ \frac{\bar{\tau}_p}{\tau_0}, \frac{\bar{\tau}_d}{\tau_0}, \frac{\bar{\tau}_n}{\tau_0}, \frac{\sigma_y}{\sigma_0}, \frac{\mu^{(2)}}{\mu^{(1)}}, n, c^{(2)} \right\}, \quad (43)$$

where the specific form of the function G is determined from (28), and $\tau_0 = \sqrt{\frac{1}{3}}\sigma_0$, $\gamma_0 = \sqrt{\frac{3}{4}}\epsilon_0$, so that $\tau_0/\gamma_0 = 2\mu^{(1)}$. Then, the relations between the three (incompressible) transversely isotropic stress invariants and the corresponding strain invariants may be computed from (29). These relations are presented in Figs 2–5 for the following values of the four parameters appearing in (43):

$$\frac{\sigma_y}{\sigma_0} = 1, \quad \frac{\mu^{(2)}}{\mu^{(1)}} = 5, \quad n = 3 \quad \text{and} \quad c^{(2)} = 0.2.$$

We recall that there are only two independent modes for the incompressible laminated composites; they are the longitudinal shear stress $\bar{\tau}_n$ and the following combination of the other two shear modes $\sqrt{\bar{\tau}_p^2 + \bar{\tau}_d^2}$ (i.e. the transverse and deviatoric shear modes, respectively). For simplicity, we will refer to this combination of the two modes at $\bar{\tau}$ and to the corresponding combination of the strain modes, $\sqrt{\bar{\gamma}_p^2 + \bar{\gamma}_d^2}$, as $\bar{\gamma}$. Thus, it suffices to consider the relations among the stress modes $\bar{\tau}_n$, $\bar{\tau}$ and the strain modes $\bar{\gamma}_n$, $\bar{\gamma}$ in order to have a complete description of the constitutive behavior of the incompressible laminate. In order to highlight the effect of nonlinearity, results are included in Figs 2–5 in the form of short-

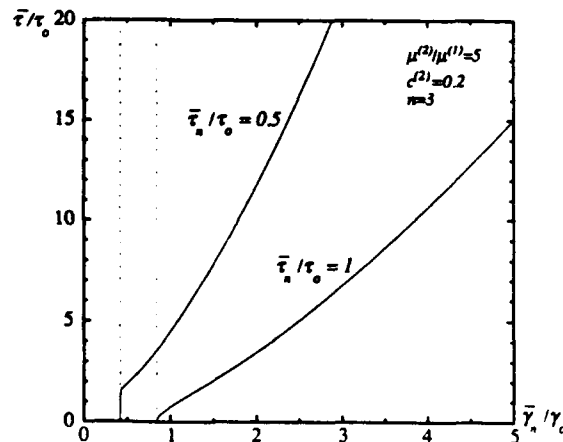


Fig. 3. The inter-relations between the shear stress $\bar{\tau}$ and the longitudinal shear strain $\bar{\gamma}_n$ of the incompressible, nonlinear laminate (continuous lines), and the reference linear laminate (short-dashed lines) for two different values of the longitudinal shear stress $\bar{\tau}_n$; $\bar{\tau}_n / \tau_0 = 0.5$ and $\bar{\tau}_n / \tau_0 = 1$.

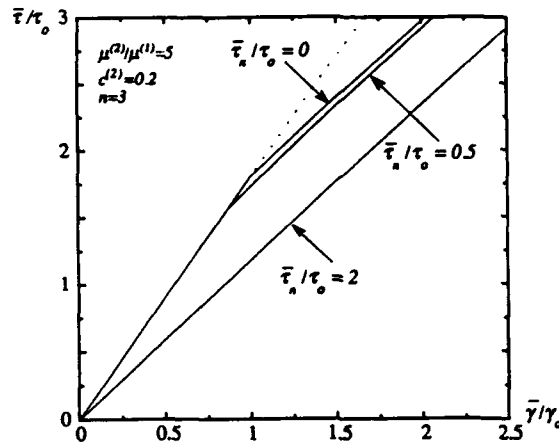


Fig. 4. The relations between the shear stress $\bar{\tau}$ and the corresponding shear strain $\bar{\gamma}$ of the incompressible, nonlinear laminate (continuous lines), and the reference linear laminate (short-dashed lines) for three different values of the longitudinal shear stress $\bar{\tau}_n$; $\bar{\tau}_n/\tau_0 = 0$, $\bar{\tau}_n/\tau_0 = 0.5$ and $\bar{\tau}_n/\tau_0 = 2$.

dashed curves for a linear laminated composite with the same shear moduli as the nonlinear laminate. Thus, the phases of this linear *reference* laminate are similar to those of the nonlinear one with the only difference that in phase I $\sigma_y = \infty$.

Figure 2 shows a plot of the longitudinal shear stress $\bar{\tau}_n$ versus the longitudinal shear strain $\bar{\gamma}_n$ for three different values of $\bar{\tau}$ ($\bar{\tau}/\tau_0 = 0, 2, 5$). We observe that when there is no preloading of the laminate ($\bar{\tau}/\tau_0 = 0$), the behavior of the stress/strain curve of the nonlinear laminate is initially the same as that of the reference linear laminate (short-dash line) until phase I yields. After yielding, the two curves diverge with the nonlinear phase controlling the behavior for large longitudinal shear stresses. That this should be so is seen from the fact that shear parallel to the layers should be controlled by the less stiff phase (in this case, the nonlinear phase). The effect of increasing $\bar{\tau}$ is to saturate the linear range of phase I, forcing the effective stress/strain curve of the laminate to be controlled by the nonlinear phase even for small values of the longitudinal shear stresses $\bar{\tau}_n$.

Figure 3 shows a plot of $\bar{\tau}$ versus the longitudinal shear strain $\bar{\gamma}_n$ for two different values of the longitudinal shear stress ($\bar{\tau}_n/\tau_0 = 0.5, 1$), and serves to emphasize the coupling between the two shear modes. Thus, a small preload in the form of a longitudinal shear stress $\bar{\tau}_n$ applied to the nonlinear laminate can lead to large increases in the longitudinal strain $\bar{\gamma}_n$ as the other shear stress mode $\bar{\tau}$ is increased; in fact, the growth is unbounded and can be shown to be proportional to $(\bar{\tau}/\tau_0)^{(n-1)/n}$.

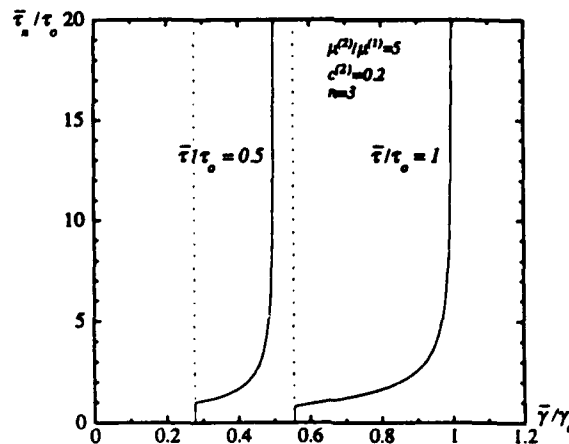


Fig. 5. The inter-relations between the longitudinal shear stress $\bar{\tau}_n$ and the shear strain $\bar{\gamma}$ of the incompressible, nonlinear laminate (continuous lines), and the reference linear laminate (short-dashed lines) for two different values of the stress mode $\bar{\tau}$; $\bar{\tau}/\tau_0 = 0.5$, and $\bar{\tau}/\tau_0 = 1$.

Figure 4 shows a plot of the shear stress $\bar{\tau}$ versus the shear strain $\bar{\gamma}$ for three different values of longitudinal shear stress $\bar{\tau}_n$ ($\bar{\tau}_n/\tau_0 = 0, 0.5, 1$). We observe that when there is no preloading of the laminate ($\bar{\tau}_n/\tau_0 = 0$), the behavior of the effective stress-strain curve of the nonlinear laminate is initially the same as that of the reference linear laminate (short-dash line) until phase 1 yields. After yielding, however, the two curves diverge with the linear phase controlling the behavior for large shear stresses $\bar{\tau}$. In fact, it can be demonstrated that the slope of the stress/strain curve in question reaches an asymptotic value of $2c^{(2)}\mu^{(2)}$ (corresponding to a linear Voigt estimate with $\mu^{(1)} \rightarrow 0$) as the shear stress $\bar{\tau}$ becomes large. Evidently, the weaker nonlinear phase is acting as if it was not present for large enough $\bar{\tau}$. The effect of increasing $\bar{\tau}_n$ is then to saturate the linear range of phase 1, reducing the effect of the nonlinear phase on the effective stress/strain curve of the composite (the laminate behaves almost linearly with modulus $c^{(2)}\mu^{(2)}$ for sufficiently large preload $\bar{\tau}_n$). That the nonlinear laminate should be controlled by the stiffer linear phase for large magnitudes of the transverse shear stress $\bar{\tau}_p$ (and fixed longitudinal shear stress $\bar{\tau}_n$) is easy to visualize, but that exactly the same behavior should be observed for the deviatoric mode $\bar{\tau}_d$ (the other component of $\bar{\tau}$) is perhaps less intuitive. The reason, however, is related to the Poisson effect. Thus, for example, if the laminate is compressed along the normal direction (which may seem to be controlled by the less stiff nonlinear phase), tensile strains are set up in the plane of the layers, which must be continuous across the phases, thus providing the required stiffening effect in the normal direction (because the linear phase controls the in-plane behavior of the laminate).

Figure 5 shows the relation between the longitudinal shear stress mode $\bar{\tau}_n$ and the strain mode $\bar{\gamma}$ for different values of the shear stress $\bar{\tau}$ ($\bar{\tau}/\tau_0 = 0.5, 2$). We observe that while there is significant coupling between the two modes (by comparison with the linear reference laminate), the coupling is not as significant as in Fig. 3. Thus, the shear strain $\bar{\gamma}$ reaches a maximum level for a given shear preload $\bar{\tau}$ as the longitudinal shear $\bar{\tau}_n$ is increased. This is because the nonlinear phase is dominated by the linear phase in this mode of deformation as observed previously in connection with Fig. 4. The effect of increasing preload $\bar{\tau}$ is to increase (in both absolute and relative terms) the increments in the shear strain $\bar{\gamma}$ with increasing shear stress $\bar{\tau}_n$.

6.2. The aluminum/alumina laminated composite

In this subsection, we demonstrate the behavior of a nonlinear, compressible laminated composite made up of aluminum layers reinforced with layers of alumina. Aluminum is a ductile material with uniaxial stress/strain curves that can be approximated by a "linear-plus-power" law with hardening exponent n varying between 4.2 and 5.8. Thus, we will assume the following form for the energy-density function of the aluminum layers (phase 1)

$$\psi^{(1)}(\tau_e, \sigma_m) = \int_0^{\sqrt{3}\tau_e} F^{(1)}(s) ds + \frac{1}{2\kappa^{(1)}} \sigma_m^2, \quad (44)$$

where $F^{(1)}$ is the same as in (41), and thus the only difference between (40) and (44) is the compressibility of aluminum accounted for in (44) through the bulk modulus $\kappa^{(1)}$. Alumina (phase 2) is a brittle material that behaves in a linear fashion up to the point of failure. Its energy-density function is represented by

$$\psi^{(2)}(\tau_e, \sigma_m) = \frac{1}{2\mu^{(2)}} \tau_e^2 + \frac{1}{2\kappa^{(2)}} \sigma_m^2, \quad (45)$$

where $\mu^{(2)}$ and $\kappa^{(2)}$ denote the shear and bulk moduli of the alumina, respectively.

With this choice of $\psi^{(1)}$ and $\psi^{(2)}$ (for the behaviors of the two phases), the effective energy-density function of the composite can be represented in dimensionless form via

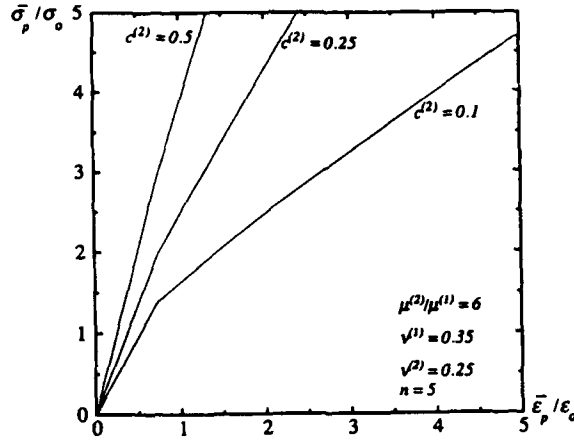


Fig. 6. The relations between the in-plane hydrostatic stress $\bar{\sigma}_p$ and strain $\bar{\epsilon}_p$ of the compressible laminate (where no other stress modes are present) for three different values of the volume fraction of the linear phase, $c^{(2)}$.

$$\frac{\bar{U}(\bar{\sigma})}{\tau_0 \gamma_0} = G \left\{ \frac{\bar{\sigma}_p}{\sigma_0}, \frac{\bar{\sigma}_n}{\sigma_0}, \frac{\bar{\tau}_p}{\tau_0}, \frac{\bar{\tau}_n}{\tau_0}, \frac{\sigma_y}{\sigma_0}, \frac{\mu^{(2)}}{\mu^{(1)}}, \nu^{(1)}, \nu^{(2)}, n, c^{(2)} \right\}, \quad (46)$$

where G is obtained from (38), $\tau_0 = \sqrt{3}\sigma_0$, $\gamma_0 = \sqrt{2}\epsilon_0$, such that $\tau_0/\gamma_0 = 2\mu^{(1)}$, and $\nu^{(1)}$, $\nu^{(2)}$ are the (dimensionless) Poisson's ratios of the two phases defined by

$$\nu^{(r)} = \frac{3\kappa^{(r)} - 2\mu^{(r)}}{6\kappa^{(r)} + 2\mu^{(r)}}.$$

In the results to follow, we will make the following choices (which are representative of the aluminum/alumina composite) for the material parameters in (46):

$$\frac{\sigma_y}{\sigma_0} = 1, \quad \frac{\mu^{(2)}}{\mu^{(1)}} = 6, \quad \nu^{(1)} = 0.35, \quad \nu^{(2)} = 0.25 \text{ and } n = 5.$$

The results are presented in Figs 6-9 in terms of plots of the four transversely isotropic stress modes versus the corresponding strain modes for three different values of the volume fraction of alumina $c^{(2)}$ (0.1, 0.25 and 0.5).

Figure 6 shows a plot of the in-plane hydrostatic stress $\bar{\sigma}_p$ versus the corresponding hydrostatic strain $\bar{\epsilon}_p$, when all other stress modes vanish, for the three values of the volume

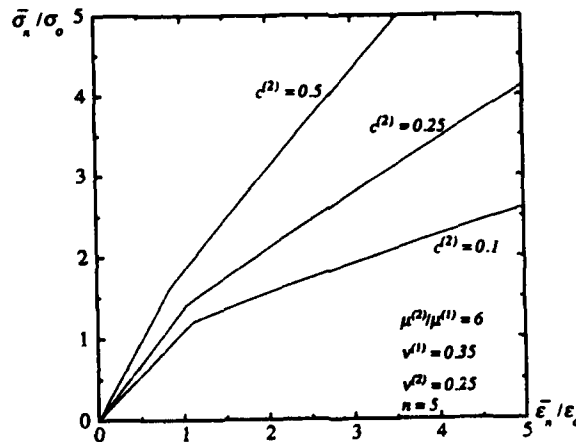


Fig. 7. The relation between the normal tensile stress $\bar{\sigma}_n$ and strain $\bar{\epsilon}_n$ of the compressible laminate (where no other stress modes are present) for three different values of the volume fraction of the linear phase, $c^{(2)}$.

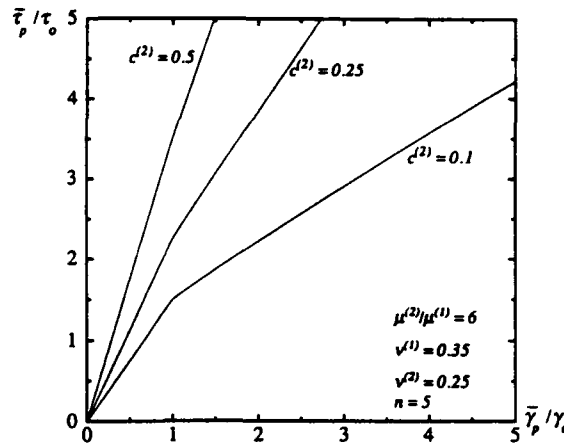


Fig. 8. The relations between the transverse shear stress $\bar{\tau}_p$ and strain $\bar{\gamma}_p$ of the compressible laminate (where no other stress modes are present) for three different values of the volume fraction of the linear phase, $c^{(2)}$.

fraction of alumina $c^{(2)}$. We observe that the laminate has a linear range with effective modulus $2\bar{\eta}$ (recall that $\eta = 9\kappa\mu/(3\kappa + 4\mu)$) up to yielding of the aluminum phase. However, the laminate behaves almost linearly even after yielding with modulus approaching $2c^{(2)}\eta^{(2)}$ for large values of $\bar{\sigma}_p$. This behavior is expected on physical grounds due to the fact that the stiffer material (alumina) should dominate the behavior in tension (compression) parallel to the layers. The effect of increasing volume fractions of alumina is of course to stiffen the effective behavior of the composite.

Figure 7 shows a plot of the normal tensile stress $\bar{\sigma}_n$ versus the corresponding tensile strain $\bar{\epsilon}_n$, when all other stress modes vanish, for the three previous values of the volume fraction of alumina $c^{(2)}$. The structure of the plots is very similar to that of Fig. 6; however, the effective moduli are different. Before phase 1 reaches yielding, the laminate has uniaxial modulus given by the expression

$$\left[\left(\frac{3}{3\kappa + 4\mu} \right) + \frac{1}{\bar{\eta}} \left(\frac{3\kappa - 2\mu}{3\kappa + 4\mu} \right)^2 \right]^{-1},$$

while after the yielding of phase 1, the modulus for large stress $\bar{\sigma}_n$ is reduced to the level

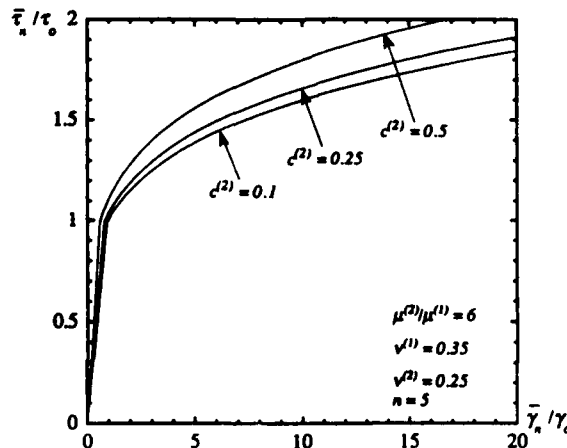


Fig. 9. The relations between the longitudinal shear stress $\bar{\tau}_n$ and strain $\bar{\gamma}_n$ of the compressible laminate (where no other stress modes are present) for three different values of the volume fraction of the linear phase, $c^{(2)}$.

$$\left[\frac{1}{c^{(2)}\eta^{(2)}} + \left(\frac{1}{\kappa} \right) - \frac{4}{3\kappa^{(2)}} \right]^{-1}.$$

In this case, it is not evident that the linear phase should govern the effective behavior of the laminate for large stresses. The reason, however, is the same as discussed in the previous subsection in connection with Fig. 4: continuity of the tangential strains across the interfaces together with the Poisson effect.

Figure 8 shows the corresponding plots for the transverse shear stress $\bar{\tau}_p$ versus the transverse shear strain $\bar{\gamma}_p$, when no other stress modes are present, for the three values of the volume fraction of alumina. In this case, the results are similar to those of Fig. 6 for clear physical reasons: the stiffer phase controls the behavior of the laminate under transverse shear loading.

Figure 9 shows plots of the longitudinal shear stress $\bar{\tau}_n$ versus the corresponding strain mode $\bar{\gamma}_n$, with no other stress modes present. The behavior in this case is dramatically different, as the study of the corresponding case for the incompressible laminate demonstrated earlier (Fig. 2). Thus, after an initial linear range before yielding of phase 1, the weaker nonlinear phase governs the effective behavior of the laminate. In contrast to the other three modes, we observe that the dependence on the volume fraction of alumina is fairly weak, so that the three curves (corresponding to different values of $c^{(2)}$) are quite close to each other.

Clearly, a study of the inter-relations between the different modes would be required to have a complete picture of the effective behavior of the nonlinear compressible laminate. However, the behavior of these inter-modal relations is similar to those already explored for the incompressible laminate. Thus, the inter-modes relations that involve the longitudinal shear strain $\bar{\gamma}_n$ are of the form of the relations presented in Fig. 3 while all other inter-modal stress/strain relations are in the form of Fig. 5.

6.3. The rigid/perfectly plastic laminated composite

In this subsection, we consider the case of an incompressible laminated composite made up of two rigid/perfectly plastic phases with yield stresses $\tau_0^{(1)}$ and $\tau_0^{(2)}$, chosen such that $\tau_0^{(1)} < \tau_0^{(2)}$, in given volume fractions $c^{(1)}$ and $c^{(2)}$. The behavior of the phases may then be characterized in terms of the convex energy-density functions

$$\psi^{(r)}(\tau_e) = \begin{cases} 0, & \tau_e \leq \tau_0^{(r)}, \\ \infty, & \tau_e > \tau_0^{(r)}, \end{cases} \quad (47)$$

($r = 1, 2$), where τ_e denotes the effective shear stress. These energy functions may be obtained directly from pure power-law energy functions of the form

$$\psi^{(r)}(\tau_e) = \frac{1}{n+1} \varepsilon_0 \sigma_0^{(r)} \left(\frac{\tau_e}{\tau_0^{(r)}} \right)^{n+1}, \quad (48)$$

in the limit as $n \rightarrow \infty$. Further, these energy functions define "yield functions" for the phase materials that may be described in the usual way via

$$\phi^{(r)}(\sigma) = \tau_e - \tau_0^{(r)} = 0. \quad (49)$$

Here, we will proceed *formally* and make use of expression (28) to determine an expression for the effective energy function of the laminated material $\bar{U}$, from which we will be able to determine a yield function for the laminated composite Φ . For a rigorous treatment of homogenization theory for rigid/perfectly plastic composites, and in particular for a discussion concerning the validity of the normality condition for the effective yield

function of the composite, the reader is referred to Suquet (1985). Because of incompressibility and transverse isotropy, we will find that such a yield function may be represented as a curve in the $(\bar{\tau}_n, \bar{\tau})$ -space, where $\bar{\tau} = \sqrt{\bar{\tau}_p^2 + \bar{\tau}_d^2}$. Thus, application of (28) leads to the expression

$$\bar{U}(\bar{\sigma}) = \min_{\omega} \{G(\bar{\tau}_n, \bar{\tau}, \omega)\}, \quad \text{where} \quad G = \begin{cases} 0, & \tau^{(1)} \leq \tau_0^{(1)} \text{ and } \tau^{(2)} \leq \tau_0^{(2)} \\ \infty, & \tau^{(1)} > \tau_0^{(1)} \text{ or } \tau^{(2)} > \tau_0^{(2)} \end{cases} \quad (50, 51)$$

and where $\tau^{(1)} = \sqrt{(1 - c^{(2)}\omega)^2 \bar{\tau}^2 + \bar{\tau}_n^2}$ and $\tau^{(2)} = \sqrt{(1 + c^{(1)}\omega)^2 \bar{\tau}^2 + \bar{\tau}_n^2}$.

The above optimization problem for ω then reduces to determining all possible combinations of $\bar{\tau}$ and $\bar{\tau}_n$ for which $\bar{U} = 0$, which in turn defines the yield function for the composite Φ . First, we note that, independent of $\bar{\tau}$ and ω , $\bar{U}$ can only vanish if

$$\bar{\tau}_n - \tau_0^{(1)} \leq 0, \quad (52)$$

for otherwise $\tau^{(1)} \geq \bar{\tau}_n > \tau_0^{(1)}$. Thus, inequality (52) is a *necessary* condition for $\bar{U}$ to vanish. However, the condition (52) is not *sufficient* to ensure that $\bar{U}$ vanishes since the condition $\tau^{(2)} \leq \tau_0^{(2)}$ may be violated. Thus, assuming that condition (52) is satisfied, we ask the question of whether there are values of ω , depending on $\bar{\tau}$ and $\bar{\tau}_n$, such that conditions $\tau^{(1)} \leq \tau_0^{(1)}$ and $\tau^{(2)} \leq \tau_0^{(2)}$ are satisfied simultaneously. The answer is affirmative, provided that $\bar{\tau}$ and $\bar{\tau}_n$ (for given volume fractions $c^{(1)}$ and $c^{(2)}$) satisfy the condition

$$\bar{\tau} \leq c^{(1)} \sqrt{(\tau_0^{(1)})^2 - \bar{\tau}_n^2} + c^{(2)} \sqrt{(\tau_0^{(2)})^2 - \bar{\tau}_n^2}. \quad (53)$$

Thus, conditions (52) and (53) define an effective yield function for the composite, $\Phi = 0$, such that

$$\Phi(\bar{\sigma}) \equiv \begin{cases} \bar{\tau} - [c^{(1)} \sqrt{(\tau_0^{(1)})^2 - \bar{\tau}_n^2} + c^{(2)} \sqrt{(\tau_0^{(2)})^2 - \bar{\tau}_n^2}], & \bar{\tau}_n < \tau_0^{(1)} \\ \bar{\tau}_n - \tau_0^{(1)}, & \bar{\tau}_n = \tau_0^{(1)}. \end{cases} \quad (54)$$

We note that when $\tau_0^{(2)} = \tau_0^{(1)}$, the expression above reduces to the von Mises yield criterion.

Plots of the yield surfaces in the $(\bar{\tau}_n, \bar{\tau})$ -space of applied stresses are given in Figs 10 and 11. Figure 10 shows the exact yield surface Φ for the choice of parameters, $\tau_0^{(2)}/\tau_0^{(1)} = 2$ and $c^{(2)} = 0.5$. The isotropic Reuss and Voigt (also known as Bishop-Hill estimate) bounds for the yield surfaces are also given for comparison. We note that the exact yield surface Φ is close to the Voigt upper bounding surface Φ_v for low values of the longitudinal shear stress ($\bar{\tau}_n < \frac{1}{2}\tau_0^{(1)}$), and close to the Reuss lower bounding surface Φ_R for

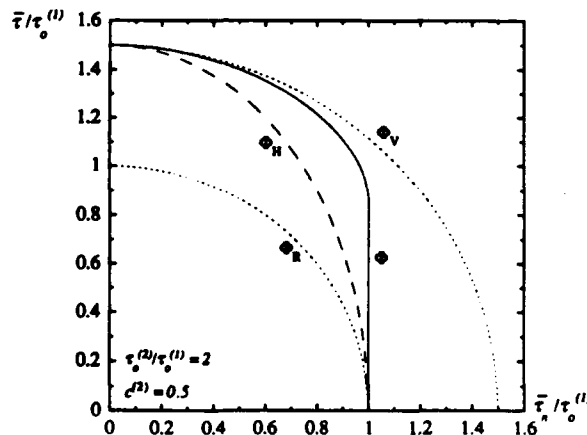


Fig. 10. Plots of the exact estimate, the anisotropic elliptic estimate of Hill, and the Voigt and Reuss isotropic estimates for the effective yield surface of a laminated composite with $\tau_0^{(2)}/\tau_0^{(1)} = 2$ and $c^{(2)} = 0.5$.

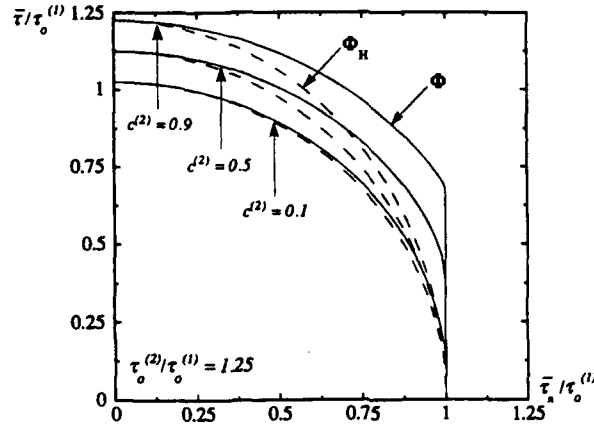


Fig. 11. Plots of the exact estimates (continuous lines) and the corresponding anisotropic elliptic estimates of Hill (dashed lines) for the effective yield surfaces of laminated composites with $\tau_0^{(2)}/\tau_0^{(1)} = 1.25$ and three different values of $c^{(2)}$ (0.1, 0.5 and 0.9).

low values of $\bar{\tau}$. We also include in Fig. 10 an estimate for the yield surface, Φ_H , which is based on the approximation of Hill (1951) for slightly anisotropic materials. This approximate yield surface is given by

$$\Phi_H(\bar{\sigma}) \equiv \frac{\bar{\tau}^2}{(c^{(1)}\tau_0^{(1)} + c^{(2)}\tau_0^{(2)})^2} + \frac{\bar{\tau}_n^2}{(\tau_0^{(1)})^2} - 1, \quad (55)$$

and we note that it amounts to an elliptic interpolation between the Voigt and Reuss yield functions.

We observe that for the largely anisotropic case depicted in Fig. 10 ($\tau_0^{(2)}/\tau_0^{(1)} = 2$ and $c^{(2)} = 0.5$), Hill's elliptic approximation severely underestimates the ultimate yield strength of the laminated composite for combined longitudinal and transverse loading. Figure 11 shows plots of the exact yield surfaces (continuous lines) and Hill's approximate yield surfaces (dashed lines) for a laminated composite with slight anisotropy ($\tau_0^{(2)}/\tau_0^{(1)} = 1.25$) and three values of $c^{(2)}$ (0.1, 0.5 and 0.9). For all values of $c^{(2)}$, the exact yield criterion bounds a larger region of the $(\bar{\tau}_n, \bar{\tau})$ -plane than the Hill approximate criterion, and the two curves are only in good agreement for small volume fractions of the stronger phase ($c^{(2)} = 0.1$).

7. CLOSURE

In this paper, we have described the application of a new variational method, developed by Ponte Castañeda (1991a, 1992), to determine the effective constitutive behavior of laminated composites with elastoplastic phases in prescribed volume fractions. It constitutes one of the first applications of the method to composite materials with anisotropic symmetries [see also Ponte Castañeda (1992) and deBotton and Ponte Castañeda (1992) for the corresponding results for fiber-reinforced materials]. Because of the simplicity of the laminated microstructure, allowing for the determination of the *exact* effective properties of laminated composites, this work is of interest—not only on account of its practical significance—but also because it provides a simple case to evaluate the power of the new method. Additionally, the results of this paper suggest that when dealing with strongly anisotropic materials, it is *not* enough to consider the behavior of the composite under special loading conditions, since the behavior of the composite under different types of loading conditions may be *dramatically* different. Thus, we found that nonlinearity highlighted the differences in the constitutive response of laminated composites under transverse and longitudinal shear loading. Further, this study also underlined the significant coupling that may arise between different loading modes in nonlinear anisotropic composites. Thus, it was found that a small fixed preload of a laminate in the longitudinal direction leads to

continued increase of the longitudinal shear strain as the level of transverse shear stress (for example) is increased. It is anticipated that the features uncovered by the present analysis of nonlinear laminated composites will also be important in other types of nonlinear composites with anisotropic symmetries, such as the practically important class of fiber-reinforced materials.

Acknowledgements—This research was supported by the Air Force Office of Scientific Research (Grant No. 91-0161). Additional support in the form of computing equipment by the Research Foundation of the University of Pennsylvania is gratefully acknowledged. The motivation for Appendix D arose from a comment by Professor J. R. Willis in connection with analogous results in electrostatics.

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APPENDIX A: ON THE CHARACTERIZATION OF TRANSVERSELY ISOTROPIC MATERIALS

The purpose of this appendix is to gather some results relevant to the analysis of linearly elastic materials with transversely isotropic symmetry. These results are used extensively throughout the body of the paper in the development of effective stress/strain relations for nonlinear laminated composites. The emphasis of this section is on representations for the transversely isotropic invariants of the stress and strain tensors. The reason is that nonlinear transversely isotropic materials are most efficiently characterized in terms of energy-density functions depending on these invariants.

A.1. Isotropic invariants

It is well known that there are three isotropic invariants for a symmetric, second-order tensor. However, only two of these—those that are of quadratic order, or less—are relevant to linearly elastic behavior. These invariants may be expressed [see, for example, Walpole (1981)] in terms of two fourth-order projection tensors $\mathbf{J}$ and $\mathbf{K}$, such that $\mathbf{I} = \mathbf{J} + \mathbf{K}$, $\mathbf{J}\mathbf{J} = \mathbf{J}$, $\mathbf{K}\mathbf{K} = \mathbf{K}$ and $\mathbf{J}\mathbf{K} = \mathbf{0}$. Their Cartesian components are given by

$$J_{ijkl} = \frac{1}{2}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} - \frac{2}{3}\delta_{ij}\delta_{kl}), \quad K_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}), \quad (A1)$$

where δ_{ij} is the Kronecker delta symbol. Then, in terms of these projection tensors, we define two isotropic invariants of the stress tensor via

$$\sigma_m = \frac{1}{3}J_{ijkl}\sigma_{ij}\sigma_{kl} \quad \text{and} \quad \tau_c^2 = \frac{1}{2}K_{ijkl}\sigma_{ij}\sigma_{kl}, \quad (A2)$$

called the hydrostatic (mean) stress, and the effective shear stress, respectively. We also define the hydrostatic strain ϵ_m , and the effective shear strain γ_c by relations completely analogous to (A2).

It is important to note that the elasticity tensor $\mathbf{L}$ of an isotropic, linearly elastic material admits a spectral decomposition

$$\mathbf{L} = 3\kappa\mathbf{J} + 2\mu\mathbf{K}, \quad (A3)$$

where $\mathbf{J}$ and $\mathbf{K}$ play the role of the eigenprojections, and the bulk and shear moduli of the material, κ and μ , are the corresponding eigenvalues. As we will see next, the situation for transversely isotropic materials is different.

A.2. Transversely isotropic invariants

There are in general five transversely isotropic invariants of a symmetric, second-order tensor (Spencer, 1971). However, only four of these invariants are linear, or quadratic, in order. They may be represented in terms of the four projections tensors [see Walpole (1981)] $\mathbf{E}^{(1)}$, $\mathbf{E}^{(2)}$, $\mathbf{E}^{(3)}$ and $\mathbf{E}^{(4)}$, satisfying the relations $\mathbf{E}^{(p)}\mathbf{E}^{(q)} = \mathbf{E}^{(p)}$; $\mathbf{E}^{(p)}\mathbf{E}^{(q)} = \mathbf{0}$, $p \neq q$; and $\mathbf{E}^{(1)} + \mathbf{E}^{(2)} + \mathbf{E}^{(3)} + \mathbf{E}^{(4)} = \mathbf{I}$. The components of these four projections tensors are given respectively by

$$\begin{aligned} E_{ijkl}^{(1)} &= \frac{1}{2}\beta_{ij}\beta_{kl}, \\ E_{ijkl}^{(2)} &= \alpha_{ij}\alpha_{kl}, \\ E_{ijkl}^{(3)} &= \frac{1}{2}(\beta_{ik}\beta_{jl} + \beta_{il}\beta_{jk} - \beta_{ij}\beta_{kl}), \\ E_{ijkl}^{(4)} &= \frac{1}{2}(\beta_{ik}\alpha_{jl} + \beta_{il}\alpha_{jk} + \beta_{jl}\alpha_{ik} + \beta_{jk}\alpha_{il}), \end{aligned} \quad (A4)$$

where $\alpha_{ij} = n_i n_j$ and $\beta_{ij} = \delta_{ij} - n_i n_j$, with $\mathbf{n}$ denoting the axis of transverse isotropy. Then, the four transversely isotropic invariants of the stress tensor σ may be expressed in the forms

$$\begin{aligned} \sigma_p &= \frac{1}{2}E_{ijkl}^{(1)}\sigma_{ij}\sigma_{kl} = \frac{1}{2}\sigma_{ij}\beta_{ij}, \quad \{\frac{1}{2}(\sigma_{11} + \sigma_{22})\}, \\ \sigma_n &= E_{ijkl}^{(2)}\sigma_{ij}\sigma_{kl} = \sigma_{ij}\alpha_{ij}, \quad \{\sigma_{33}\}, \\ \tau_p^2 &= \frac{1}{2}\sigma_{ij}E_{ijkl}^{(3)}\sigma_{kl} = \frac{1}{2}[\sigma_{ij}\sigma_{kl}\beta_{ik}\beta_{jl} - \frac{1}{2}(\sigma_{ij}\beta_{ij})^2], \quad \{\sigma_{12}^2 + \frac{1}{4}(\sigma_{11} - \sigma_{22})^2\}, \\ \tau_n^2 &= \frac{1}{2}\sigma_{ij}E_{ijkl}^{(4)}\sigma_{kl} = [\sigma_{ij}\sigma_{kl}\alpha_{ik}\alpha_{jl} - (\sigma_{ij}\alpha_{ij})^2], \quad \{(\sigma_{13}^2 + \sigma_{23}^2)\}, \end{aligned} \quad (A5)$$

which correspond physically to the in-plane hydrostatic stress, the normal tensile stress, the (in-plane) transverse shear stress, and the (anti-plane) longitudinal shear stress (given in brackets are the corresponding representations for a choice of $\mathbf{n}$ aligned with the 3-direction). Analogous relations apply for the transversely isotropic invariants of the strain tensor ϵ , denoted respectively ϵ_p , ϵ_n , γ_p and γ_n . We also note for latter reference that the following two relations hold between the transversely isotropic invariants of (A5) and the isotropic invariants of (A2), namely,

$$\sigma_m = \frac{1}{3}(2\sigma_p + \sigma_n), \quad \tau_c^2 = \tau_p^2 + \tau_n^2 + \frac{1}{3}(\sigma_p - \sigma_n)^2. \quad (A6)$$

Contrary to the situation for isotropic materials, the above four projection tensors are not the eigentensors of the spectral decomposition of an arbitrary transversely isotropic material (Mehrabadi and Cowin, 1990). Such eigentensors would unfortunately involve the material moduli. Therefore, it is necessary to introduce [see Walpole (1981)] two other tensors, that are *not* projections, $\mathbf{E}^{(5)}$ and $\mathbf{E}^{(6)}$, with components

$$E_{ijkl}^{(3)} = \alpha_{ij}\beta_{kl}, \quad E_{ijkl}^{(4)} = \beta_{ij}\alpha_{kl}. \quad (A7)$$

Then, the elasticity tensor L of an arbitrary transversely isotropic material may be expressed in terms of these six tensors. It is worth mentioning that the above tensors satisfy the relation

$$J = \frac{1}{3}E^{(1)} + \frac{1}{3}E^{(2)} + \frac{1}{3}(E^{(3)} + E^{(4)}), \quad (A8)$$

and that we can additionally define for later reference an additional tensor E' such that

$$E' = E^{(3)} + E^{(4)} - K. \quad (A9)$$

This last tensor is a projection tensor, which is orthogonal to $E^{(3)}$ and $E^{(4)}$.

Finally, we remark that the energy density function of a transversely isotropic, linearly elastic material may be represented in the form

$$U(\sigma) = \psi(\sigma_p, \sigma_n, \tau_p, \tau_n), \quad (A10)$$

where ψ is a quadratic function. Then, the relation between the transversely isotropic stress and strain invariants is given by

$$\epsilon_p = \frac{1}{2} \frac{\partial \psi}{\partial \sigma_p}, \quad \epsilon_n = \frac{\partial \psi}{\partial \sigma_n}, \quad \gamma_p = \frac{1}{2} \frac{\partial \psi}{\partial \tau_p} \quad \text{and} \quad \gamma_n = \frac{1}{2} \frac{\partial \psi}{\partial \tau_n}. \quad (A11)$$

A.3. Incompressible, transversely isotropic invariants

For incompressible, transversely isotropic materials, it suffices to consider the three invariants of order less than quadratic on the space of traceless, symmetric, second-order tensors. These may be obtained in terms of the three orthogonal projection tensors $E^{(3)}$, $E^{(4)}$ and E' , defined in the previous subsection. Thus, the incompressible, transversely isotropic invariants of the stress tensors σ are τ_p , τ_n , and the deviatoric shear stress

$$\tau_d = \frac{1}{\sqrt{3}}(\sigma_p - \sigma_n), \quad (A12)$$

corresponding to the three above projections, respectively. We note further that from (A6), we have the following identity relating the effective shear stress and the incompressible, transversely isotropic invariants, $\tau_e^2 = \tau_p^2 + \tau_n^2 + \tau_d^2$. The corresponding strain invariants are denoted by γ_p , γ_n and γ_d .

Finally, we note that the elasticity tensor L of an incompressible, transversely isotropic, linearly elastic material admits a spectral decomposition of the form

$$L = 2\mu_p E^{(3)} + 2\mu_n E^{(4)} + 2\mu_d E', \quad (A13)$$

where μ_p , μ_n , μ_d , are the three shear moduli that suffice to characterize the behavior of such a material [see Lipton (1991a)].

APPENDIX B: A USEFUL IDENTITY

In this appendix, we demonstrate the following identity, which is used repeatedly in the body of the paper, namely

$$\frac{1}{\alpha} = \min_{\omega^{(r)}, \theta^{(r)}=1} \left\{ \sum_{r=1}^n \frac{c^{(r)}}{\alpha^{(r)}} (\omega^{(r)})^2 \right\}, \quad (B1)$$

where the variables $\alpha^{(r)} > 0$ ($r = 1, \dots, n$) are constant, and where the variables $\omega^{(r)}$ ($r = 1, \dots, n$) are subject to the constraint $\bar{\omega} = 1$.

We begin by letting g be the function defined by

$$g(\omega^{(r)}) = \sum_{r=1}^n \frac{c^{(r)}}{\alpha^{(r)}} (\omega^{(r)})^2. \quad (B2)$$

The choice of the set, $\omega^{(r)} = \alpha^{(r)}/\bar{\alpha}$, satisfies the constraint and is such that $g(\omega^{(r)}) = 1/\bar{\alpha}$. Consider next a second, arbitrary set, distinct from the first set, $\hat{\omega}^{(r)}$ ($r = 1, \dots, n$), such that $\hat{\omega} = 1$, and let $\theta^{(r)} = \hat{\omega}^{(r)} - \omega^{(r)}$. Then, substitution of this second set into (B2) leads to

$$g(\hat{\omega}^{(r)}) = \sum_{r=1}^n \frac{c^{(r)}}{\alpha^{(r)}} (\hat{\omega}^{(r)})^2 = \sum_{r=1}^n \frac{c^{(r)}}{\alpha^{(r)}} (\omega^{(r)})^2 + \sum_{r=1}^n \frac{c^{(r)}}{\alpha^{(r)}} (\theta^{(r)})^2 > g(\omega^{(r)}), \quad (B3)$$

where we have used the fact that $\theta = 0$. Hence, identity (B1) is demonstrated. In the body of the paper, we replace $\omega^{(r)}$ by $(1 - \omega^{(r)})$, with an appropriate modification for the constraint.

APPENDIX C: A SIMPLIFIED EXPRESSION FOR THE EFFECTIVE STRESS/STRAIN RELATIONS

Consider the following form for the effective energy function (26) of the incompressible laminated composite

$$\bar{U}(\bar{\sigma}) = \min_{\omega^{(r)}, \bar{\omega}=0} \left\{ \sum_{s=1}^n c^{(s)} \psi^{(s)}(\tau^{(s)}) \right\}, \quad (C1)$$

where

$$\tau^{(s)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_d^2)(1 - \omega^{(s)})^2 + \bar{\tau}_n^2}.$$

As shown in the body of the paper, we can eliminate the constraint $\bar{\omega} = 0$ by letting

$$\omega^{(n)} = -\frac{1}{c^{(n)}} \sum_{r=1}^{n-1} c^{(r)} \omega^{(r)}, \quad (C2)$$

and rewriting (C1) in terms of the $n-1$ optimization variables $\omega^{(r)}$ ($r = 1, \dots, n-1$) via

$$\bar{U}(\bar{\sigma}) = \min_{\omega^{(r)}, r=1, \dots, n-1} \left\{ \sum_{s=1}^{n-1} c^{(s)} \psi^{(s)}(\tau^{(s)}) + c^{(n)} \psi^{(n)}(\tau^{(n)}) \right\}, \quad (C3)$$

where the variables $\tau^{(s)}$ ($s = 1, \dots, n-1$) are the same as before, but on the other hand

$$\tau^{(n)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_d^2) \left(1 + \frac{1}{c^{(n)}} \sum_{r=1}^{n-1} c^{(r)} \omega^{(r)} \right)^2 + \bar{\tau}_n^2}.$$

Then, the $n-1$ optimization conditions of (C3) are given by the relations

$$-\frac{1}{\tau^{(r)}} (\psi^{(r)})'(\tau^{(r)}) (1 - \omega^{(r)}) + \frac{1}{\tau^{(n)}} (\psi^{(n)})'(\tau^{(n)}) \left(1 + \frac{1}{c^{(n)}} \sum_{s=1}^{n-1} c^{(s)} \omega^{(s)} \right) = 0, \quad (r = 1, \dots, n-1). \quad (C4)$$

If we now denote the optimal variables $\omega^{(r)}$, satisfying (C4), by $\hat{\omega}^{(r)}$ ($r = 1, \dots, n-1$), the effective energy function of the incompressible laminate may then be written in the form

$$\bar{U}(\bar{\sigma}) = \sum_{s=1}^{n-1} c^{(s)} \psi^{(s)}(\hat{\tau}^{(s)}) + c^{(n)} \psi^{(n)}(\hat{\tau}^{(n)}), \quad (C5)$$

where

$$\hat{\tau}^{(s)} = \tau^{(s)}(\omega^{(s)} = \hat{\omega}^{(s)}) \quad \text{and} \quad \hat{\tau}^{(n)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_d^2) \left(1 + \frac{1}{c^{(n)}} \sum_{r=1}^{n-1} c^{(r)} \hat{\omega}^{(r)} \right)^2 + \bar{\tau}_n^2}.$$

It follows that effective stress/strain relations of the laminated composite may be computed from the relations

$$\begin{aligned} \bar{\epsilon} = & \sum_{r=1}^{n-1} \frac{c^{(r)}}{\hat{\tau}^{(r)}} (\psi^{(r)})'(\hat{\tau}^{(r)}) \left[(1 - \hat{\omega}^{(r)})^2 \left(\bar{\tau}_p \frac{\partial \bar{\tau}_p}{\partial \bar{\sigma}} + \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right) + \bar{\tau}_n \frac{\partial \bar{\tau}_n}{\partial \bar{\sigma}} \right] + \dots \\ & \dots + \frac{c^{(n)}}{\hat{\tau}^{(n)}} (\psi^{(n)})'(\hat{\tau}^{(n)}) \left[\left(1 + \frac{1}{c^{(n)}} \sum_{s=1}^{n-1} c^{(s)} \hat{\omega}^{(s)} \right)^2 \left(\bar{\tau}_p \frac{\partial \bar{\tau}_p}{\partial \bar{\sigma}} + \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right) + \bar{\tau}_n \frac{\partial \bar{\tau}_n}{\partial \bar{\sigma}} \right] + \dots \\ & \dots + \sum_{r=1}^{n-1} c^{(r)} (\bar{\tau}_p^2 + \bar{\tau}_d^2) \frac{\partial \hat{\omega}^{(r)}}{\partial \bar{\sigma}} \left[-\frac{1}{\hat{\tau}^{(r)}} (\psi^{(r)})'(\hat{\tau}^{(r)}) (1 - \hat{\omega}^{(r)}) + \frac{1}{\hat{\tau}^{(n)}} (\psi^{(n)})'(\hat{\tau}^{(n)}) \left(1 + \frac{1}{c^{(n)}} \sum_{s=1}^{n-1} c^{(s)} \hat{\omega}^{(s)} \right) \right]. \quad (C6) \end{aligned}$$

We note that each of the terms in the last summation of (C6) is identical to zero by virtue of the optimizations conditions (C4). Thus, in the computation of the effective stress/strain relations, we may regard the optimizations variables as constants as far as derivatives with respect to $\bar{\sigma}$ are concerned, to obtain the final result

$$\bar{\epsilon} = \sum_{r=1}^n \frac{c^{(r)}}{\hat{\tau}^{(r)}} (\psi^{(r)})'(\hat{\tau}^{(r)}) \left[(1 - \hat{\omega}^{(r)})^2 \left(\bar{\tau}_p \frac{\partial \bar{\tau}_p}{\partial \bar{\sigma}} + \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right) + \bar{\tau}_n \frac{\partial \bar{\tau}_n}{\partial \bar{\sigma}} \right], \quad (C7)$$

where $\hat{\omega}^{(n)}$ is defined via the relation (C2) in terms of the other $\hat{\omega}^{(r)}$ ($r = 1, \dots, n-1$).

It can be shown that an analogous result may be obtained for the nonlinear compressible composite with effective energy function $\bar{U}$ given by (37). In fact, we may write the effective stress/strain relations for the nonlinear compressible laminate in the form

$$\bar{\epsilon} = \sum_{r=1}^n c^{(r)} \frac{\partial \psi^{(r)}}{\partial \bar{\sigma}} (\bar{\epsilon}_e^{(r)}, \bar{\sigma}_m^{(r)}). \quad (C8)$$

where $\bar{\epsilon}_e^{(r)}$, and $\bar{\sigma}_m^{(r)}$ are evaluated from (36) at the optimal values of $\omega_e^{(r)}$, and $\omega_m^{(r)}$, denoted by $\bar{\omega}_e^{(r)}$, and $\bar{\omega}_m^{(r)}$, respectively. Here, the derivatives with respect to the average stress $\bar{\sigma}$ are evaluated with $\bar{\omega}_e^{(r)}$, and $\bar{\omega}_m^{(r)}$ fixed.

APPENDIX D: AN ALTERNATIVE DERIVATION OF EXPRESSION (37)

Having obtained expressions (35) and (37) for the effective energy functions of the linear and nonlinear laminated composites $\bar{U}_0$ and $\bar{U}$, respectively, we note that the form of these expressions is reminiscent of the type of result that one would expect from direct utilization of the principle of minimum complementary energy (4). In this appendix, we briefly show that results (35) and (37) for $\bar{U}_0$ and $\bar{U}$, respectively, can indeed be alternatively obtained from the principle of minimum complementary energy. It is important to emphasize, however, that while the derivations given in the body of the paper result from straightforward computations, the present derivations based on the principle of minimum complementary energy rely more directly on the physics of the problem, and were motivated by the prior derivations. Additionally, the case of a laminated composite is a very special microstructure; in general, we do not expect that we will be able to use the approach of this appendix for nonlinear composites with more general anisotropic microstructures.

We begin with the derivation of the linear result (35). We have already mentioned that the stress field within the laminated composite is piecewise constant, i.e. of the form

$$\sigma = \sum_{r=1}^n \chi^{(r)}(\mathbf{x} \cdot \mathbf{n}) \sigma^{(r)},$$

where $\sigma^{(r)}$ corresponds to the constant stress field in phase r . The problem then reduces to that of finding these unknown phase stresses $\sigma^{(r)}$, together with the corresponding constant strain fields $\epsilon^{(r)}$ (related to the stresses by the phase constitutive relations), and satisfying the conditions of continuity of the traction stresses and tangential strains across the interfaces between the phases, as well as the averaging conditions stated in Section 2.

In this connection, the interior and exterior projection operators of Hill (1972, 1983) $\mathbf{F} = \mathbf{E}^{(1)} + \mathbf{E}^{(3)}$ and $\mathbf{E} = \mathbf{E}^{(2)} + \mathbf{E}^{(4)}$ (refer to Appendix A), respectively, turn out to be useful because they allow the decomposition of any symmetric, second-order tensor into its tangential (interior) and traction (exterior) components (with reference to a boundary with normal $\mathbf{n}$). Thus, the tangential components of the strain (which must be continuous across interfacial boundaries on the laminated composite) are given by $\mathbf{F}\epsilon$, and, correspondingly, the traction components of the stress (which must also be continuous across the interfacial boundaries) are given by $\mathbf{E}\sigma$. Alternatively, we may state that $\mathbf{E}^{(2)}\sigma$, $\mathbf{E}^{(4)}\sigma$ and $\mathbf{E}^{(1)}\epsilon$, $\mathbf{E}^{(3)}\epsilon$ must also be continuous across such boundaries.

Next, we apply the above results to the laminated composite, for which the interfacial boundaries are all perpendicular to a fixed vector $\mathbf{n}$. Since the traction stresses must be continuous from phase to phase, we have that

$$\mathbf{E}^{(2)}\sigma^{(r)} = \mathbf{E}^{(2)}\bar{\sigma}, \quad \text{and} \quad \mathbf{E}^{(4)}\sigma^{(r)} = \mathbf{E}^{(4)}\bar{\sigma}, \quad (D1)$$

where we have additionally made use of the average stress condition given in Section 2. We continue by noting that for an isotropic material (as are all the phases in the our laminate), $\mathbf{E}^{(3)}\sigma^{(r)} = 2\mu_0^{(r)}\mathbf{E}^{(3)}\epsilon^{(r)}$ within each linear phase, and therefore for an isotropic phase the $\mathbf{E}^{(3)}$ projection of the stress tensor must have the same direction in all phases. Thus, applying the averaging condition for the stresses, we arrive at

$$\mathbf{E}^{(3)}\sigma^{(r)} = (1 - \omega_e^{(r)})\mathbf{E}^{(3)}\bar{\sigma}, \quad (D2)$$

where the variables $\omega_e^{(r)}$ must satisfy the condition that $\bar{\omega}_e = 0$. Additionally, since the $\mathbf{E}^{(1)}$ -projection is one-dimensional, it follows that the $\mathbf{E}^{(1)}$ -projections of the stress tensor must also be parallel from phase to phase. Therefore, applying the averaging condition for the stresses, we have that

$$\mathbf{E}^{(1)}\sigma^{(r)} = (1 - \omega_m^{(r)})\mathbf{E}^{(1)}\bar{\sigma}, \quad (D3)$$

where the variables $\omega_m^{(r)}$ must satisfy the condition that $\bar{\omega}_m = 0$. We note, however, that if $\mathbf{E}^{(1)}\bar{\sigma} = 0$ (or, equivalently, if $\bar{\sigma}_p = 0$), the above result does not hold, because in this case the corresponding projections of the stress in the phases need not vanish (only their average needs to vanish).

Applying the results of Appendix A [in particular, (A6)], we conclude that the isotropic invariants of the stress tensor within each phase $\epsilon^{(r)}$ and $\sigma^{(r)}$ (on which the energy-density functions of each isotropic phase depend) are precisely those given by relations (36). Therefore, it follows from the principle of minimum complementary energy—by minimizing over the set of admissible stresses (i.e. over the optimizing variables $\omega_e^{(r)}$ and $\omega_m^{(r)}$ subject to the constraints $\bar{\omega}_e = 0$ and $\bar{\omega}_m = 0$)—that the effective energy function $\bar{U}_0$ of the linear composite is indeed given by expression (35).

For the nonlinear laminated composite, we observe that the same analysis given above would also work, leading to expression (37) for $\bar{U}$. The only modification that is required in this analysis is that for a nonlinear isotropic phase (say phase r), the relation $\mathbf{E}^{(3)}\sigma^{(r)} = 2\mu_0^{(r)}\mathbf{E}^{(3)}\epsilon^{(r)}$ would not hold, but it can be easily shown that for the nonlinear isotropic material of the type considered in this work, the conclusion (D2) would still hold, and hence the final form for $\bar{U}$ would be the same as that for the linear laminated composite $\bar{U}_0$.

We conclude this appendix by stating an alternative form of (35) and (37) that works even when $\bar{\sigma}_p = 0$. This is accomplished by redefining the optimizing variables $\omega_m^{(r)}$ in terms of the new variables

$$\dot{\omega}_m^{(i)} = \frac{\bar{\sigma}_p \omega_m^{(i)} - \bar{\sigma}_n}{\bar{\sigma}_p - \bar{\sigma}_n}, \quad (D4)$$

where we now need to have $\bar{\sigma}_p - \bar{\sigma}_n \neq 0$. In terms of the new variables $\dot{\omega}_m^{(i)}$ (the variables $\omega_c^{(i)}$ do not change), relation (37) is expressed in the form

$$\bar{U}(\bar{\sigma}) = \min_{\substack{\omega_c^{(i)}, \dot{\omega}_m^{(i)} \\ \dot{\omega}_c = \dot{\omega}_m = 0}} \left\{ \sum_{i=1}^n c^{(i)} \psi^{(i)}(\bar{\epsilon}_c^{(i)}, \bar{\sigma}_m^{(i)}) \right\}, \quad (D5)$$

where

$$\bar{\epsilon}_c^{(i)} = \sqrt{(1 - \omega_c^{(i)})^2 \bar{\epsilon}_p^2 + \bar{\epsilon}_n^2 + \frac{1}{2}[(\bar{\sigma}_n - \bar{\sigma}_p)(1 - \dot{\omega}_m^{(i)})]^2} \quad \text{and} \quad \bar{\sigma}_m^{(i)} = \bar{\sigma}_n + \frac{1}{2}(\bar{\sigma}_p - \bar{\sigma}_n)(1 - \dot{\omega}_m^{(i)}). \quad (D6)$$

Note that when $\bar{\sigma}_p = \bar{\sigma}_n = 0$, we are guaranteed that $\sigma_p^{(i)} = \sigma_n^{(i)} = 0$ in each phase, and then both forms are equally valid.

Reference [7]

ELASTOPLASTIC CONSTITUTIVE RELATIONS FOR FIBER-REINFORCED SOLIDS

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Abstract

In this paper, we make use of a procedure for estimating the effective properties of nonlinear composite materials, proposed recently by Ponte Castañeda (1991a), to study the effective constitutive behavior of ductile fiber-reinforced composites. Both estimates and rigorous bounds are obtained for the effective energy functions of multiple-phase fiber composites with general plastic behaviors (in the context of deformation theory of plasticity) for the isotropic constituent phases. The resulting expressions for the energy functions may be differentiated in a straightforward manner to obtain corresponding estimates for the anisotropic effective stress-strain relations. Explicit calculations are carried out for the case of an aluminum-matrix composite reinforced with boron fibers. The results reveal some interesting features distinguishing the constitutive behavior of plastic fiber composites from that of linear-elastic fiber composites. One such feature is the strong coupling between the dilatational and distortional modes for the plastic fiber composites. Finally, comparisons are made with available experimental data.

1. Introduction

Fiber-reinforced composites are commonly used materials, and their mechanical properties have been the subject of extensive investigations. However, most of the work to date has addressed exclusively the *linear*-elastic behavior of these materials; details and references can be found in a report by Hashin (1972) and review articles by Willis (1981, 1982) and Hashin (1983). In this paper, we are concerned with the overall behavior of fiber-reinforced composites in which one or more of the phases undergoes *plastic* deformation. The number of papers dealing with this aspect of the behavior of fiber composites is comparatively small. Next, we give a brief review of some of the relevant contributions.

Among the first contributions, Hill (1964b) extended analogous results for the overall elastic moduli of linear-elastic fiber composites (Hill 1964a) to obtain corresponding estimates for the incremental moduli of ductile fiber composites (in the context of flow theory of plasticity). An alternative approach using the methods of limit analysis to estimate the overall yield strength of composites (see Drucker 1959) was applied by Shu and Rosen (1967), Majumdar and McLaughlin (1975) and de Buhan *et al.* (1990) to fiber composites. Micromechanical models involving some empirical adjustments were developed by Hashin *et al.* (1974), Dvorak and Bahei-El-Din (1987) and Sun and Chen (1991), among others, to predict the yielding and post-yielding behavior of fiber composites. The predictions of some of these models were tested experimentally by Dvorak *et al.* (1988) on a boron/aluminum system. More recently, Zhao and Weng (1990) developed an approximate procedure, based on the Mori-Tanaka (1973) method, for estimating the effective constitutive relations of composites reinforced by aligned spheroidal inclusions, which include fibers as a special case. On the other hand, Talbot and Willis (1991) provided rigorous bounds for the effective energy functions of ductile fiber-reinforced composites by application of the nonlinear generalization of the Hashin and Shtrikman (1962) variational principles due to Talbot and Willis (1985). These authors carried out explicit calculations for the case of incompressible fiber-reinforced composites. Ponte Castañeda (1992) has also obtained simple bounds and estimates of the Hashin-Shtrikman type for the effective energy functions of incompressible fiber composites

through a different variational procedure, developed by Ponte Castañeda (1991a). Finally, Suquet (1992) made use of an altogether different method to obtain bounds for the overall energy functions of power-hardening materials weakened by cylindrical voids.

Our aim in this paper is to develop and generalize the results of Ponte Castañeda (1992) for fiber-reinforced composites. The method used is based on a variational principle that enables the expression of the effective energy functions of nonlinear composites in terms of optimization problems involving the effective moduli of appropriate families of linear *comparison* composites. Thus, the variational principle suggests a procedure for generating bounds and estimates for the effective behavior of nonlinear composites from *known* estimates and bounds for linear composites with similar microstructures. The procedure was applied by Ponte Castañeda (1991a, b; 1992) to statistically isotropic composites with *general* isotropic constitutive behaviors for the phases. The specific examples of nonlinear materials reinforced by rigid and linear-elastic inclusions, or weakened by voids, were considered in these references. The procedure may also be applied to anisotropic composites. This has been done in some detail by deBotton and Ponte Castañeda (1992) for ductile laminated composites. In this work, we continue our study of the behavior of anisotropic nonlinear composites by considering the application of the procedure to transversely isotropic, fiber-reinforced composites with general *compressible* behavior for the isotropic phases.

The rest of the paper is arranged as follows. In Section 2, we review the definition of effective properties and recall the variational principle of Ponte Castañeda (1992). Next, in Section 3, we make use of the bounds of Hill (1964a) and Hashin (1965) for linear-elastic fiber composites to generate corresponding bounds and estimates for ductile fiber composites. In sections 4, 5 and 6, we consider some special classes of fiber-reinforced composites, for which the expressions for the bounds and estimates of Section 3 may be simplified further. Thus, we consider the cases of general multiphase *incompressible* fiber composites, hollow-fibers composites, and two-phase, compressible metal-matrix composites. Finally, in Section 7, we compute the effective stress-strain relations of a specific aluminum-matrix composite, reinforced by linear-elastic boron fibers.

2. Effective properties and their variational characterization

In this section, we briefly review the definition of effective properties and their variational characterization. More general discussions, in the context of the inelastic behavior of composite materials, may be found in the articles of Hill (1967) and Suquet (1985). For our purposes, a composite is a heterogeneous material made up of two or more distinct phases, and characterized by two separate length scales: a macroscopic scale L , and a microscopic scale l , such that $l \ll L$. The macroscopic scale describes the gross size of the specimen and the scale of variation of the applied loading, and the microscopic scale characterizes the size of the typical inhomogeneity in the material. Thus, a composite is microscopically heterogeneous, but macroscopically homogeneous.

We consider a representative specimen of the composite Ω , with boundary $\partial\Omega$. For simplicity, we choose units such that the volume of the specimen is unity. We assume that the constitutive behavior of the distinct phases in the composite is characterized by the deformation theory of plasticity or, equivalently, by nonlinear infinitesimal elasticity. However, we note that the usual *approximate* extensions may be made for composite materials characterized by the flow theory of plasticity (see Budiansky 1959 and Hashin *et al.* 1974). Thus, at a point $\mathbf{x} \in \Omega$, the relation between the strain field $\boldsymbol{\varepsilon}(\mathbf{x})$ and the stress field $\boldsymbol{\sigma}(\mathbf{x})$ is given by

$$(1) \quad \boldsymbol{\varepsilon}(\mathbf{x}) = \frac{\partial U(\mathbf{x}, \boldsymbol{\sigma})}{\partial \boldsymbol{\sigma}},$$

where $U(\mathbf{x}, \boldsymbol{\sigma})$ is the local complementary energy-density function of the composite.

Then (following Hill 1963), when the composite is subjected to the uniform traction condition

$$(2) \quad \boldsymbol{\sigma} \mathbf{n} = \bar{\boldsymbol{\sigma}} \mathbf{n}, \quad \mathbf{x} \in \partial\Omega,$$

where $\mathbf{n}$ is the outward unit normal to $\partial\Omega$ and $\bar{\boldsymbol{\sigma}}$ is a constant symmetric tensor, its effective behavior may be characterized in terms of the *effective complementary-energy function* $\tilde{U}$, such that

$$(3) \quad \bar{\boldsymbol{\varepsilon}} = \frac{\partial \tilde{U}}{\partial \bar{\boldsymbol{\sigma}}},$$

where $\bar{\epsilon}$ is the mean value of the strain field. We also recall that under the boundary condition (2), the mean value of the stress field is precisely $\bar{\sigma}$.

In view of (3), the problem of characterizing the effective behavior of the composite reduces to that of computing its effective complementary energy-density function $\tilde{U}$. This may be accomplished directly by means of the principle of minimum complementary energy, stating that

$$(4) \quad \tilde{U}(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} \int_{\Omega} U(\mathbf{x}, \sigma) dv,$$

where

$$(5) \quad S(\bar{\sigma}) = \{ \sigma \mid \nabla \cdot \sigma = 0 \text{ in } \Omega, \text{ and } \sigma \mathbf{n} = \bar{\sigma} \mathbf{n} \text{ on } \partial\Omega \}$$

is the set of *statically* admissible stress fields. The variational principle (4) is equivalent to a standard boundary value problem, governed by the equilibrium and the compatibility equations, together with the boundary conditions (2). We note that composite materials may exhibit sharp interfaces across which material properties are discontinuous. Consequently, at these interfaces, the equilibrium and compatibility equations must be interpreted in their weak forms enforcing continuity of the tractions and of the tangential components of the strain tensor, respectively.

In addition to the analytical difficulties associated with the heterogeneity of the problem, difficulties also arise because of the nonlinearity of the problem. Precisely to deal with this later difficulty, a variational procedure was introduced by Ponte Castañeda (1991a, 1992). This procedure is based upon a variational principle that expresses the effective energy function of a given nonlinear composite in terms of an optimization problem involving the effective energy functions of a class of *linear comparison composites*. Consequently, well-known estimates and bounds for the effective energy functions of linear composites may be used to generate corresponding estimates and bounds for the effective energy functions of nonlinear composites. Here, we will make use of this method, together with existing results for linear-elastic fiber composites, to obtain bounds and estimates for the behavior of nonlinear, ductile fiber composites.

We will restrict ourselves to composites where all the individual constituents are isotropic with energy-density functions depending only on the first and second isotropic invariants of the stress tensor. Thus, the local complementary energy function may be expressed in the form

$$(6) \quad U(\mathbf{x}, \boldsymbol{\sigma}) = \psi(\mathbf{x}; \tau_e, \sigma_m),$$

where ψ is a nonnegative function, which is convex in its last two arguments and satisfies the condition $\psi(\mathbf{x}; 0, 0) = 0$ for all $\mathbf{x}$. The precise definitions of the two isotropic invariants, the *mean* (hydrostatic) stress σ_m and the *effective shear* stress τ_e , are given by relation (I.2). Additionally, we assume that there exists a function $f(\mathbf{x}, v_e, v_m) = \psi(\mathbf{x}, \tau_e, \sigma_m)$ with $v_e = \tau_e^2$ and $v_m = \sigma_m^2$, such that f is convex in its last two arguments (this is the so-called strong convexity hypothesis). This assumption, implying that the dependence of U on the magnitude of the stress tensor is *stronger than quadratic*, is consistent with the anticipated behavior of elastoplastic materials.

In the remainder of this section, we briefly review the variational principle. For further details, we refer the reader to Ponte Castañeda (1992). First, we note that under the above strong convexity assumption, the energy-density function U admits the representation

$$(7) \quad U(\mathbf{x}, \boldsymbol{\sigma}) = \max_{\mu_o, \kappa_o \geq 0} \{U_o(\mathbf{x}, \boldsymbol{\sigma}) - V(\mathbf{x}; \mu_o, \kappa_o)\},$$

where

$$(8) \quad U_o(\mathbf{x}, \boldsymbol{\sigma}) = \frac{1}{2\mu_o(\mathbf{x})} \tau_e^2 + \frac{1}{2\kappa_o(\mathbf{x})} \sigma_m^2,$$

corresponds to the local energy-density function of a linear-elastic solid with shear modulus $\mu_o(\mathbf{x})$ and bulk modulus $\kappa_o(\mathbf{x})$, and where

$$(9) \quad V(\mathbf{x}; \mu_o, \kappa_o) = \max_{\boldsymbol{\sigma}} \{U_o(\mathbf{x}, \boldsymbol{\sigma}) - U(\mathbf{x}, \boldsymbol{\sigma})\}.$$

Then, substitution of (7) into (4) yields the variational statement

$$(10) \quad \tilde{U}(\bar{\boldsymbol{\sigma}}) = \max_{\mu_o(\mathbf{x}), \kappa_o(\mathbf{x}) \geq 0} \left\{ \tilde{U}_o(\bar{\boldsymbol{\sigma}}) - \int_{\Omega} V(\mathbf{x}; \mu_o(\mathbf{x}), \kappa_o(\mathbf{x})) dx \right\},$$

where

$$(11) \quad \tilde{U}_o(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma} \in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U_o(\mathbf{x}, \boldsymbol{\sigma}) dx,$$

is the effective energy function of the linear comparison composite with local energy-density function U_o , as given by relation (8). We emphasize that, under the strong convexity hypothesis, the variational statement (10) (in terms of the linear comparison composite) is completely equivalent to the classical principle of minimum complementary energy.

Next, we specialize the above variational statement to the case of composites with distinct homogeneous phases. Thus, we consider composites made up of n isotropic phases. Each phase is governed by an arbitrary complementary energy-density function satisfying the strong convexity assumption, with $U^{(r)}(\boldsymbol{\sigma}) = \psi^{(r)}(\boldsymbol{\tau}_e, \boldsymbol{\sigma}_m)$ ($r = 1, \dots, n$). Then, the local energy function of the composite may be written

$$(12) \quad U(\mathbf{x}, \boldsymbol{\sigma}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) U^{(r)}(\boldsymbol{\sigma}),$$

where $\chi^{(r)}$ (equals 1 when $\mathbf{x}$ is in phase r , and 0 otherwise) is the characteristic function of the r th phase. The volume fraction of the r th phase is given by

$$(13) \quad c^{(r)} = \int_{\Omega} \chi^{(r)}(\mathbf{x}) d\mathbf{x}.$$

An estimate for the effective energy function of the nonlinear composite may be obtained by restricting the set of arbitrary comparison moduli $\mu_o(\mathbf{x})$ and $\kappa_o(\mathbf{x})$ in (10), to the set of piecewise constant moduli (with a different, but constant, modulus over each phase). Consequently, the variational principle (10) yields a bound for the effective energy function of the nonlinear composite (Ponte Castañeda 1992), given by

$$(14) \quad \tilde{U}(\bar{\boldsymbol{\sigma}}) \geq \max_{\mu_o^{(r)}, \kappa_o^{(r)} \geq 0} \left\{ \tilde{U}_o(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^n c^{(r)} V^{(r)}(\mu_o^{(r)}, \kappa_o^{(r)}) \right\},$$

where

$$(15) \quad V^{(r)}(\mu_o^{(r)}, \kappa_o^{(r)}) = \max_{\boldsymbol{\sigma}} \{ U_o^{(r)}(\boldsymbol{\sigma}) - U^{(r)}(\boldsymbol{\sigma}) \},$$

and

$$(16) \quad U_o^{(r)}(\boldsymbol{\sigma}) = \frac{1}{2\mu_o^{(r)}} \tau_e^2 + \frac{1}{2\kappa_o^{(r)}} \sigma_m^2.$$

In (14), $\tilde{U}_o$ corresponds to the effective complementary-energy function of a linear composite with the same distribution of phases as the nonlinear composite, *i.e.*,

$$\tilde{U}_o(\bar{\sigma}) = \min_{\sigma \in S(\bar{\sigma})} \int_{\Omega} \sum_{r=1}^n \chi^{(r)}(x) U_o^{(r)}(\sigma) dx.$$

Thus, each phase is homogeneous and isotropic with shear and bulk moduli $\mu_o^{(r)}$ and $\kappa_o^{(r)}$, respectively. We emphasize that expressions for $\tilde{U}_o$, in the form of bounds and estimates of various types, are available in the literature for several classes of composite materials, including fiber composites. We note that lower bounds for $\tilde{U}_o$ lead through (14) to corresponding lower bounds for $\tilde{U}$, while, on the other hand, upper bounds for $\tilde{U}_o$ lead only to estimates for the corresponding upper bounds for $\tilde{U}$ (we call such estimates *upper estimates*).

3. Application to elastoplastic fiber composites

In this section, we apply the procedure described in the previous section to obtain bounds and estimates for the effective behavior of elastoplastic fiber composites. Henceforth, the term *fiber composites*, is used to describe the class of n -phase composites with prescribed volume fractions of the isotropic phases, and overall transversely isotropic symmetry. Thus, the microstructure of this class of materials is characterized by a statistically isotropic distribution of the phases in the plane transverse to the symmetry axis $\mathbf{n}$ (see Figure 1). In Appendix I, we briefly review some of the properties of transversely isotropic materials, which will be quoted as needed in the developments to follow.

We begin by considering the general case of n -phase fiber composites. Thus, we provide rigorous lower bounds, as well as upper estimates, for the effective energy functions of such composites. In addition, we also provide corresponding expressions for the effective stress-strain relations of such composites. The lower bounds and upper estimates for the effective energy functions of the nonlinear composites are obtained via the procedure described in Section 2 in terms of the corresponding bounds for the effective energy functions of the class of linear, n -phase, comparison fiber composites.

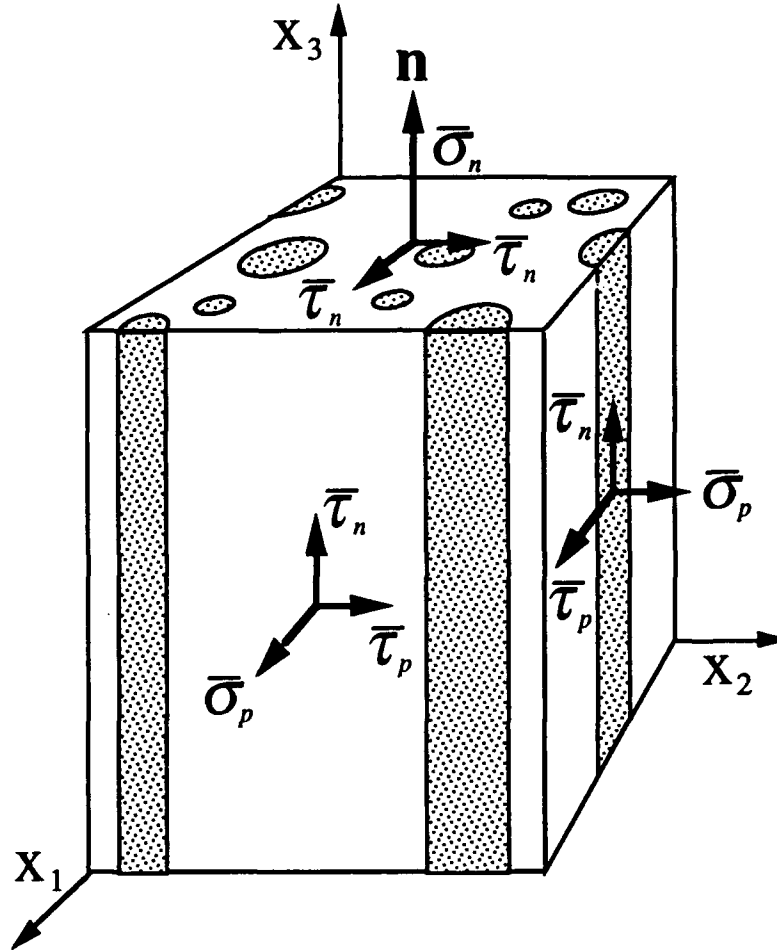


Fig. 1. A two phase fiber-reinforced composite.

The constitutive behaviors of the phases of the linear comparison composite may be expressed via the fourth-order elasticity tensors

$$(17) \quad \mathbf{M}_o^{(r)} = \frac{1}{2\mu_o^{(r)}} \mathbf{K} + \frac{1}{2\kappa_o^{(r)}} \mathbf{J},$$

where the expressions for $\mathbf{K}$ and $\mathbf{J}$, the isotropic projections of the identity tensor, are given by relation (I.1). The corresponding energy-density functions $U_o^{(r)}$ are given by (16). Bounds of the Hashin-Shtrikman (1962) type for this class of linear composites were given by Hill (1964a), Hashin (1965) and Walpole (1969).

Following Walpole's representation, we have the following lower bound for the effective energy function of the linear comparison composite, namely,

$$(18) \quad \tilde{U}_o^{(HS-)}(\bar{\sigma}) = \frac{1}{2} \bar{\sigma}_{ij} \left(\tilde{M}_o^{(HS-)} \right)_{ijkl} \bar{\sigma}_{kl},$$

where

$$(19) \quad \tilde{M}_o^{(HS-)}(\mu_o^{(r)}, \kappa_o^{(r)}) = \left[\sum_{r=1}^n c^{(r)} \left[M_o^{(r)} + M_o^*(\mu_o^{(+)}, \kappa_o^{(+)}) \right]^{-1} \right]^{-1} - M_o^*(\mu_o^{(+)}, \kappa_o^{(+)}).$$

In this later relation,

$$(20) \quad M_o^*(\mu, \kappa) = \frac{1}{2\mu} E^{[1]} + \left(\frac{1}{2\mu} + \frac{1}{\kappa + \frac{1}{3}\mu} \right) E^{[3]} + \frac{1}{2\mu} E^{[4]},$$

where the tensors $E^{[q]}$ ($q=1, \dots, 4$), the four transversely isotropic projections of the identity tensor, are given by relation (I.4), and $\mu_o^{(+)} = \max_r \{\mu_o^{(r)}\}$, $\kappa_o^{(+)} = \max_r \{\kappa_o^{(r)}\}$. Then, upon substitution of (18) into (14), the corresponding lower bound for the effective energy function of the nonlinear fiber composite becomes

$$(21) \quad \tilde{U}^{(HS-)}(\bar{\sigma}) = \max_{\mu_o^{(r)}, \kappa_o^{(r)} \geq 0} \left\{ \tilde{U}_o^{(HS-)}(\bar{\sigma}) - \sum_{r=1}^n c^{(r)} V^{(r)}(\mu_o^{(r)}, \kappa_o^{(r)}) \right\},$$

where the functions $V^{(r)}$ are given by relations (15).

Moreover, it is demonstrated in Appendix II that, once the optimization problem (21) is solved, the corresponding estimates for the effective stress-strain relations are given by

$$(22) \quad \bar{\epsilon}_{ij} = \left[\tilde{M}_o^{(HS-)}(\hat{\mu}_o^{(r)}, \hat{\kappa}_o^{(r)}) \right]_{ijkl} \bar{\sigma}_{kl},$$

where $\hat{\mu}_o^{(r)}$ and $\hat{\kappa}_o^{(r)}$ are the optimized values of the variables $\mu_o^{(r)}$ and $\kappa_o^{(r)}$, respectively. We note that, in spite of its appearance, (22) is *not* a linear relation between the average stress and strain. This is because $\tilde{M}_o^{(HS-)}$ depends on the average stress through $\hat{\mu}_o^{(r)}$ and $\hat{\kappa}_o^{(r)}$. We also note that expressions for the stress-strain relations derived from bounds on the effective energy are not guaranteed to be bounds for the effective stress-strain relations of the composite.

Expressions for an upper estimate for $\tilde{U}$, denoted $\tilde{U}^{(HS+)}$, may be obtained in an analogous manner. Thus, the upper estimate for the effective energy functions of the nonlinear fiber composites is given by a relation similar to (21) with $\tilde{U}_o^{(HS-)}$ replaced by

$$(23) \quad \tilde{U}_o^{(HS+)}(\bar{\sigma}) = \frac{1}{2} \bar{\sigma}_{ij} \left(\tilde{M}_o^{(HS+)} \right)_{ijkl} \bar{\sigma}_{kl}.$$

In this expression, $\tilde{M}_o^{(hs+)}$ is given by a relation analogous to relation (19), except that $\mu_o^{(+)}$ and $\kappa_o^{(+)}$ are replaced by $\mu_o^{(-)} = \min_r \{\mu_o^{(r)}\}$ and $\kappa_o^{(-)} = \min_r \{\kappa_o^{(r)}\}$, respectively. The associated expressions for the effective stress-strain relations are similar to relations (22), and are given in terms of $\tilde{M}_o^{(hs+)}(\hat{\mu}_o^{(r)}, \hat{\kappa}_o^{(r)})$, where $\hat{\mu}_o^{(r)}$ and $\hat{\kappa}_o^{(r)}$ are now the optimized values of the variables $\mu_o^{(r)}$ and $\kappa_o^{(r)}$ arising from the solution of the optimization problem for $\tilde{U}^{(hs+)}$.

From a practical point of view, the class of two-phase, transversely isotropic fiber-reinforced composites is probably the most important. For this case, Lipton (1991) has shown that the linear bounds (18) and (23) are optimal. Thus, the lower bound is attained by a fiber composite made up of a matrix of the stiffer phase weakened by fibers of the softer phase, while the upper bound is attained by a fiber composite made up of a matrix with the more compliant phase reinforced by fibers of the stiffer material. Therefore, the linear bounds $\tilde{U}_o^{(hs-)}$ and $\tilde{U}_o^{(hs+)}$ may be regarded as estimates for the effective energy functions of these two types of linear-elastic, fiber-reinforced composites, respectively. It follows from the discussion of Section 2 that $\tilde{U}^{(hs-)}$ and $\tilde{U}^{(hs+)}$ may be also regarded as estimates for the effective energy functions of the corresponding extremal nonlinear fiber composites. Thus, we may regard the lower bound (upper estimate) as an estimate for the effective energy function of a nonlinear fiber composite involving fibers (matrices) of the weaker phase, and matrices (fibers) of a stiffer material. In particular, the associated expressions for the stress-strain relations may also be used as estimates for the behaviors of these two types of extremal nonlinear fiber composites. In Sections 6 and 7, we will discuss this possible interpretation of the results in more detail.

To conclude this section, we note that the representations for the lower bound $\tilde{U}^{(hs-)}$ and the upper estimate $\tilde{U}^{(hs+)}$ are given in terms of $4n$ -dimensional optimization problems. From a computational point of view, obtaining the solutions of these problems is straightforward, specially because the functions $V^{(r)}$ are convex in the optimization variables $\mu_o^{(r)}$ and $\kappa_o^{(r)}$. Nevertheless, in some cases, these representations can be simplified further with the help of the identity given in Appendix III. In the following section, dealing with the special case of incompressible fiber

composites, we make use of this approach. In later sections, we consider the more complicated case of compressible fiber composites.

4. The incompressible fiber-reinforced composites

In this section, we are concerned with fiber composites that are made up of n incompressible phases (and are hence incompressible). Neglecting dependence on the third invariant of the stress, we have that the complementary energy-density functions of the phases depend only on the effective shear stress, *i.e.*,

$$(24) \quad U^{(r)}(\sigma) = \psi^{(r)}(\tau_e).$$

Then, expression (21), together with (15), reduces to the following expression for the lower bound on the effective energy function of the nonlinear fiber composite, namely,

$$(25) \quad \tilde{U}^{(HS-)}(\bar{\sigma}) = \max_{\mu_o^{(r)} \geq 0} \min_{\tau_e^{(r)}} \left\{ \tilde{U}_o^{(HS-)}(\bar{\sigma}) - \sum_{r=1}^n c^{(r)} \left[\frac{1}{2\mu_o^{(r)}} (\tau_e^{(r)})^2 - \psi^{(r)}(\tau_e^{(r)}) \right] \right\}.$$

In the above relation, the lower bound for the effective energy function of a linear comparison composite (made up of n incompressible isotropic phases) $\tilde{U}_o^{(HS-)}$ is given by relation (18), with $\kappa_o^{(r)} = \infty$ ($r = 1, \dots, n$) in expressions (19) and (20) for $\tilde{M}_o^{(HS-)}$ and M_o^* , respectively. Then, by means of identity (III.2), $\tilde{U}_o^{(HS-)}$ may be rewritten in the form

$$(26) \quad \tilde{U}_o^{(HS-)}(\bar{\sigma}) = \frac{1}{2} \min_{\substack{\Omega^{(r)} \\ \bar{\Omega} = \mathbf{I}}} \left\{ \sum_{r=1}^n c^{(r)} \bar{\sigma}_{ij} \left[\Omega_{klij}^{(r)} \left((M_o^{(r)})_{klmn} + (M_o^*(\mu_o^{(+)})_{klmn} \right) \Omega_{mnpq}^{(r)} - (M_o^*(\mu_o^{(+)})_{ijpq} \right) \bar{\sigma}_{pq} \right] \right\},$$

where the optimization variables $\Omega^{(r)}$, are subject to the constraint $\sum_{r=1}^n c^{(r)} \Omega^{(r)} = \mathbf{I}$, and satisfy the symmetry conditions $\Omega_{ijkl} = \Omega_{jikl} = \Omega_{ijlk}$. In general, expression (26) involves optimization over $36n$ entries of the $\Omega^{(r)}$. However, due to the symmetries of the tensors $M_o^{(r)}$ and M_o^* , only $6n$ non-zero entries are needed. Thus, the optimization variables may be chosen in the form

$$(27) \quad \Omega^{(r)} = \sum_{q=1}^6 \omega_q^{(r)} \mathbf{E}^{[q]},$$

where the definitions of $E^{[5]}$ and $E^{[6]}$ are given in Appendix I, and where the optimization constraint implies that $\bar{\omega}_q = 1$, ($q = 1, 2, 3, 4$) and $\bar{\omega}_q = 0$, ($q = 5, 6$). With this choice for the $\bar{\Omega}^{(r)}$, $\tilde{U}_o^{(HS-)}$ can be expressed in terms of the three *incompressible* transversely isotropic invariants of $\bar{\sigma}$, namely, the deviatoric shear stress $\bar{\tau}_d$, the transverse shear stress $\bar{\tau}_p$ and the longitudinal shear stress $\bar{\tau}_n$ (see Appendix I). Furthermore, $\tilde{U}_o^{(HS-)}$ depends on these three invariants and the $\omega_q^{(r)}$ ($q = 1, \dots, 6$) only through the following $4n$ groups

$$(28) \quad \bar{\tau}_d(\omega_1^{(r)} - 2\omega_6^{(r)}), \quad \bar{\tau}_d(\omega_5^{(r)} - \omega_2^{(r)}), \quad \bar{\tau}_p\omega_3^{(r)}, \quad \text{and} \quad \bar{\tau}_n\omega_4^{(r)}.$$

However, following a procedure similar to the one described in Appendix II [see the discussion preceeding equations (II.8)] to eliminate the optimization constraints, and using the identity $\max_{x,y} \{f(x+y) + g(x)\} = \max_x \{f(x)\} + \max_x \{g(x)\}$, the number of the optimization variables can be further reduced to $2n$ variables. In terms of these variables, the lower bound for the effective energy function of the linear comparison composite becomes

$$(29) \quad \tilde{U}_o^{(HS-)}(\bar{\sigma}) = \min_{\substack{\omega^{(r)}, \bar{\omega}=1 \\ \eta^{(r)}, \bar{\eta}=1}} \left\{ \sum_{\substack{r=1 \\ r \neq s}}^n c^{(r)} \left[\frac{1}{2\mu_o^{(r)}} \left[(\tau_p^2 + \tau_n^2)(\omega^{(r)})^2 + \tau_d^2(\eta^{(r)})^2 \right] + \frac{1}{2\mu_o^{(s)}} (\tau_p^2 + \tau_n^2)(1 - \omega^{(r)})^2 \right] \right\}.$$

Upon substitution of (29) into (25), we arrive at following expression for the lower bound for the effective energy function of the nonlinear, incompressible fiber composite $\tilde{U}^{(HS-)}$, namely,

$$(30) \quad \tilde{U}^{(HS-)}(\bar{\sigma}) = \min_s \min_{\substack{\omega^{(r)}, \bar{\omega}=1 \\ \eta^{(r)}, \bar{\eta}=1}} \left\{ \sum_{\substack{r=1 \\ r \neq s}}^n c^{(r)} \psi^{(r)}(\tau_e^{(r)}) + c^{(s)} \psi^{(s)}(\tau_e^{(s)}) \right\},$$

where

$$(31) \quad \tau_e^{(r)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_n^2)(\omega^{(r)})^2 + (\bar{\tau}_d\eta^{(r)})^2}, \quad (r = 1, \dots, n; r \neq s),$$

$$\tau_e^{(s)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_n^2) \left[(\omega^{(s)})^2 + \sum_{i=1}^n \frac{c^{(i)}}{c^{(s)}} (1 - \omega^{(i)})^2 \right] + (\bar{\tau}_d\eta^{(s)})^2}.$$

Expression (30) was obtained from (25) by interchanging the order of the optimization operations over the variables $\mu_o^{(r)}$ and the $\omega^{(r)}$, $\eta^{(r)}$ variables, respectively. This is allowed by the Saddle Point Theorem (Rockafellar 1970, Corollary 37.3.1) since the functions $(-V^{(r)})$ are concave in the

variables $\mu_o^{(r)}$, while (29) is convex in the variables $\omega^{(r)}$ and $\eta^{(r)}$. We also made use of the equality (7), specialized to each of the phases. Finally, we note that there are n "branches" (one for each phase) to the solution of (30), and that the minimum over all the branches yields the desired lower bound.

Once the minimization problem (30) is solved, an estimate for the associated stress-strain relations may be obtained by simple differentiation with respect to $\bar{\sigma}$. However, from Appendix II, in the computation of the effective stress-strain relations, we may regard the optimization variables $\omega^{(r)}$ and $\eta^{(r)}$ as constants as far as derivatives with respect to $\bar{\sigma}$ are concerned. Thus, the expressions for the effective stress-strain relations may be written

$$(32) \quad \bar{\epsilon} = \sum_{\substack{r=1, \\ r \neq s}}^n \frac{c^{(r)}}{\hat{\tau}_e^{(r)}} \frac{d\psi^{(r)}}{d\tau_e^{(r)}} (\hat{\tau}_e^{(r)}) \left[(\hat{\omega}^{(r)})^2 \left(\bar{\tau}_p \frac{\partial \bar{\tau}_p}{\partial \bar{\sigma}} + \bar{\tau}_n \frac{\partial \bar{\tau}_n}{\partial \bar{\sigma}} \right) + (\hat{\eta}^{(r)})^2 \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right] + \dots$$

$$\dots + \frac{c^{(s)}}{\hat{\tau}_e^{(s)}} \frac{d\psi^{(s)}}{d\tau_e^{(s)}} (\hat{\tau}_e^{(s)}) \left[\left((\hat{\omega}^{(s)})^2 + \sum_{i=1}^n \frac{c^{(i)}}{c^{(s)}} (1 - \hat{\omega}^{(i)})^2 \right) \left(\bar{\tau}_p \frac{\partial \bar{\tau}_p}{\partial \bar{\sigma}} + \bar{\tau}_n \frac{\partial \bar{\tau}_n}{\partial \bar{\sigma}} \right) + (\hat{\eta}^{(s)})^2 \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right],$$

where $\hat{\omega}^{(r)}$ and $\hat{\eta}^{(r)}$ are the optimized values of the variables $\omega^{(r)}$ and $\eta^{(r)}$ (from 30), respectively. The s -phase in the above expression corresponds to the branch attaining the minimum in (30), and the expressions for $\hat{\tau}_e^{(r)} = \tau_e^{(r)}(\hat{\omega}^{(r)}, \hat{\eta}^{(r)})$, and $\hat{\tau}_e^{(s)} = \tau_e^{(s)}(\hat{\omega}^{(r)}, \hat{\eta}^{(r)})$ are obtained from (31).

The upper estimate for the effective energy function of the nonlinear incompressible fiber composite may be obtained in a similar manner. Thus, the expression for the upper estimate has precisely the same form as the lower bound (30), except that the minimum over all phases is replaced by a maximum. The corresponding estimate for the effective stress-strain relation may also be obtained by means of (32), where, in this case, the s -phase corresponds to the branch attaining the maximum in (30).

The representation (30) for the lower bound (or the upper estimate) involves minimization problems over $2n$ constrained variables. However, the number of optimization variables can be further reduced to $2(n-1)$ unconstrained variables (see Appendix II). For example, in case of a two-phase, incompressible fiber composite, the optimization constraint may be eliminated by

letting $\omega^{(1)} = 1 + c^{(2)}\omega$, $\omega^{(2)} = 1 - c^{(1)}\omega$, $\eta^{(1)} = 1 + c^{(2)}\eta$, and $\eta^{(2)} = 1 - c^{(1)}\eta$. In terms of the two unconstrained variables ω and η , the two branches of (30) are

$$(33) \quad \tilde{U}^{(n)}(\bar{\sigma}) = \min_{\omega, \eta} \left\{ c^{(1)} \psi^{(1)} \left(\sqrt{\left[(1 + c^{(2)}\omega)^2 + c^{(2)}\omega^2 \right] (\bar{\tau}_p^2 + \bar{\tau}_n^2) + (1 + c^{(2)}\eta)^2 \bar{\tau}_d^2} \right) + \dots \right. \\ \left. \dots + c^{(2)} \psi^{(2)} \left(\sqrt{\left[(1 - c^{(1)}\omega)^2 + c^{(1)}\omega^2 \right] (\bar{\tau}_p^2 + \bar{\tau}_n^2) + (1 - c^{(1)}\eta)^2 \bar{\tau}_d^2} \right) \right\},$$

and

$$\tilde{U}^{(n)}(\bar{\sigma}) = \min_{\omega, \eta} \left\{ c^{(1)} \psi^{(1)} \left(\sqrt{\left[(1 + c^{(2)}\omega)^2 + c^{(2)}\omega^2 \right] (\bar{\tau}_p^2 + \bar{\tau}_n^2) + (1 + c^{(2)}\eta)^2 \bar{\tau}_d^2} \right) + \dots \right. \\ \left. \dots + c^{(2)} \psi^{(2)} \left(\sqrt{\left[(1 - c^{(1)}\omega)^2 + c^{(1)}\omega^2 \right] (\bar{\tau}_p^2 + \bar{\tau}_n^2) + (1 - c^{(1)}\eta)^2 \bar{\tau}_d^2} \right) \right\}.$$

Then, the lower bound and the upper estimate are equal to the smallest and the largest of the two branches, respectively. The corresponding estimates for the effective stress-strain relations are obtained by making appropriate use of (32).

We note that lower bound expressions for the effective energy functions of *incompressible* nonlinear fiber composites have been obtained previously by Talbot and Willis (1991). These authors made use of the Talbot-Willis variational method, resulting in a different, more complicated, form for the bounds than the form presented here. We also note that expressions (30) for the lower bound, and the analogous expression for the upper estimate, were first derived by Ponte Castañeda (1992) by application of the variational principle (10). However, the derivation given here, in terms of the identity (III.2), is different, and can be generalized to the class of *compressible*, nonlinear fiber composites. This is accomplished in the next section. We also note that expressions (32) for the effective stress-strain relations of the fiber composite are presented here for the first time.

5. The compressible fiber-reinforced composites

With the insight gained in the previous section, we attempt in this section to obtain corresponding results for n -phase fiber composites with compressible, nonlinear, isotropic phases. Again, we will make use of the identity (III.2) in the expressions for the bounds of the linear comparison composite. Thus, expressions for the lower bound and the upper estimate for the

effective energy function of the nonlinear, compressible fiber composite may be obtained by following exactly the same steps that led from (25) to (30).

We begin by making use of the same choice for the optimization variables $\mathbf{\Omega}$ as in (27) to rewrite the expression for $\tilde{U}_o^{(hs-)}$ in terms of the four transversely isotropic invariants of $\bar{\mathbf{\sigma}}$: the in-plane hydrostatic stress $\bar{\sigma}_p$, the normal tensile stress $\bar{\sigma}_n$, the transverse shear stress $\bar{\tau}_p$ and the longitudinal shear stress $\bar{\tau}_n$ (see Appendix I). Next, we observe that the dependence of $\tilde{U}_o^{(hs-)}$ on these four invariants and on the six optimization variables (for each phase) is only through the following $4n$ groups:

$$(34) \quad (\bar{\sigma}_p \omega_1^{(r)} + \bar{\sigma}_n \omega_6^{(r)}), \quad (2\bar{\sigma}_p \omega_5^{(r)} + \bar{\sigma}_n \omega_2^{(r)}), \quad \bar{\tau}_p \omega_3^{(r)}, \quad \text{and} \quad \bar{\tau}_n \omega_4^{(r)}.$$

Therefore, following the same method as in the case of the incompressible composite, the $4n$ groups of (34) may be replaced by the $4n$ groups: $\bar{\sigma}_p \eta^{(r)}$, $\bar{\sigma}_n \phi^{(r)}$, $\bar{\tau}_p \omega^{(r)}$ and $\bar{\tau}_n \theta^{(r)}$, respectively. Consequently, the lower bound for the effective energy function of the linear comparison composite may be rewritten in the form

$$(35) \quad \tilde{U}_o^{(hs-)}(\bar{\mathbf{\sigma}}) = \min_{\Pi} \left\{ \sum_{r=1}^n c^{(r)} \left[\frac{1}{2\mu_o^{(r)}} \left[(\bar{\tau}_p \omega^{(r)})^2 + (\bar{\tau}_n \theta^{(r)})^2 + \frac{1}{3} (\bar{\sigma}_p \eta^{(r)} - \bar{\sigma}_n \phi^{(r)})^2 \right] + \dots \right. \right. \\ \left. \dots + \frac{1}{2\mu_o^{(+)}} \left[\bar{\sigma}_p^2 (1 - \eta^{(r)})^2 + \beta_o(\mu_o^{(+)}, \kappa_o^{(+)}) \bar{\tau}_p^2 (1 - \omega^{(r)})^2 + \bar{\tau}_n^2 (1 - \theta^{(r)})^2 \right] + \dots \right. \\ \left. \dots + \frac{1}{2\kappa_o^{(r)}} \left[\frac{2}{3} \bar{\sigma}_p \eta^{(r)} + \frac{1}{3} \bar{\sigma}_n \phi^{(r)} \right]^2 \right] \right\},$$

where

$$(36) \quad \beta_o(\mu, \kappa) = 1 + \frac{2\mu}{\kappa + \frac{1}{3}\mu},$$

and where $\Pi = \{ \eta^{(r)}, \phi^{(r)}, \omega^{(r)}, \theta^{(r)} | \bar{\eta} = 1, \bar{\phi} = 1, \bar{\omega} = 1, \bar{\theta} = 1 \}$ is the reduced set of (constrained) optimization variables. We note that the choice of this set is not unique, and that in some cases, as when $\bar{\sigma}_p = 0$ or $\bar{\sigma}_n = 0$, other choices may be preferred (see Section 6). This is because expression (35) becomes degenerate when $\bar{\sigma}_p = 0$ or $\bar{\sigma}_n = 0$.

With expression (35) for the linear lower bound, relation (21) leads to the following form for the nonlinear lower bound

$$(37) \quad \tilde{U}^{(HS-)}(\bar{\sigma}) = \min_{s_1, s_2} \min_{\Pi} \left\{ \sum_{r=1}^n c^{(r)} \psi^{(r)}(\tau_e^{(r)}, \sigma_m^{(r)}) + \Delta_o(\bar{\sigma}, \Pi) \right\},$$

where

$$(38) \quad \tau_e^{(r)} = \sqrt{(\bar{\tau}_p \omega^{(r)})^2 + (\bar{\tau}_n \theta^{(r)})^2 + \frac{1}{3}(\bar{\sigma}_p \eta^{(r)} - \bar{\sigma}_n \phi^{(r)})^2},$$

$$\sigma_m^{(r)} = \frac{2}{3} \bar{\sigma}_p \eta^{(r)} + \frac{1}{3} \bar{\sigma}_n \phi^{(r)},$$

and where

$$(39) \quad \Delta_o = \max_{\mu_o^{(s_1)}, \kappa_o^{(s_2)} \geq 0} \left\{ \sum_{i=1}^n \frac{c^{(i)}}{2\mu_o^{(s_1)}} \left[\bar{\sigma}_p^2 (1 - \eta^{(i)})^2 + \beta_o(\mu_o^{(s_1)}, \kappa_o^{(s_2)}) \bar{\tau}_p^2 (1 - \omega^{(i)})^2 + \bar{\tau}_n^2 (1 - \theta^{(i)})^2 \right] + \dots \right.$$

$$\dots + c^{(s_1)} \left[\frac{1}{2\mu_o^{(s_1)}} (\tau_e^{(s_1)})^2 + \max_{\tau_e} \left\{ \frac{1}{2\mu_o^{(s_1)}} \tau_e^2 - \psi^{(s_1)}(\tau_e, \sigma_m^{(s_1)}) \right\} \right] + \dots$$

$$\left. \dots + c^{(s_2)} \left[\frac{1}{2\kappa_o^{(s_2)}} (\sigma_m^{(s_2)})^2 + \max_{\sigma_m} \left\{ \frac{1}{2\kappa_o^{(s_2)}} \sigma_m^2 - \psi^{(s_2)}(\tau_e^{(s_2)}, \sigma_m) \right\} \right] \right\}.$$

Here, we have made use of the Saddle Point Theorem to interchange the order of the minimum over the set Π with the maximum over the comparison moduli $\mu_o^{(r)}$ and $\kappa_o^{(r)}$. We have also made use of relation (7) specialized to each of the phases to simplify the above expression. However, it should be noted that relation (7) cannot be used in (39) due to the coupling between the two optimization variables $\mu_o^{(s_1)}$ and $\kappa_o^{(s_2)}$. The representation (37) involves a minimization problem over $4n$ constrained variables, along with the intermediate four-dimensional optimization problem (39). However, the constraint can be easily embedded in (37) to reduce the dimension of the optimization problem to $4(n-1)$ (see Appendix II). Finally, we note that the problem (37) has n^2 branches (one for each possible combination of $\mu_o^{(s_1)}$ and $\kappa_o^{(s_2)}$, $s_1, s_2 = 1, \dots, n$), and that the lower bound is then obtained by taking the minimum over all these branches.

The upper estimate for the effective energy function of the compressible, nonlinear fiber composite may be obtained in a similar manner. Thus, the expression for the upper estimate has

precisely the same form as that for the lower bound (37), except that the minimum over all possible combinations of $\mu_o^{(s_1)}$ and $\kappa_o^{(s_2)}$ is replaced by a maximum.

On the face of it, the representation (37) for the lower bound (or the upper estimate) for the effective energy function of the nonlinear fiber composite does not appear to offer much of an advantage over the previous representation (21). However, in many practical applications, associated with special classes of fiber-reinforced composites, further simplification of the above representation is possible. For example, if a particular phase, say phase s , is stiffer (weaker) than the others, only one branch of the solution needs to be evaluated since, in this case, the choice $s_1 = s_2 = s$ leads to the lower bound (upper estimate). Further, we note that whenever the intermediate optimization problem (39) can be solved analytically, the representation (37) is preferable because it involves a minimization problem, in contrast with expression (21) which requires the solution of a minimax problem. From a computational point of view, this also eliminates the need for the iterative procedure associated with the evaluation of the functions $V^{(r)}$ in (21). To illustrate this point, let us examine the class of compressible fiber composites with one or more incompressible phases.

Thus, we consider a compressible, n -phase fiber composite, with at least one incompressible phase. Then, the minimum in (37) is given by the trivial choice $\kappa_o^{(s_2)} = \infty$, which enables the evaluation of the optimization problem (39). Thus, the lower bound reduces to the $3n$ -dimensional constrained minimization problem

$$(40) \quad \tilde{U}^{(ns-)}(\bar{\sigma}) = \min_s \min_{\substack{\omega^{(r)}, \bar{\omega}=1 \\ \phi^{(r)}, \bar{\phi}=1 \\ \eta^{(r)}, \bar{\eta}=1}} \left\{ \sum_{\substack{r=1 \\ r \neq s}}^n c^{(r)} \psi^{(r)}(\tau_e^{(r)}, \sigma_m^{(r)}) + c^{(s)} \psi^{(s)}(\tau_e^{(s)}, \sigma_m^{(s)}) \right\},$$

where

$$(41) \quad \tau_e^{(r)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_n^2)(\omega^{(r)})^2 + \frac{1}{3}(\bar{\sigma}_p \eta^{(r)} - \bar{\sigma}_n \phi^{(r)})^2}, \quad (r = 1, \dots, n; r \neq s),$$

$$\tau_e^{(s)} = \sqrt{(\bar{\tau}_p^2 + \bar{\tau}_n^2)(\omega^{(s)})^2 + \frac{1}{3}(\bar{\sigma}_p \eta^{(s)} - \bar{\sigma}_n \phi^{(s)})^2 + \sum_{i=1}^n \frac{c^{(i)}}{c^{(s)}} [\bar{\sigma}_p^2 (1 - \eta^{(i)})^2 + (\bar{\tau}_p^2 + \bar{\tau}_n^2)(1 - \omega^{(i)})^2]},$$

and

$$\sigma_m^{(r)} = \frac{2}{3} \bar{\sigma}_p \eta^{(r)} + \frac{1}{3} \bar{\sigma}_n \phi^{(r)}, \quad (r = 1, \dots, n).$$

Within this class of composites, of particular interest is the sub-class of hollow-fibers composites, for which the above expression for the lower bound reduces to a simple, explicit result. Thus, we take the complementary energy-density function of the matrix (phase 1) to be of the form $U^{(1)}(\boldsymbol{\sigma}) = \psi(\tau_e)$. Correspondingly, the complementary energy-density function of the cylindrical voids (phase 2) is given by $U^{(2)}(\boldsymbol{\sigma}) = \infty$ if $\boldsymbol{\sigma} \neq \mathbf{0}$, and 0 otherwise. The minimum in (40) is trivially attained by the choice $s = 1$ and $\omega^{(2)} = \phi^{(2)} = \eta^{(2)} = 0$. The values of the variables $\omega^{(1)}$, $\phi^{(1)}$ and $\eta^{(1)}$ then follows from the optimization constraint. Substitution of these values into (40), leads to the following expression for the lower bound of the hollow-fibers composites

$$(42) \quad \tilde{U}^{(hs-)}(\bar{\boldsymbol{\sigma}}) = (1-c) \psi \left(\frac{1}{(1-c)} \sqrt{(1+c)(\bar{\tau}_p^2 + \bar{\tau}_n^2) + \frac{1}{3}(\bar{\sigma}_p - \bar{\sigma}_n)^2 + c\bar{\sigma}_p^2} \right),$$

where $c = c^{(2)}$ is the volume fraction of the voids. The corresponding estimate for the stress-strain relation may be easily derived with the help of (1.12). We note that when $\psi(\tau_e) = a(\tau_e)^{n+1}$, where a is a non-negative constant and $n > 1$, the above result reduces to the lower bound derived independently by Suquet (1992) for power-law materials containing cylindrical voids.

Another class of composites, of great practical significance, for which the representation of the lower bound may be simplified, by explicit evaluation of (39), is the class of n -phase fiber composites where the stiffest phase is linear. In the following section, we are concerned with the special case of two-phase, fiber-reinforced composites of this type.

6. Application to metal-matrix composites

Among the various classes of fiber-reinforced composites, metal-matrix composites is one of the most common classes. In this section, we restrict our attention to this important class of composites, which are made up of ductile matrices reinforced by stiffer, linear-elastic fibers. Since the plastic strains in the metal phase are independent of the hydrostatic stresses, we assume that the behavior of the matrix (phase 1) is governed by a complementary energy-density function of the form

$$(43) \quad \psi^{(1)}(\tau_e, \sigma_m) = \varphi(\tau_e) + \frac{1}{2\kappa^{(1)}} \sigma_m^2,$$

where the function φ is nonnegative and satisfies the strong convexity assumption described in Section 2, and where $\kappa^{(1)}$ is the bulk modulus. Further, to account for the initial linear-elastic behavior of the metal phase, we assume that $\varphi(\tau_e) = \frac{1}{2\mu^{(1)}} \tau_e^2 + f(\tau_e)$, where $\mu^{(1)}$ is the elastic shear modulus. On the other hand, the behavior of the fiber material (phase 2) is governed by the quadratic complementary energy-density function

$$(44) \quad \psi^{(2)}(\tau_e, \sigma_m) = \frac{1}{2\mu^{(2)}} \tau_e^2 + \frac{1}{2\kappa^{(2)}} \sigma_m^2,$$

where $\mu^{(2)}$ and $\kappa^{(2)}$ are the corresponding shear and bulk moduli, respectively. Finally, to enforce the assumption that phase 2 is stiffer than phase 1, we let $\mu^{(2)} > \mu^{(1)}$ and $\kappa^{(2)} > \kappa^{(1)}$.

We begin by considering the lower bound $\tilde{U}^{(HS-)}$. As mentioned in the previous section, since phase 2 is linear and stiffer than phase 1, the intermediate optimization (39) may be evaluated explicitly. Further, because of the particular choice for the energy-density functions $\psi^{(1)}$ and $\psi^{(2)}$, a different choice for the reduced set of optimization variables in (34) leads to additional simplification of the problem. Thus, with the new choice for the optimization variables, the lower bound for the effective energy function of the composite may be given in terms of the following 3-dimensional minimization problem

$$(45) \quad \tilde{U}^{(HS-)}(\bar{\sigma}) = \min_{\eta, \omega, \theta} \left\{ c^{(1)} \left[\varphi(\tau_e^{(1)}) + \frac{1}{2\kappa^{(1)}} (\sigma_m^{(1)})^2 \right] + c^{(2)} \left[\frac{1}{2\mu^{(2)}} (\tau_e^{(2)})^2 + \frac{1}{2\kappa^{(2)}} (\sigma_m^{(2)})^2 \right] \right\},$$

where

$$(46) \quad \begin{aligned} (\tau_e^{(1)})^2 &= \bar{\tau}_p^2 (1 + c^{(2)} \omega)^2 + \bar{\tau}_n^2 (1 + c^{(2)} \theta)^2 + \frac{1}{3} (\bar{\sigma}_p - \bar{\sigma}_n)^2 (1 + c^{(2)} \eta)^2, \\ (\tau_e^{(2)})^2 &= \bar{\tau}_p^2 \left[(1 - c^{(1)} \omega)^2 + c^{(1)} \beta_o (\mu_o^{(2)}, \kappa_o^{(2)}) \omega^2 \right] + \bar{\tau}_n^2 \left[(1 - c^{(1)} \theta)^2 + c^{(1)} \theta^2 \right] + \dots \\ &\quad \dots + \frac{1}{3} (\bar{\sigma}_p - \bar{\sigma}_n)^2 (1 - c^{(1)} \eta)^2 + c^{(1)} \left[\frac{1}{3} \bar{\sigma}_p (2\phi + \eta) + \frac{1}{3} \bar{\sigma}_n (\phi - \eta) \right]^2, \\ \sigma_m^{(1)} &= \left(\frac{2}{3} \bar{\sigma}_p + \frac{1}{3} \bar{\sigma}_n \right) (1 + c^{(2)} \phi), \\ \sigma_m^{(2)} &= \left(\frac{2}{3} \bar{\sigma}_p + \frac{1}{3} \bar{\sigma}_n \right) (1 - c^{(1)} \phi). \end{aligned}$$

In the above expression, β_o is given by relation (36) and

$$(47) \quad \phi = \frac{\bar{\sigma}_p \left(\frac{2}{\kappa^{(2)}} - \frac{2}{\kappa^{(1)}} - \frac{\eta}{\mu^{(2)}} \right) + \bar{\sigma}_n \left(\frac{1}{\kappa^{(1)}} - \frac{1}{\kappa^{(2)}} + \frac{\eta}{\mu^{(2)}} \right)}{(2\bar{\sigma}_p + \bar{\sigma}_n) \left(\frac{c^{(2)}}{\kappa^{(1)}} + \frac{c^{(1)}}{\kappa^{(2)}} + \frac{1}{\mu^{(2)}} \right)}.$$

Once the minimization problem (45) is solved, the corresponding estimate for the effective stress-strain relation may be obtained by following the same procedure followed as for the incompressible fiber composite. Thus, the estimate for the stress-strain relation may be written in the form

$$(48) \quad \bar{\epsilon} = \frac{\partial}{\partial \bar{\sigma}} \left\{ c^{(1)} \left[\varphi(\hat{\tau}_e^{(1)}) + \frac{1}{2\kappa^{(1)}} (\hat{\sigma}_m^{(1)})^2 \right] + c^{(2)} \left[\frac{1}{2\mu^{(2)}} (\hat{\tau}_e^{(2)})^2 + \frac{1}{2\kappa^{(2)}} (\hat{\sigma}_m^{(2)})^2 \right] \right\},$$

where $\hat{\tau}_e^{(1)}$, $\hat{\tau}_e^{(2)}$, $\hat{\sigma}_m^{(1)}$ and $\hat{\sigma}_m^{(2)}$ are evaluated from (46) at the optimal values of η , ω and θ . Further, in (48), the derivatives with respect to $\bar{\sigma}$ are evaluated with η , ω and θ fixed. We emphasize that due to the different choice of the optimization variables, the above representation for the lower bound is subject to the restrictions $(2\bar{\sigma}_p + \bar{\sigma}_n) \neq 0$ and $(\bar{\sigma}_p - \bar{\sigma}_n) \neq 0$ [instead of the restrictions $\bar{\sigma}_p \neq 0$ and $\bar{\sigma}_n \neq 0$, for the representation (37)]. For consistency, we will make use of the same set of optimization variables in the following representation for the upper estimate (although, in this case, the number of the optimization variables cannot be reduced).

The evaluation of the upper estimate is more complicated since in this case the intermediate optimization problem (39) can not be solved explicitly. However, if the ratio of the initial shear modulus to the bulk modulus of the weaker, matrix phase is small (i.e., $\mu_o^{(1)}/\kappa_o^{(1)} \ll 1$), great simplification may be achieved by means of the following approximation for β_o , namely,

$$(49) \quad \beta_o(\mu_o^{(1)}, \kappa_o^{(1)}) = 1 + 2 \frac{\mu_o^{(1)}}{\kappa_o^{(1)}} + o\left(\frac{\mu_o^{(1)}}{\kappa_o^{(1)}}\right)^2.$$

Then, the estimate for the upper bound for the effective energy function reduces to

$$(50) \quad \tilde{U}^{(HS+)}(\bar{\sigma}) \equiv \min_{\eta, \phi, \omega, \theta} \left\{ c^{(1)} \left[\varphi(\tau_e^{(1)}) + \frac{1}{2\kappa^{(1)}} (\sigma_m^{(1)})^2 \right] + c^{(2)} \left[\frac{1}{2\mu^{(2)}} (\tau_e^{(2)})^2 + \frac{1}{2\kappa^{(2)}} (\sigma_m^{(2)})^2 \right] \right\},$$

where

$$(51) \quad \begin{aligned} (\tau_e^{(1)})^2 &= \bar{\tau}_p^2 \left[(1 + c^{(2)}\omega)^2 + c^{(2)}\omega^2 \right] + \bar{\tau}_n^2 \left[(1 + c^{(2)}\theta)^2 + c^{(2)}\theta^2 \right] + \dots \\ &\quad \dots + \frac{1}{3} (\bar{\sigma}_p - \bar{\sigma}_n)^2 (1 + c^{(2)}\eta)^2 + c^{(2)} \left[\frac{1}{3} \bar{\sigma}_p (2\phi + \eta) + \frac{1}{3} \bar{\sigma}_n (\phi - \eta) \right]^2, \\ (\tau_e^{(2)})^2 &= \bar{\tau}_p^2 (1 - c^{(1)}\omega)^2 + \bar{\tau}_n^2 (1 - c^{(1)}\theta)^2 + \frac{1}{3} (\bar{\sigma}_p - \bar{\sigma}_n)^2 (1 - c^{(1)}\eta)^2, \\ (\sigma_m^{(1)})^2 &= \left(\frac{2}{3} \bar{\sigma}_p + \frac{1}{3} \bar{\sigma}_n \right)^2 (1 + c^{(2)}\phi)^2 + 2c^{(1)} \bar{\tau}_p^2 \omega^2, \\ \sigma_m^{(2)} &= \left(\frac{2}{3} \bar{\sigma}_p + \frac{1}{3} \bar{\sigma}_n \right) (1 - c^{(1)}\phi). \end{aligned}$$

We recall that (50) is subject to the restrictions $(2\bar{\sigma}_p + \bar{\sigma}_n) \neq 0$ and $(\bar{\sigma}_p - \bar{\sigma}_n) \neq 0$. Also, fortunately, the above approximation holds for most metal matrix composites. For example, when $\mu^{(1)}/\kappa^{(1)} < 0.45$ (or in terms of the associated Poisson's ratio, $\nu^{(1)} > 0.3$), the maximum difference between (50) and the exact solution of the expression for $\tilde{U}^{(hs*)}$ is about 2% (see also Figure 3). It should also be noted that when $\bar{\tau}_p = 0$ the two forms are equal.

The associated expressions for the stress-strain relations may be evaluated from the upper estimate for the effective energy function in the usual way. Thus, the expressions for the effective stress-strain relations have the same form as (48), but with $\hat{\tau}_e^{(1)}$, $\hat{\tau}_e^{(2)}$, $\hat{\sigma}_m^{(1)}$ and $\hat{\sigma}_m^{(2)}$ evaluated from (51) at the optimal values of the variables η , ϕ , ω and θ [from (50)].

To conclude this section, we note that the lower bound (45) is actually an optimal bound (i.e., no bound that is better can be given for this class of composites). This can be shown by following arguments analogous to those given by Ponte Castañeda (1991b) in the context of statistically isotropic, two-phase, nonlinear, composites with one linear phase. From a more practical point of view, however, the above results may be used as estimates for the effective energy functions of two-phase composites with nonlinear fibers and a linear matrix (see Section 3). Thus, the lower bound (45), together with the corresponding estimate for the stress-strain relations, serve as estimates for the behavior of composites with a linear-elastic matrix weakened by softer, ductile fibers. Alternatively, the upper estimate (50), together with the corresponding estimate for the stress-strain relations, serve as estimates for the behavior of composites with a soft, ductile matrix reinforced by stiffer, linear-elastic fibers.

We note that the microstructure associated with the upper estimates (for the effective energy functions) corresponds to the that of metal-matrix composites. This suggests that the predictions for the effective stress-strain relation obtained from the upper estimate $\tilde{U}^{(hs*)}$ may be used as estimates for the behavior of metal-matrix composites. Indeed, the study of the effective behavior of an aluminum-matrix composite, that is carried out in the next section, confirms this expectation.

7. Application to an aluminum-matrix composite reinforced with boron fibers

In this section, we specialize the results of the previous section to the case of a nonlinear, compressible, fiber composite made up of an aluminum-matrix reinforced with boron fibers. The effective behavior of the composite is presented in terms of relations between the four transversely isotropic stress modes and the corresponding four strain modes (see Appendix I). These relations show the overall response of the anisotropic composite to different loading modes and reveal the coupling among the different modes.

Aluminum (phase 1) is a ductile material with a uniaxial stress-strain curve that can be approximated by a "linear-plus-power" law (see Figure 2 in Adams 1970). Accordingly, we assume the following form for the energy-density function of the aluminum matrix,

$$(52) \quad \psi^{(1)}(\tau_e, \sigma_m) = \varepsilon_o \int_0^{\sqrt{3}\tau_e} \left[\frac{s}{\sigma_o} + \left(\frac{s}{\sigma_o} - \frac{\sigma_y}{\sigma_o} \right)^n H(s - \sigma_y) \right] ds + \frac{1}{2\kappa^{(1)}} \sigma_m^2.$$

Here, H is the unit step function (equal to 0 when $s \leq \sigma_y$, or to 1 otherwise) and ε_o and σ_o are the strain and stress normalization factors, respectively, such that $\sigma_o/\varepsilon_o = 3\mu^{(1)}$, with $\mu^{(1)}$ denoting the elastic shear modulus. Also, $\kappa^{(1)}$ is the bulk modulus and σ_y is the yield stress in tension. Boron (phase 2) is a brittle material that behaves linearly up to failure. Its energy-density function is given by (44), with $\mu^{(2)}$ and $\kappa^{(2)}$ denoting the shear and bulk modulus of boron, respectively.

The lower bound and the upper estimate for the effective energy function of the composite may then be represented in dimensionless form via the relation

$$(53) \quad \frac{\tilde{U}^{(ns+)}(\bar{\sigma})}{\sigma_o \varepsilon_o} = G^{(+)} \left\{ \frac{\bar{\sigma}_p}{\sigma_o}, \frac{\bar{\sigma}_n}{\sigma_o}, \frac{\bar{\tau}_p}{\sigma_o}, \frac{\bar{\tau}_n}{\sigma_o}, \frac{\sigma_y}{\sigma_o}, \frac{\mu^{(2)}}{\mu^{(1)}}, \nu^{(1)}, \nu^{(2)}, n, c^{(2)} \right\},$$

where $G^{(-)}$ and $G^{(+)}$ are obtained from (45) and (50), respectively. The Poisson's ratios of the two phases $\nu^{(1)}$ and $\nu^{(2)}$ are defined by

$$(54) \quad \nu^{(r)} = \frac{3\kappa^{(r)} - 2\mu^{(r)}}{6\kappa^{(r)} + 2\mu^{(r)}} \quad (r = 1, 2).$$

The corresponding estimates for the four transversely isotropic strain modes are then determined by making use of (48) along with (I.5).

The numerical values for the six parameters in (54) are chosen so that a comparison with the corresponding experimental results of Adams (1970) may be carried out. Thus, the properties of the aluminum matrix are taken from Figure 2 in Adams (1970), which presents the uniaxial stress-strain curve for the aluminum. Similarly, the numerical values for the elastic constants of the boron fibers, as well as their volume fraction, were also taken from the same reference. These parameters are

$$\frac{\sigma_y}{\sigma_o} = 14.9, \quad \frac{\mu^{(2)}}{\mu^{(1)}} = 7.47, \quad \nu^{(1)} = 0.32, \quad \nu^{(2)} = 0.20, \quad n = 1.82 \text{ and } c^{(2)} = 0.34,$$

where the normalization factors are $\sigma_o = 5.89 \text{ MPa}$ and $\epsilon_o = 9.29 \times 10^{-5}$.

Results for the different loading modes of the nonlinear fiber composites, under several loading combinations, are given in Figures 2, 4, 5 and 6. To highlight the effect of nonlinearity, results are also given in the form of short-dash curves for linear fiber composites with the same elastic properties as the nonlinear fiber composite. Thus, the phases of these linear *reference* composites are identical to those of the nonlinear composite with the only difference that σ_y is taken to be unbounded for phase 1.

Figure 2 shows a plot of the normalized transverse shear stress $\bar{\tau}_p/\tau_y$ versus the normalized transverse shear strain $\bar{\gamma}_p/\gamma_y$ for three different loading combinations. Here, $\tau_y = \sigma_y/\sqrt{3}$ and $\gamma_y = \tau_y/2\mu^{(1)}$ denote the yield stress and strain in shear of the aluminum matrix, respectively. The continuous curves correspond to estimates derived from the lower bound for the effective energy function, while the long-dash curves correspond to estimates derived from the upper estimate for the effective energy function. In all three loading combinations, $\bar{\sigma}_p = 0$. The curves denoted by $\bar{\sigma}_n/\bar{\tau}_p = \bar{\tau}_n/\bar{\tau}_p = 0$ correspond to a pure transverse shear load. Initially, the behaviors of these two stress-strain curves for the nonlinear composites are the same as those of the corresponding curves for the reference linear composites (short-dash lines). However, the predictions for the effective yield points of the two types of estimates are different, with both the yield stresses and the yield strains being larger for the estimate associated with the lower bound on the energy.

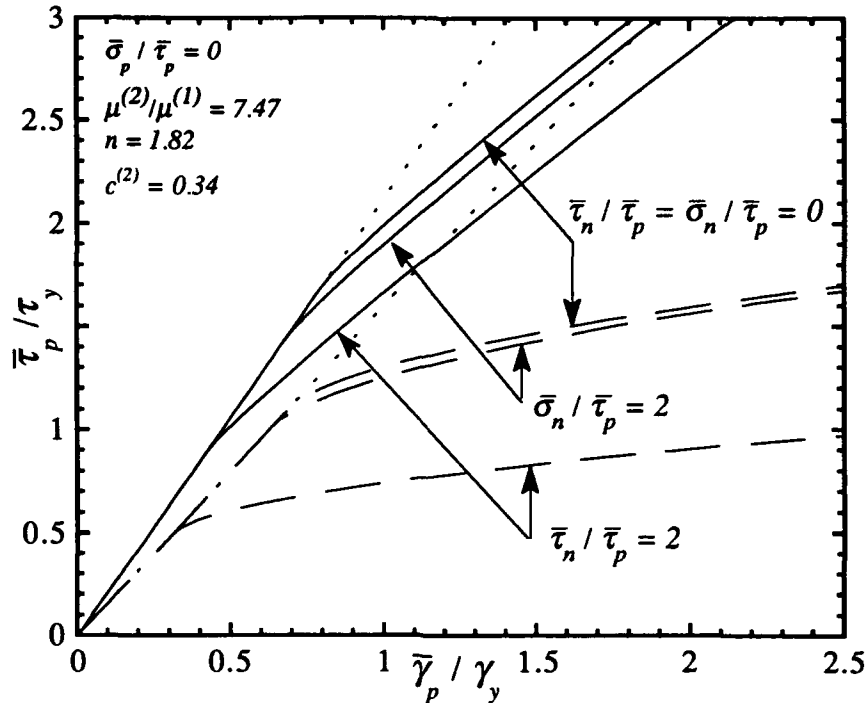


Fig. 2. Lower estimates (continuous curves) and upper estimates (dash curves) for the relations between the transverse shear stress $\bar{\tau}_p$ and the composite, as well as for the reference li combinations: $\bar{\sigma}_n / \bar{\tau}_p = \bar{\tau}_n / \bar{\tau}_p = 0$, $\bar{\tau}_n / \bar{\tau}_p$, $\bar{\sigma}_n / \bar{\tau}_p = 2$, $\bar{\tau}_n / \bar{\tau}_p = 2$ for all cases.

The effective stress-strain relations $\bar{\sigma}_p / \bar{\tau}_p$ and $\bar{\tau}_n / \bar{\tau}_p$ show post-yield behavior with almost linear hardening with $\bar{\gamma}_p / \gamma_y$.

$$(55) \quad \frac{2c^{(2)}\bar{\tau}_p}{(2-c^{(2)})\kappa}$$

The corresponding stress-strain relation for the post-yield behavior with power hardening, where the growth of the stress is proportional to $(\bar{\tau}_n / \tau_y)^n$ for large $\bar{\tau}_n$.

As discussed in the previous section, the stress-strain relations derived from the upper estimate for the effective energy function may be used as estimates for the behavior of composites with a ductile matrix phase and linear-elastic fibers. In this context, it is interesting to compare our results with corresponding predictions of Hashin (1980) (see also Dvorak and Bahei-El-Din 1987), who argues, on physical grounds, the existence of two distinct failure modes for fiber-reinforced

composites with a weaker matrix phase: a *fiber* mode in which the composite fails due to fiber failure in tension or compression along the fibers, and a *matrix* mode in which the matrix fails in shear transverse to, or along, the fibers. According to this model, when the composite is subjected to transverse shear loads, its yield stress and post-yield behavior are dominated by the behavior of the ductile matrix. We note that the prediction for the behavior of the composite, from the stress-strain curve associated with the upper energy estimate, agrees with this model. Thus, the effective yield stress is only slightly higher than the yield stress of the matrix, and the post-yield behavior is dominated by the behavior of the ductile matrix as well.

The coupling between the shear modes $\bar{\tau}_n$ and $\bar{\tau}_p$ is demonstrated by the two curves for the proportional loading path $\bar{\tau}_n/\bar{\tau}_p = 2$, and the coupling between $\bar{\sigma}_n$ and $\bar{\tau}_p$ by the two curves for the proportional loading path $\bar{\sigma}_n/\bar{\tau}_p = 2$. We note that the effect of a combination of transverse shear loads with any of the other modes is to saturate the linear range of phase 1, accelerating the post-yield behavior of the composite. Thus, the resistance of the composite to transverse shear loads is reduced with the growth of any of the other loading modes. We note that this softening effect is more marked for the stress-strain curves obtained from the upper energy estimate. Also, the effect of the superimposed longitudinal shear $\bar{\tau}_n$ on the transverse shear mode $\bar{\tau}_p$ is more pronounced than that of the superimposed uniaxial tensile stress $\bar{\sigma}_n$.

To illustrate the consequences of this softening phenomenon, let us consider a cylindrical fiber composite bar, subjected to a uniaxial tension load in the fiber direction combined with a torque aligned with the symmetry axis. The stress field that develops within this bar, on the microscopic level, may be represented by a combination of a uniaxial tensile stress $\bar{\sigma}_n$ and a transverse shear stress $\bar{\tau}_p$. Thus, our results predict that any increment in the uniaxial tensile stress (with fixed transverse shear stress) will cause additional growth of the resultant transverse shear strain. The overall contributions of the shear strain increments (within the composite) is to increase the twisting angle of the bar. Thus, on the macroscopic level, we would observe growth of the twisting angle in response to an increment in the tensile load (with a fixed applied torque).

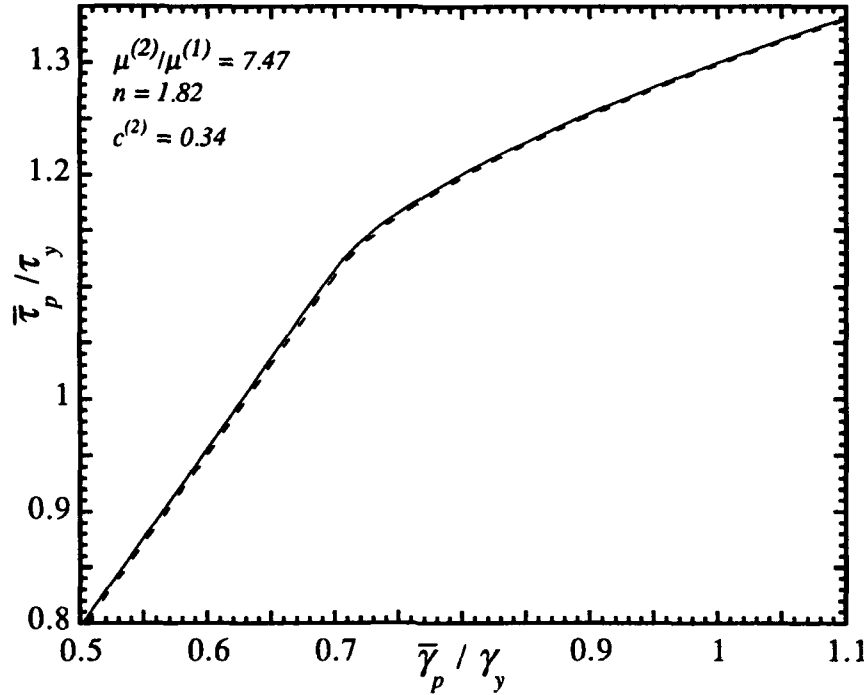


Fig. 3. Comparison of the upper estimates for the relations between the transverse shear stress $\bar{\tau}_p$ and the corresponding shear strain $\bar{\gamma}_p$ (continuous curve) with the corresponding approximation (50) (short-dash curve), based on neglecting terms of order $(\mu_o^{(1)}/\kappa_o^{(1)})^2$.

Figure 3 shows a plot of the normalized shear stress $\bar{\tau}_p/\tau_y$ versus the normalized shear strain $\bar{\gamma}_p/\gamma_y$, in the vicinity of the predicted yield point, when all other stress modes vanish. Here, we compare the predictions for $\bar{\gamma}_p$ obtained from the exact solution for the minimax problem for $\tilde{U}^{(ns*)}$ (continuous curves) with the corresponding approximation (50) (short-dash curve), based on neglecting terms of order $(\mu_o^{(1)}/\kappa_o^{(1)})^2$. The curves are very close, with a maximum error of about 0.5%. We note that the maximum error between the two curves always occurs near the predicted yield points.

Figure 4 shows a plot of the normalized longitudinal shear stress $\bar{\tau}_n/\tau_y$ versus the normalized longitudinal shear strain $\bar{\gamma}_n/\gamma_y$ for three different loading combinations. The continuous curves correspond to the estimates derived from the lower bound for the effective energy function, while the long-dash curves correspond to the estimates derived from the upper estimate for the effective energy function. For all three loading combinations, $\bar{\sigma}_p = 0$.

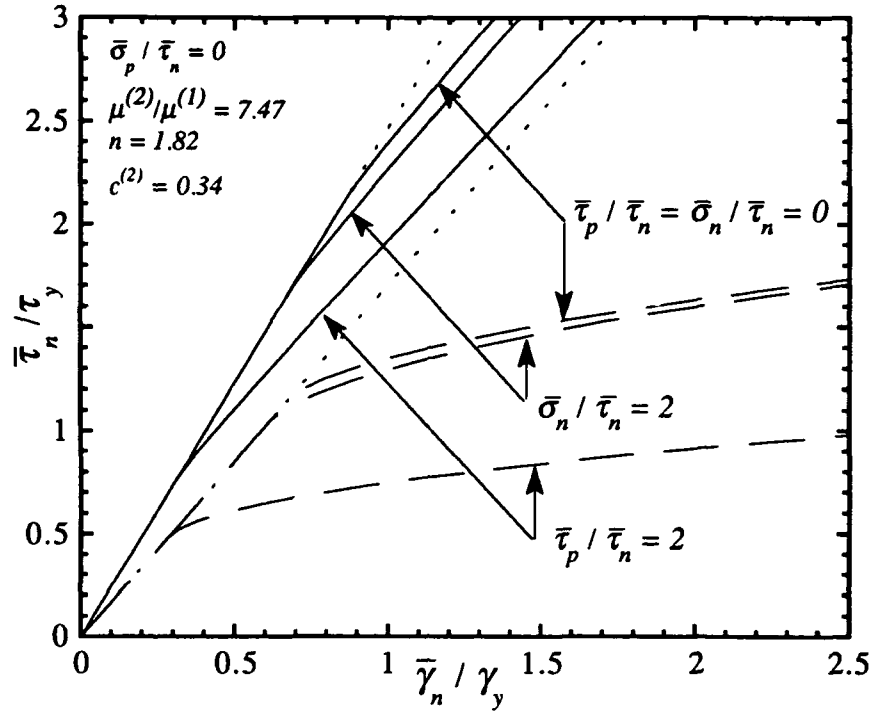


Fig. 4. Lower estimates (continuous curves) and upper estimates (long-dash curves) for the relations between the longitudinal shear stress $\bar{\tau}_n$ and the corresponding shear strain $\bar{\gamma}_n$ for the nonlinear fiber composites, as well as for the reference linear fiber composites (short-dash lines), for three different loading combinations: $\bar{\sigma}_n/\bar{\tau}_n = \bar{\tau}_p/\bar{\tau}_n = 0$, $\bar{\tau}_p/\bar{\tau}_n = 2$ and $\bar{\sigma}_n/\bar{\tau}_n = 2$. Here, $\bar{\sigma}_p = 0$ for all cases.

The curves denoted by $\bar{\tau}_p/\bar{\tau}_n = \bar{\sigma}_n/\bar{\tau}_n = 0$ correspond to a pure longitudinal shear load along the fibers. We observe that the predictions for the yield stresses, the yield strains and the post-yield behaviors obtained from the upper energy estimates are very similar to the predictions of the corresponding curve in Figure 2 (denoted by $\bar{\sigma}_n/\bar{\tau}_p = \bar{\tau}_n/\bar{\tau}_p = 0$). This similarity is anticipated on grounds of our previous observation concerning the interpretation of the upper energy estimates as estimates for ductile-matrix composites. Thus, when the composite is subjected to shear loads along the fibers, it is expected that its yield and post-yield behaviors will be dominated by the corresponding behaviors of the aluminum matrix, and hence, the similarity between the two curves.

On the other hand, the curves associated with the lower bounds, in Figures 2 and 4, are not as close. Although the (nearly) linear-hardening, post-yielding behavior is common for both cases, there is a significant difference between the asymptotic slopes of the curves as the stress increases. In Figure 4, the slope of the longitudinal shear stress-strain curve approaches the value $2c^{(2)}\mu^{(2)}/(2 - c^{(2)})$, whereas the corresponding slope for the transverse shear stress curve, in Figure 2, approaches a lower value which is given by relation (55).

The coupling between the shear modes $\bar{\tau}_n$ and $\bar{\tau}_p$ is demonstrated by two curves for the proportional loading path $\bar{\tau}_p/\bar{\tau}_n = 2$, whereas the coupling between $\bar{\sigma}_n$ and $\bar{\tau}_p$ is demonstrated by the two curves for the proportional loading path $\bar{\sigma}_n/\bar{\tau}_n = 2$. We observe that the effect of adding any of the other modes to the longitudinal shear mode is to saturate earlier the linear range of phase 1, accelerating the post-yield behavior of the composite. Thus, in a manner similar to the previous case (Figure 2), the resistance of the composite to longitudinal shear loads is reduced with the growth of any of the other loading modes.

Figure 5 shows plots of the normalized tensile stress $\bar{\sigma}_n/\sigma_y$ versus the normalized tensile strain $\bar{\epsilon}_n/\epsilon_y$ for three different loading combinations. Here, $\epsilon_y = \sigma_y/2\mu^{(1)}(1 + \nu^{(1)})$ denotes the yield strain of the aluminum matrix under uniaxial tensile load. The continuous curves correspond to the estimates derived from the lower bound for the effective energy function, while the long-dash curves correspond to the estimates derived from the upper estimate for the effective energy function. For all three loading combinations, $\bar{\tau}_p = 0$.

The curves denoted by $\bar{\sigma}_p/\bar{\sigma}_n = \bar{\tau}_n/\bar{\sigma}_n = 0$ correspond to a uniaxial tensile load in the fiber direction. Initially, the behaviors of the two curves are the same as those for the reference linear composite (short-dash lines), until phase 1 yields. The predictions for the effective yield stress and strain are almost the same, and after yielding, according to both estimates the plastic hardening of the composite is nearly linear. As discussed by Hashin (1980), this post-yield behavior is expected on physical grounds since the stiff phase dominates the behavior of the composite in tension (or compression) along the fibers.

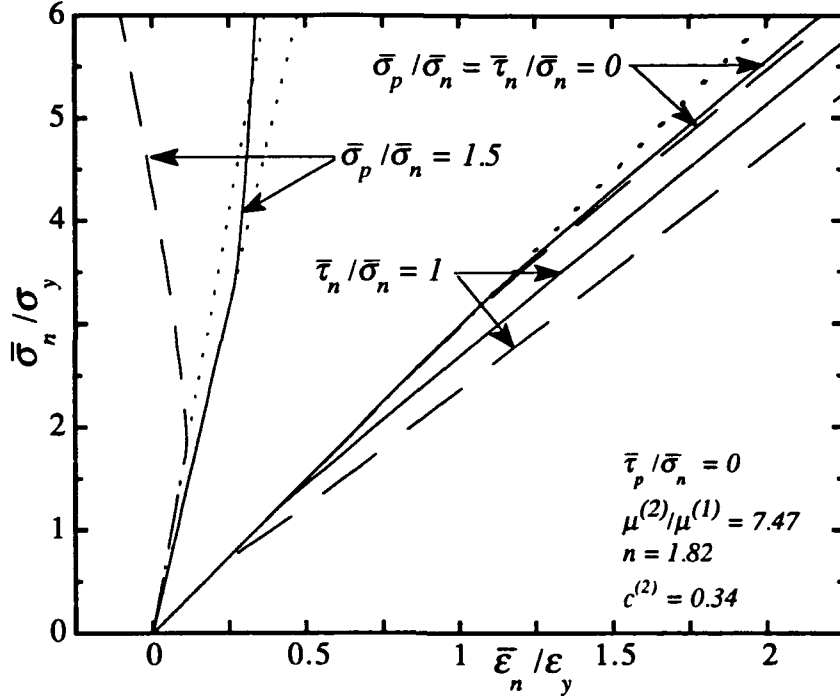


Fig. 5. Lower estimates (continuous curves) and upper estimates (long-dash curves) for the relations between the normal tensile stress $\bar{\sigma}_n$ and the corresponding strain $\bar{\epsilon}_n$ for the nonlinear fiber composites, as well as for the reference linear fiber composites (short-dash lines), for three different loading combinations: $\bar{\sigma}_p / \bar{\sigma}_n = \bar{\tau}_n / \bar{\sigma}_n = 0$, $\bar{\sigma}_p / \bar{\sigma}_n = 1.5$ and $\bar{\tau}_n / \bar{\sigma}_n = 1$. Here, $\bar{\tau}_p = 0$ for all cases.

The coupling between the two dilatational modes is demonstrated by the two curves for the proportional loading path $\bar{\sigma}_p / \bar{\sigma}_n = 1.5$. Thus, we see that the superimposed $\bar{\sigma}_p$ leads to significant stiffening of the composite. We observe that the two curves (for $\bar{\sigma}_p / \bar{\sigma}_n = 1.5$) are initially fairly close to each other (and behave the same way as the corresponding curves of the reference linear composites). However, after the yielding of phase 1, the predictions based on the upper energy estimate are markedly different from the corresponding predictions from the lower energy bound. Thus, according to the lower energy bound, after yielding, there is only little growth of the tensile strain; while according to the upper energy estimate, the normal tensile strain actually *decreases*. This interesting behavior is due to the plastic incompressibility of the aluminum phase. Since the plastic strains in phase 1 are proportional to the stress deviators, and under the above combination of loads, the normal component of the stress deviator (in the fiber direction) is negative, the plastic

part of the normal tensile strain decreases. According to the lower bound, the magnitudes of the elastic and the plastic parts of the tensile strain are almost the same, and hence $\bar{\epsilon}_n$ (the sum of the two parts) is almost fixed. However, according to the upper estimate, under these loading conditions the magnitude of the plastic part of the tensile strain is larger than that of the elastic part, and hence $\bar{\epsilon}_n$ decreases.

The coupling between the normal tensile stress and the longitudinal shear stress $\bar{\tau}_n$ is demonstrated by the two curves corresponding to the proportional loading path $\bar{\tau}_n/\bar{\sigma}_n = 1$. The effect of increasing the shear load $\bar{\tau}_n$ is to saturate the linear range of phase 1, hence accelerating the post-yield behavior of the composite. We note that the difference between the curves for the lower bound and the upper estimate are relatively small.

Figure 6 shows plots of the normalized in-plane hydrostatic stress $\bar{\sigma}_p/\sigma_y$ versus the normalized in-plane hydrostatic strain $\bar{\epsilon}_p/\epsilon_y$ for three different loading combinations. The continuous curves correspond to the estimates derived from the lower bound for the effective energy function, while the long-dash curves to the estimates derived from the upper estimate for the effective energy function. For all three loading combinations, $\bar{\tau}_p = 0$.

The curves denoted by $\bar{\sigma}_n/\bar{\sigma}_p = \bar{\tau}_n/\bar{\sigma}_p = 0$ correspond to a pure, plane hydrostatic load in the transverse direction. Initially, the behaviors of the two curves are the same as those of the reference linear composite (short-dash lines), until the yielding of phase 1. According to both types of estimates, the plastic hardening of the composites is almost linear. This suggests that the in-plane hydrostatic strains are governed by the stiff linear phase. In the case of normal tensile loads (Figure 5), it is easy to visualize that the tensile strains along the fibers should be controlled by the stiffer phase; however, that the same behavior should be observed for the in-plane hydrostatic strains is perhaps less intuitive. The reason is related to the Poisson effect. Thus, when the fiber composite is subjected to in-plane hydrostatic loads, tensile strains are set up in the fiber direction (due to the Poisson's effect), which must be continuous across the phases, thus providing the required stiffening effect in the transverse direction (because the linear phase controls the strains along the fibers).

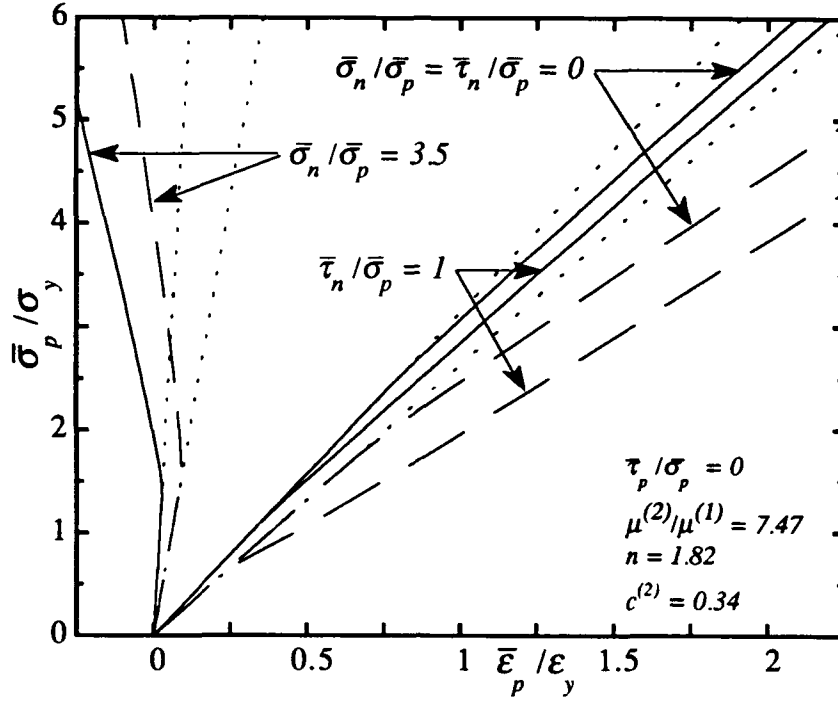


Fig. 6. Lower estimates (continuous curves) and upper estimates (long-dash curves) for the relations between the in-plane hydrostatic stress $\bar{\sigma}_p$ and the corresponding strain $\bar{\epsilon}_p$ for the nonlinear fiber composites, as well as for the reference linear fiber composites (short-dash lines), for three different loading combinations: $\bar{\sigma}_n / \bar{\sigma}_p = \bar{\tau}_n / \bar{\sigma}_p = 0$, $\bar{\sigma}_n / \bar{\sigma}_p = 3.5$ and $\bar{\tau}_n / \bar{\sigma}_p = 1$. Here, $\bar{\tau}_p = 0$ for all cases.

The coupling between the two dilatational modes is demonstrated by the two curves for the proportional loading path $\bar{\sigma}_n / \bar{\sigma}_p = 3.5$. We observe that the two curves have the same behaviors as the corresponding curves for the reference linear composite (short-dash lines), until the yielding of phase 1. After yielding, however, the in-plane hydrostatic strain decreases for both types of estimates. The reason is the same as discussed previously, namely, the independence of the plastic strains (in the aluminum phase) on the hydrostatic stress. The curves for the proportional loading path $\bar{\tau}_n / \bar{\sigma}_p = 1$ shows the coupling between the in-plane hydrostatic stress and the longitudinal shear stress $\bar{\tau}_n$. These curves resemble the corresponding curves for $\bar{\tau}_n / \bar{\sigma}_n = 1$ in Figure 5.

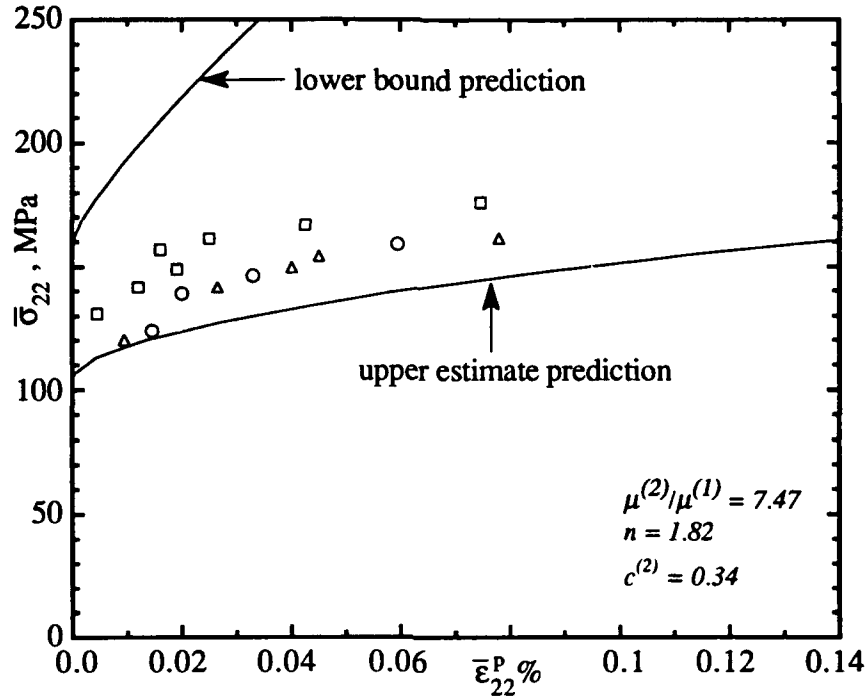


Fig. 7. Lower and upper estimates (continuous curves) for the relations between the uniaxial tensile stress $\bar{\sigma}_{22}$ and the uniaxial plastic tensile strain $\bar{\epsilon}_{22}^p$ for the nonlinear fiber composite, and results for three sets of experimental data points (squares, triangles and circles), corresponding to three tests carried out on an aluminum-matrix composite reinforced with boron fibers (Adams 1970).

Finally, we make a comparison between our theoretical results and the experimental data of Adams (1970). For simplicity, we consider a Cartesian coordinate frame, where the x_3 axis is aligned with the fiber direction $\mathbf{n}$, and the other two axes lie in the transverse plane (see Figure 1). We make use the results of Appendix I to determine the values of the four transversely isotropic stress (strain) invariants in terms of the Cartesian components for a given stress (strain) tensor. In this case, the applied load $\bar{\sigma}_{22}$ is a tensile stress in the transverse direction, and we have $\bar{\sigma}_p = \frac{1}{2} \bar{\sigma}_{22}$, $\bar{\tau}_p = \frac{1}{2} \bar{\sigma}_{22}$. Therefore, by means of the chain rule, the corresponding tensile strain component is

$$\bar{\epsilon}_{22} = \frac{\partial \bar{U}}{\partial \bar{\sigma}_{22}} = \bar{\epsilon}_p + \bar{\gamma}_p.$$

Figure 7 shows plots of the transverse tensile stress $\bar{\sigma}_{22}$ versus the plastic part of the corresponding tensile strain $\bar{\epsilon}_{22}^p$. Three sets of experimental data points are shown (squares,

triangles and circles), representing the three different tests carried out on the aluminum-matrix composite (Adams 1970). Two curves representing the corresponding two estimates obtained from the lower bound and the upper estimate for the effective energy function are also shown. We observe that the two curves bound the experimental data points from above and below. However, the curve obtained from the upper energy estimate is much closer to the experimental data points. This is in agreement with our prior discussions stating that the stress-strain relations derived from the upper energy estimate can be used as estimates for the behavior of two-phase fiber composites with the ductile phase playing the role of the matrix and the brittle phase that of the fibers.

8. Concluding remarks

In this work, we have obtained bounds and estimates for the effective behavior of nonlinear fiber-reinforced composites by means of a variational procedure introduced by Ponte Castañeda (1991a, 1992). This procedure enabled the extension of upper and lower bounds of the Hashin-Shtrikman (1962) type for the effective energy functions of n -phase *linear* fiber-reinforced composites to corresponding lower bounds and upper estimates (estimates for the upper bound) for the effective energy functions of n -phase *nonlinear* fiber composites. Simple representations for these bounds and estimates, generalizing earlier results by Ponte Castañeda (1992) for the class of *incompressible* fiber composites, were obtained for the class of *compressible* fiber composites. We note that different, equivalent expressions for the lower bound for the effective energy functions of *incompressible* fiber composites have also been obtained by Talbot and Willis (1991), using a different procedure. However, the form of the lower bounds obtained in this work is simpler, and, additionally, the nonlinear Hashin-Shtrikman upper estimates are also new.

The important case of a compressible, two-phase fiber composites made up of a ductile and a brittle phase was studied in detail. The general expression for the lower bound was specialized for this class of composites, and a similar expression, based on neglecting terms of order $(\mu_o^{(1)}/\kappa_o^{(1)})^2$, was obtained for the upper estimate. Corresponding estimates for the effective stress-strain relations were derived by straightforward differentiation of these expressions. Explicit calculations

were carried out for an aluminum-boron system, highlighting significant couplings that arise between the different loading modes.

Finally, we proposed alternative (approximate) interpretations for the constitutive relations derived from the lower bounds and upper estimates for the effective energy functions of two-phase, nonlinear fiber composites. Thus, the results based on the lower bounds were given the interpretation of estimates for the effective behavior of fiber composites made up of stiffer linear-elastic matrices weakened by ductile fibers. Correspondingly, the results based on the upper estimates for the effective energy functions were given the interpretation of estimates for the effective behavior of fiber composites made up of ductile matrices reinforced by stiffer linear-elastic fibers. This latter interpretation was confirmed through comparison with available experimental results for a metal-matrix, fiber-reinforced composite.

Acknowledgments

This work was supported by the Air Force Office of Scientific Research (Grant No. 91-0161). The authors thank Mr. Kingshuk Bose for his valuable comments on an earlier version of the paper.

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Appendix I: On the characterization of transversely isotropic materials

The purpose of this appendix is to gather some results relevant to the analysis of materials with transversely isotropic symmetry. These results are used extensively throughout the body of the paper in the development of estimates for the effective behavior of nonlinear fiber composites, which constitute a special class of transversely isotropic materials. The emphasis of this section is on representations for the transversely isotropic invariants of the stress and strain tensors. The reason is that *nonlinear* transversely isotropic materials are most efficiently characterized in terms of energy-density functions depending on these invariants.

I.1 Isotropic invariants

As is well-known, there are three isotropic invariants for a symmetric, second-order tensor. However, only two of these, which are of quadratic order or less, are relevant to linear-elastic behavior. These invariants may be expressed (see, for example, Walpole 1981) in terms of two fourth-order *projection* tensors $\mathbf{J}$ and $\mathbf{K}$, such that $\mathbf{I} = \mathbf{J} + \mathbf{K}$, $\mathbf{J} = \mathbf{J}\mathbf{J}$, $\mathbf{K} = \mathbf{K}\mathbf{K}$ and $\mathbf{J}\mathbf{K} = \mathbf{0}$. Their Cartesian components are given by

$$(I.1) \quad J_{ijkl} = \frac{1}{3} \delta_{ij} \delta_{kl}, \quad K_{ijkl} = \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl}),$$

where δ_{ij} is the Kronecker delta symbol. Then, in terms of these projection tensors, we define two isotropic invariants of the stress tensor via

$$(I.2) \quad \sigma_m = \frac{1}{3} J_{kkij} \sigma_{ij} \quad \text{and} \quad \tau_e^2 = \frac{1}{2} K_{ijkl} \sigma_{ij} \sigma_{kl},$$

called the hydrostatic (mean) stress, and the effective shear stress, respectively. We also define the hydrostatic strain ϵ_m , and the effective shear strain γ_e by relations analogous to (I.2).

It is important to note that the elasticity tensor $\mathbf{L}$ of an isotropic, linear-elastic material admits a spectral decomposition

$$(I.3) \quad \mathbf{L} = 3\kappa\mathbf{J} + 2\mu\mathbf{K},$$

where $\mathbf{J}$ and $\mathbf{K}$ play the role of the eigenprojections, and the bulk and shear moduli of the material, κ and μ , are the corresponding eigenvalues.

II.2 Transversely isotropic invariants

There are in general five transversely isotropic invariants of a symmetric, second-order tensor (Spencer 1971). However, only four of these invariants are linear, or quadratic, in order. They may be represented in terms of the four projection tensors (Walpole 1981) $\mathbf{E}^{[1]}$, $\mathbf{E}^{[2]}$, $\mathbf{E}^{[3]}$, and $\mathbf{E}^{[4]}$, satisfying the relations $\mathbf{E}^{[p]}\mathbf{E}^{[p]} = \mathbf{E}^{[p]}$, $\mathbf{E}^{[p]}\mathbf{E}^{[q]} = \mathbf{0}$, $p \neq q$; and $\mathbf{E}^{[1]} + \mathbf{E}^{[2]} + \mathbf{E}^{[3]} + \mathbf{E}^{[4]} = \mathbf{I}$.

The Cartesian components of these four projection tensors are given by

$$(I.4) \quad \begin{aligned} E_{ijkl}^{[1]} &= \frac{1}{2} \beta_{ij} \beta_{kl}, \\ E_{ijkl}^{[2]} &= \alpha_{ij} \alpha_{kl}, \\ E_{ijkl}^{[3]} &= \frac{1}{2} (\beta_{ik} \beta_{jl} + \beta_{jk} \beta_{il} - \beta_{ij} \beta_{kl}), \\ E_{ijkl}^{[4]} &= \frac{1}{2} (\beta_{ik} \alpha_{jl} + \beta_{il} \alpha_{jk} + \beta_{jl} \alpha_{ik} + \beta_{jk} \alpha_{il}), \end{aligned}$$

where $\alpha_{ij} = n_i n_j$ and $\beta_{ij} = \delta_{ij} - n_i n_j$, with $\mathbf{n}$ denoting the axis of transverse isotropy. Then, the four transversely isotropic invariants of the stress tensor $\boldsymbol{\sigma}$ may be expressed in the forms

$$(I.5) \quad \begin{aligned} \sigma_p &= \frac{1}{2} E_{iikl}^{[1]} \sigma_{kl} = \frac{1}{2} \sigma_{ij} \beta_{ij}, & \left\{ \frac{1}{2} (\sigma_{11} + \sigma_{22}) \right\}, \\ \sigma_n &= E_{iikl}^{[2]} \sigma_{kl} = \sigma_{ij} \alpha_{ij}, & \left\{ \sigma_{33} \right\}, \\ \tau_p^2 &= \frac{1}{2} \sigma_{ij} E_{ijkl}^{[3]} \sigma_{kl} = \frac{1}{2} \left[\sigma_{ij} \sigma_{kl} \beta_{ik} \beta_{jl} - \frac{1}{2} (\sigma_{ij} \beta_{ij})^2 \right], & \left\{ \sigma_{12}^2 + \frac{1}{4} (\sigma_{11} - \sigma_{22})^2 \right\}, \\ \tau_n^2 &= \frac{1}{2} \sigma_{ij} E_{ijkl}^{[4]} \sigma_{kl} = \left[\sigma_{ij} \sigma_{kl} \alpha_{jk} - (\sigma_{ij} \alpha_{ij})^2 \right], & \left\{ (\sigma_{13}^2 + \sigma_{23}^2) \right\}. \end{aligned}$$

Physically, these invariants correspond to the in-plane hydrostatic stress, the normal tensile stress, the (in-plane) transverse shear stress, and the (anti-plane) longitudinal shear stress (given in brackets are the corresponding representations for a choice of $\mathbf{n}$ aligned with the 3-direction).

Analogous relations apply for the transversely isotropic invariants of the strain tensor $\boldsymbol{\epsilon}$, denoted respectively by ϵ_p , ϵ_n , γ_p , and γ_n . We also note for latter reference that the following relations

hold between the transversely isotropic invariants of (I.5) and the isotropic invariants of (I.2),

$$(I.6) \quad \sigma_m = \frac{1}{3} (2\sigma_p + \sigma_n), \quad \tau_e^2 = \tau_p^2 + \tau_n^2 + \frac{1}{3} (\sigma_p - \sigma_n)^2.$$

Contrary to the situation for isotropic materials, the above four projection tensors are not the eigentensors of the spectral decomposition of an arbitrary transversely isotropic elasticity tensor (Mehrabadi and Cowin 1990). Such eigentensors would involve the material moduli. Therefore, it

is necessary to introduce (Walpole 1981) two other tensors, that are *not* projections, $\mathbf{E}^{[5]}$ and $\mathbf{E}^{[6]}$, with components

$$(I.7) \quad E_{ijkl}^{[5]} = \alpha_{ij}\beta_{kl}, \quad \text{and} \quad E_{ijkl}^{[6]} = \beta_{ij}\alpha_{kl}.$$

The elasticity tensor $\mathbf{L}$ of an arbitrary transversely isotropic material may be expressed in terms of these six tensors, via

$$(I.8) \quad \mathbf{L} = a_1 \mathbf{E}^{[1]} + a_2 \mathbf{E}^{[2]} + a_3 \mathbf{E}^{[3]} + a_4 \mathbf{E}^{[4]} + a_5 (\mathbf{E}^{[5]} + \mathbf{E}^{[6]}),$$

where a_q ($q = 1, \dots, 5$) are the five moduli that suffice to characterize the behavior of the material. The compliance tensor $\mathbf{M}$, of an arbitrary transversely isotropic material may be expressed in a similar form. It is worth mentioning that the isotropic projection $\mathbf{J}$, can be represented in the form

$$(I.9) \quad \mathbf{J} = \frac{2}{3} \mathbf{E}^{[1]} + \frac{1}{3} \mathbf{E}^{[2]} + \frac{1}{3} (\mathbf{E}^{[5]} + \mathbf{E}^{[6]}),$$

and that we can additionally define, for latter reference, the tensor $\mathbf{E}'$ such that

$$(I.10) \quad \mathbf{E}' = \mathbf{E}^{[3]} + \mathbf{E}^{[4]} - \mathbf{K}.$$

The tensor $\mathbf{E}'$ is a projection tensor, orthogonal to $\mathbf{E}^{[3]}$ and $\mathbf{E}^{[4]}$.

Finally, we remark that the energy density function of a transversely isotropic, linear-elastic materials may be represented in the form

$$(I.11) \quad U(\boldsymbol{\sigma}) = \psi(\sigma_p, \sigma_n, \tau_p, \tau_n).$$

Then, the relations between the transversely isotropic stress and strain invariants are given by

$$(I.12) \quad \epsilon_p = \frac{1}{2} \frac{\partial \psi}{\partial \sigma_p}, \quad \epsilon_n = \frac{\partial \psi}{\partial \sigma_n}, \quad \gamma_p = \frac{1}{2} \frac{\partial \psi}{\partial \tau_p} \quad \text{and} \quad \gamma_n = \frac{1}{2} \frac{\partial \psi}{\partial \tau_n}.$$

I.3 Incompressible, transversely isotropic invariants

For incompressible, transversely isotropic materials, it suffices to consider the three invariants of quadratic order or less, on the space of traceless, symmetric, second-order tensors. These may be obtained in terms of the three orthogonal projection tensors $\mathbf{E}^{[3]}$, $\mathbf{E}^{[4]}$ and $\mathbf{E}'$, defined in the previous subsection. Thus, the incompressible, transversely isotropic invariants of the stress tensor $\boldsymbol{\sigma}$ are τ_p , τ_n , and the deviatoric (axisymmetric) shear stress

$$(I.13) \quad \tau_d = \frac{1}{\sqrt{3}} (\sigma_p - \sigma_n),$$

corresponding to the three above projections, respectively. We note that from (I.6)₂ we have the following identity relating the effective shear stress and the incompressible, transversely isotropic invariants, $\tau_e^2 = \tau_p^2 + \tau_n^2 + \tau_d^2$. The corresponding strain invariants are denoted γ_p , γ_n and γ_d .

Finally, we note that the elasticity tensor $\mathbf{L}$ of an incompressible, transversely isotropic, linear-elastic material admits a spectral decomposition of the form

$$(I.14) \quad \mathbf{L} = 2\mu_p \mathbf{E}^{[3]} + 2\mu_n \mathbf{E}^{[4]} + 2\mu_d \mathbf{E},$$

where μ_p , μ_n , μ_d , are the three shear moduli characterizing the behavior of such a material (Lipton, 1992).

Appendix II: Estimates for the effective stress-strain relations

In this appendix, we demonstrate the process of obtaining estimates for the effective stress-strain relations of nonlinear fiber composites from corresponding expressions for bounds and estimates for the effective energy functions, which are given in the body of the paper. We begin by considering expression (21) for the lower bound for the effective energy function. Since all the phases in the composite are assumed to be isotropic, the functions $V^{(r)}$ in expression (21) may be written in the form

$$(II.1) \quad V^{(r)}(\mu_o^{(r)}, \kappa_o^{(r)}) = \max_{\tau_e^{(r)}, \sigma_m^{(r)}} \left\{ \frac{1}{2\mu_o^{(r)}} (\tau_e^{(r)})^2 + \frac{1}{2\kappa_o^{(r)}} (\sigma_m^{(r)})^2 - \psi^{(r)}(\tau_e^{(r)}, \sigma_m^{(r)}) \right\}.$$

Assuming sufficient smoothness for the functions $\psi^{(r)}$, the corresponding optimization conditions can be expressed in terms of the following $2n$ implicit equations for the variables $\tau_e^{(r)}$ and $\sigma_m^{(r)}$,

$$(II.2) \quad \frac{1}{\mu_o^{(r)}} \tau_e^{(r)} - \frac{\partial \psi^{(r)}}{\partial \tau_e^{(r)}}(\tau_e^{(r)}, \sigma_m^{(r)}) = 0, \quad \text{and} \quad \frac{1}{\kappa_o^{(r)}} \sigma_m^{(r)} - \frac{\partial \psi^{(r)}}{\partial \sigma_m^{(r)}}(\tau_e^{(r)}, \sigma_m^{(r)}) = 0,$$

and we denote by $\hat{\tau}_e^{(r)}(\mu_o^{(r)}, \kappa_o^{(r)})$ and $\hat{\sigma}_m^{(r)}(\mu_o^{(r)}, \kappa_o^{(r)})$ the solutions for these equations. Then, substitution of the solutions for $V^{(r)}$ into (21), leads to

$$(II.3) \quad \tilde{U}^{(ns-)}(\bar{\sigma}) = \max_{\mu_o^{(r)}, \kappa_o^{(r)} \geq 0} \left\{ F(\bar{\sigma}; \mu_o^{(r)}, \kappa_o^{(r)}) \right\},$$

where

$$(II.4) \quad F = \left[\frac{1}{2} \bar{\sigma}_{ij} (\tilde{M}_o^{(ns-)})_{ijkl} \bar{\sigma}_{kl} - \sum_{r=1}^n c^{(r)} \left[\frac{1}{2\mu_o^{(r)}} (\hat{\tau}_e^{(r)})^2 + \frac{1}{2\kappa_o^{(r)}} (\hat{\sigma}_m^{(r)})^2 - \psi^{(r)}(\hat{\tau}_e^{(r)}, \hat{\sigma}_m^{(r)}) \right] \right].$$

Once again, the optimization conditions for the above problem can be stated in terms of $2n$ equations for the variables $\mu_o^{(r)}$ and $\kappa_o^{(r)}$, namely,

$$(II.5) \quad \frac{\partial F(\bar{\sigma}; \mu_o^{(r)}, \kappa_o^{(r)})}{\partial \mu_o^{(r)}} = 0, \quad \text{and} \quad \frac{\partial F(\bar{\sigma}; \mu_o^{(r)}, \kappa_o^{(r)})}{\partial \kappa_o^{(r)}} = 0.$$

We denote the optimal values of the variables $\mu_o^{(r)}$ and $\kappa_o^{(r)}$, satisfying (II.5), by $\hat{\mu}_o^{(r)}(\bar{\sigma})$ and $\hat{\kappa}_o^{(r)}(\bar{\sigma})$, respectively. In terms of these optimal values, the lower bound for the effective energy function may be expressed in the form

$$(II.6) \quad \tilde{U}^{(ns-)}(\bar{\sigma}) = F(\bar{\sigma}; \hat{\mu}_o^{(r)}, \hat{\kappa}_o^{(r)}),$$

and the corresponding estimate for the stress-strain relations is given by

$$(II.7) \quad \bar{\epsilon}_{ij} = \frac{\partial \tilde{U}^{(ns-)}(\bar{\sigma})}{\partial \sigma_{ij}} \\ = \left[\tilde{M}_o^{(ns-)}(\hat{\mu}_o^{(r)}, \hat{\kappa}_o^{(r)}) \right]_{ijkl} \bar{\sigma}_{kl} + \sum_{r=1}^n \frac{\partial F(\bar{\sigma}; \hat{\mu}_o^{(r)}, \hat{\kappa}_o^{(r)})}{\partial \mu_o^{(r)}} \frac{\partial \hat{\mu}_o^{(r)}(\bar{\sigma})}{\partial \sigma_{ij}} + \sum_{r=1}^n \frac{\partial F(\bar{\sigma}; \hat{\mu}_o^{(r)}, \hat{\kappa}_o^{(r)})}{\partial \kappa_o^{(r)}} \frac{\partial \hat{\kappa}_o^{(r)}(\bar{\sigma})}{\partial \sigma_{ij}}.$$

However, due to the optimization conditions (II.5), the last two sums in (II.7) vanish, and the estimate (22) for the effective stress-strain relations is obtained. Analogous expressions for the upper estimate, effective stress-strain relations may be obtained similarly.

Next, we consider the estimate for effective stress-strain relations (32), obtained from the lower bound for the effective energy function of the incompressible fiber composite (30). For simplicity, we assume that the n th branch attains the minimum in (30) (*e.g.* $s = n$), and additionally we define $\bar{\tau}^2 = \bar{\tau}_p^2 + \bar{\tau}_n^2$. Then, the optimization constraints $\bar{\omega} = 1$ and $\bar{\eta} = 1$ may be eliminated by letting

$$(II.8) \quad \omega^{(n)} = \frac{1}{c^{(n)}} \left[1 - \sum_{r=1}^{n-1} c^{(r)} \omega^{(r)} \right], \quad \text{and} \quad \eta^{(n)} = \frac{1}{c^{(n)}} \left[1 - \sum_{r=1}^{n-1} c^{(r)} \eta^{(r)} \right].$$

In terms of the $2(n-1)$ optimizations variables $\omega^{(r)}, \eta^{(r)}$ ($r = 1, \dots, n-1$), expression (30) may be rewritten in the form

$$(II.9) \quad \tilde{U}^{(ns-)}(\bar{\sigma}) = \min_{\omega^{(r)}, \eta^{(r)}} \left\{ \sum_{r=1}^{n-1} c^{(r)} \psi^{(r)}(\tau_e^{(r)}) + c^{(n)} \psi^{(n)}(\tau_e^{(n)}) \right\},$$

where the variables $\tau_e^{(r)}$ ($r = 1, \dots, n-1$) are given by relation (31)₁, and

$$(II.10) \quad \tau_e^{(n)} = \sqrt{\bar{\tau}^2 \left[1 + 2 \left(\frac{1}{c^{(n)}} - \sum_{i=1}^{n-1} \frac{c^{(i)}}{c^{(n)}} \omega^{(i)} \right)^2 - \sum_{i=1}^n \frac{c^{(i)}}{c^{(n)}} (1 - (\omega^{(i)})^2) \right] + \bar{\tau}_d^2 \left(\frac{1}{c^{(n)}} - \sum_{i=1}^{n-1} \frac{c^{(i)}}{c^{(n)}} \eta^{(i)} \right)^2}.$$

It follows that the $2(n-1)$ optimization conditions in (II.9) are given by the relations

$$(II.11) \quad \frac{1}{\tau_e^{(r)}} (\psi^{(r)})' (\tau_e^{(r)}) \omega^{(r)} + \frac{1}{\tau_e^{(n)}} (\psi^{(n)})' (\tau_e^{(n)}) \left[\omega^{(n)} - \frac{2}{c^{(n)}} \left(1 - \sum_{i=1}^{n-1} c^{(i)} \omega^{(i)} \right) \right] = 0,$$

and

$$\frac{1}{\tau_e^{(r)}} (\psi^{(r)})' (\tau_e^{(r)}) \eta^{(r)} + \frac{1}{c^{(n)} \tau_e^{(n)}} (\psi^{(n)})' (\tau_e^{(n)}) \left(1 - \sum_{i=1}^{n-1} c^{(i)} \eta^{(i)} \right) = 0.$$

If we now denote the optimal values of $\omega^{(r)}$ and $\eta^{(r)}$, satisfying (II.11), by $\hat{\omega}^{(r)}$ and $\hat{\eta}^{(r)}$, respectively, the lower bound for the effective energy function may be written in the form

$$(II.12) \quad \tilde{U}^{(ns-)}(\bar{\sigma}) = \sum_{r=1}^{n-1} c^{(r)} \psi^{(r)}(\hat{\tau}_e^{(r)}) + c^{(n)} \psi^{(n)}(\hat{\tau}_e^{(n)}),$$

where $\hat{\tau}_e^{(r)} = \sqrt{\bar{\tau}^2 (\hat{\omega}^{(r)})^2 + \bar{\tau}_d^2 (\hat{\eta}^{(r)})^2}$ ($r = 1, \dots, n-1$), and $\hat{\tau}_e^{(n)} = \tau_e^{(n)}(\hat{\omega}^{(r)}, \hat{\eta}^{(r)})$ [as given by (II.10)]. The corresponding estimate for the effective stress-strain relation of the incompressible fiber composite then becomes

$$(II.13) \quad \begin{aligned} \bar{\epsilon} = & \sum_{r=1}^{n-1} \frac{c^{(r)}}{\hat{\tau}_e^{(r)}} (\psi^{(r)})' (\hat{\tau}_e^{(r)}) \left[(\hat{\omega}^{(r)})^2 \bar{\tau} \frac{\partial \bar{\tau}}{\partial \bar{\sigma}} + (\hat{\eta}^{(r)})^2 \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right] + \dots \\ & \dots + \frac{c^{(n)}}{\hat{\tau}_e^{(n)}} (\psi^{(n)})' (\hat{\tau}_e^{(n)}) \left[\left[1 + 2 \left(\frac{1}{c^{(n)}} - \sum_{i=1}^{n-1} \frac{c^{(i)}}{c^{(n)}} \hat{\omega}^{(i)} \right)^2 - \sum_{i=1}^n \frac{c^{(i)}}{c^{(n)}} (1 - (\hat{\omega}^{(i)})^2) \right] \bar{\tau} \frac{\partial \bar{\tau}}{\partial \bar{\sigma}} + \dots \right. \\ & \left. \dots + \left(\frac{1}{c^{(n)}} - \sum_{i=1}^{n-1} \frac{c^{(i)}}{c^{(n)}} \hat{\eta}^{(i)} \right)^2 \bar{\tau}_d \frac{\partial \bar{\tau}_d}{\partial \bar{\sigma}} \right] + \dots \\ & \dots + \sum_{r=1}^{n-1} c^{(r)} \bar{\tau}^2 \frac{\partial \hat{\omega}^{(r)}}{\partial \bar{\sigma}} \left[\frac{1}{\hat{\tau}_e^{(r)}} (\psi^{(r)})' (\hat{\tau}_e^{(r)}) \hat{\omega}^{(r)} + \frac{1}{\hat{\tau}_e^{(n)}} (\psi^{(n)})' (\hat{\tau}_e^{(n)}) \left[\hat{\omega}^{(n)} - \frac{2}{c^{(n)}} \left(1 - \sum_{i=1}^{n-1} c^{(i)} \hat{\omega}^{(i)} \right) \right] \right] + \dots \\ & \dots + \sum_{r=1}^{n-1} c^{(r)} \bar{\tau}_d^2 \frac{\partial \hat{\eta}^{(r)}}{\partial \bar{\sigma}} \left[\frac{1}{\hat{\tau}_e^{(r)}} (\psi^{(r)})' (\hat{\tau}_e^{(r)}) \hat{\eta}^{(r)} + \frac{1}{c^{(n)} \hat{\tau}_e^{(n)}} (\psi^{(n)})' (\hat{\tau}_e^{(n)}) \left(1 - \sum_{i=1}^{n-1} c^{(i)} \hat{\eta}^{(i)} \right) \right]. \end{aligned}$$

We note that each of the terms in the last two sums in (II.13) is identical to zero by virtue of the optimization conditions (II.11). Thus, in the computation of the effective stress-strain relations, we

may regard the optimization variables as constants (as far as derivatives with respect to $\bar{\sigma}$ are concerned). The final result is given by relation (32), where $\hat{\omega}^{(n)}$ and $\hat{\eta}^{(n)}$ are defined via relations (II.8) in terms of the $\hat{\omega}^{(r)}$ and $\hat{\eta}^{(r)}$ ($r = 1, \dots, n-1$), respectively.

The corresponding expressions for the upper estimates for the effective stress-strain relations of the incompressible fiber composite are computed similarly. It can also be shown that analogous results may be obtained for the *compressible* nonlinear fiber composites.

Appendix III: A useful identity

In many applications, expressions for the effective elasticity tensors of linear-elastic composites involve the evaluation of a fourth-order tensor $\hat{\mathbf{M}}$, defined by

$$(III.1) \quad \hat{\mathbf{M}} = \left[\sum_{r=1}^n c^{(r)} [\mathbf{M}^{(r)}]^{-1} \right]^{-1},$$

where $\mathbf{M}^{(r)}$ are positive definite, satisfying the conditions $M_{ijkl}^{(r)} = M_{jikl}^{(r)} = M_{ijlk}^{(r)} = M_{klij}^{(r)}$, and where $\sum_{r=1}^n c^{(r)} = 1$ with $0 \leq c^{(r)} \leq 1$, (see, for example, Hill 1965 and Walpole 1969). For later reference, we note that all the symmetries of the tensors $\mathbf{M}^{(r)}$ are carried over to the tensor $\hat{\mathbf{M}}$.

The aim of this appendix is to demonstrate the following identity, which is used repeatedly in the body of the paper,

$$(III.2) \quad \sigma_{ij} \hat{M}_{ijst} \sigma_{st} = \min_{\Omega^{(r)}, \bar{\Omega}=1} \left\{ \sum_{r=1}^n c^{(r)} \sigma_{ij} \Omega_{klij}^{(r)} M_{klpq}^{(r)} \Omega_{pqst}^{(r)} \sigma_{st} \right\},$$

for all second-order symmetric tensors σ , where the variables $\Omega^{(r)}$ ($r = 1, \dots, n$), are subject to the optimization constraint $\sum_{r=1}^n c^{(r)} \Omega^{(r)} = \mathbf{I}$, and satisfy the symmetry conditions $\Omega_{ijkl} = \Omega_{jikl} = \Omega_{ijlk}$.

This identity is a generalization of an analogous identity, first presented in deBotton and Ponte Castañeda (1992).

We begin by letting $G(\Omega^{(r)})$ be the function defined by

$$(III.3) \quad G_{ijst}(\Omega^{(r)}) = \sum_{r=1}^n c^{(r)} \Omega_{klij}^{(r)} M_{klpq}^{(r)} \Omega_{pqst}^{(r)},$$

where the variables $\Omega^{(r)}$ are subject to the constraint $\bar{\Omega} = \mathbf{I}$, and where the tensors $\mathbf{M}^{(r)}$ and the constants $c^{(r)}$ are as given above. The choice of the set

$$(III.4) \quad \Omega_{ijkl}^{(r)} = \left(M^{(r)}\right)_{ijpq}^{-1} \hat{M}_{pqkl},$$

satisfies both the optimization constraints and the symmetry conditions stated in (III.2), and is such that $\mathbf{G}(\Omega^{(r)}) = \hat{\mathbf{M}}$. We consider next a second, arbitrary set, distinct from the first set, $\hat{\Omega}^{(r)}$ ($r = 1, \dots, n$), such that $\bar{\hat{\Omega}}^{(r)} = \mathbf{I}$, and we let $\Theta^{(r)} = \hat{\Omega}^{(r)} - \Omega^{(r)} \neq \mathbf{0}$. Then, substitution of this second set into (III.3) leads to

$$(III.5) \quad \begin{aligned} G_{ijst}(\hat{\Omega}^{(r)}) &= \sum_{r=1}^n c^{(r)} (\Omega_{klij}^{(r)} + \Theta_{klij}^{(r)}) M_{klpq}^{(r)} (\Omega_{pqst}^{(r)} + \Theta_{pqst}^{(r)}) \\ &= \sum_{r=1}^n c^{(r)} \Omega_{klij}^{(r)} M_{klpq}^{(r)} \Omega_{pqst}^{(r)} + \sum_{r=1}^n c^{(r)} \Theta_{klij}^{(r)} M_{klpq}^{(r)} \Theta_{pqst}^{(r)}, \end{aligned}$$

where we have used the fact that $\bar{\Theta} = \mathbf{0}$. In addition, since the tensors $\mathbf{M}^{(r)}$ are positive definite, it follows that for all nonzero, second-order, symmetric tensors σ ,

$$(III.6) \quad \sigma_{ij} \Theta_{klij}^{(r)} M_{klpq}^{(r)} \Theta_{pqst}^{(r)} \sigma_{st} = \left(\Theta_{klij}^{(r)} \sigma_{ij}\right) M_{klpq}^{(r)} \left(\Theta_{pqst}^{(r)} \sigma_{st}\right) > 0.$$

The identity (III.2) then follows from (III.5) and (III.6), because

$$(III.7) \quad \sigma_{ij} G_{ijkl}(\hat{\Omega}^{(r)}) \sigma_{kl} > \sigma_{ij} G_{ijkl}(\Omega^{(r)}) \sigma_{kl} = \sigma_{ij} \hat{M}_{ijkl} \sigma_{kl},$$

for all possible $\hat{\Omega}^{(r)} \neq \Omega^{(r)}$ satisfying the optimization constraint and the symmetry conditions, and nonzero, second-order, symmetric tensors σ .

Finally, we note that this identity can be further generalized to the case of a tensor $\mathbf{M}$ with arbitrary dependence on the position vector $\mathbf{x}$. In this case the summations in (III.1) and (III.2) are replaced by integrations over the volume Ω . The optimization variables $\Omega = \Omega(\mathbf{x})$ satisfy the symmetry conditions stated in (III.2) and the optimization constraint $\int_{\Omega} \Omega(\mathbf{x}) d\mathbf{x} = \mathbf{I}$.

Reference [16]

CONSTITUTIVE MODELS FOR DUCTILE SOLIDS REINFORCED BY RIGID SPHEROIDAL INCLUSIONS

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Abstract— We study the effective constitutive response of composite materials made of rigid spheroidal inclusions dispersed in a ductile matrix phase. Given a general convex potential characterizing the plastic (in the context of J_2 -deformation theory) behavior of the matrix material, we derive expressions for the corresponding effective potentials of the rigidly reinforced composites, under *general* loading conditions. The derivation of the effective potentials for the nonlinear composites is based on a variational procedure developed recently by Ponte Castañeda (1991a). We consider two classes of composites. In the first class, the spheroidal inclusions are aligned, resulting in overall transversely isotropic symmetry for the composite. In the second class, the inclusions are randomly oriented, and thus the composite is macroscopically isotropic. The effective response of composites with aligned inclusions depends on both the orientation of the loading and on the inclusion concentration and shape. Comparing the strengthening effects of rigid oblate and prolate spheroids, we find that pro-

late spheroids give rise to stiffer effective response under axisymmetric (relative to the axis of transverse isotropy) deformations, while oblate spheroids provide greater reinforcement for materials loaded in transverse shear (relative to the axial direction). On the other hand, nearly spherical (slightly prolate) spheroids are most effective in strengthening the composite under longitudinal shear deformations. Thus, the optimal shape for strengthening composites with aligned inclusions depends strongly on the deformation mode. Alternatively, the properties of composites with randomly oriented spheroidal inclusions, being isotropic, depend only on the concentration and shape of the inclusions. We find that both oblate and prolate inclusions lead to significant strengthening for this class of composites.

1. INTRODUCTION

In this work, we seek to estimate the effective, or overall, constitutive response of composite materials comprised of an incompressible, ductile matrix phase reinforced by rigid, spheroidal inclusions. The matrix and inclusions are assumed to be perfectly bonded, and the inclusions are further assumed to be small compared to the overall dimension of the specimen under consideration. Here, we are interested in predicting the effects of the volume fraction, aspect ratio and orientation of the inclusions on the effective properties of the composite. A potential application of this study would be in the optimal design of materials in which a nonlinear ductile phase is reinforced by relatively stiff inclusions. These materials include some metal-matrix composites and intermetallics reinforced by stiff particles.

Composites with linear constitutive behavior for both the matrix and inclusion phases have been studied extensively. In this connection, Eshelby's (1957) celebrated solution for the constant stress and strain fields within an ellipsoidal inclusion in an infinite matrix, subjected

to uniform remote tractions, was instrumental in many developments, including the determination of the effective moduli of elastic composites with dilute concentrations of ellipsoidal inclusions. This fundamental result can also be used to compute the effective properties of multi-phase elastic composites with nondilute concentrations of the phases. Thus, for this purpose, Budiansky (1965) and Hill (1965) proposed the self-consistent method and obtained explicit expressions for the effective moduli of composites with isotropic distributions of spherical inclusions. Wu (1966) extended the use of the self-consistent method to predict the effective behavior of isotropic composites with randomly oriented ellipsoidal inclusions. Another remarkable achievement in the field of linear composite materials was the developments of new variational principles by Hashin and Shtrikman (1962). These variational principles allowed the derivation, by Hashin and Shtrikman (1963), of upper and lower bounds for the effective moduli of isotropic composite materials with prescribed volume fractions of the phases, but otherwise arbitrary microstructure. Extensions of this class of bounds for fiber-reinforced composites with overall transversely isotropic symmetry were first given by Hill (1964) and Hashin (1965). Further generalization of the Hashin-Shtrikman bounds and self-consistent estimates for anisotropic linear elastic composites have been given by Walpole (1966a,b, 1969) and Willis (1977). An alternative approach in the study of composite materials is to assume periodic microstructures and to compute the effective properties of the composite from a boundary-value problem defined on the unit cell with periodic boundary conditions. This approach has been pursued by Nemat-Nasser and Taya (1981) and Nemat-Nasser *et al.* (1982). For a more complete review of the field of linear composites, the reader is referred to the articles by Hashin (1983) and Willis (1982) and to the monograph by Sanchez-Palencia (1980).

Nonlinear composite materials have recently been the subject of increased attention. Talbot and Willis (1987) and Ponte Castañeda and Willis (1988) (see also Willis 1991) developed bounds and self-consistent estimates for the effective behavior of nonlinear composites via an extension of the Hashin-Shtrikman variational principles to nonlinear materials, proposed by Talbot and Willis (1985). More recently, Ponte Castañeda (1991a) proposed a new, conceptually simple, variational procedure which leads to more general bounds and estimates for the properties of nonlinear composites. The procedure makes use of arbitrary bounds and estimates for classes of *linear* comparison composites to generate bounds and estimates for the corresponding classes of nonlinear composites. This variational procedure has been used by Ponte Castañeda (1991a,b, 1992) and deBotton and Ponte Castañeda (1992) to obtain estimates for the properties of isotropic particulate composites and anisotropic laminated and fiber-reinforced composites. Other contributions in the field of nonlinear composites include the work of Qiu and Weng (1991) dealing with the shape effect on the overall properties of two-phase elastic-plastic composite materials. They make use of an appropriate adaptation of the method of Mori and Tanaka (1973). In addition, a number of works have appeared recently (see, for example, Christman et al. 1989 and Bao et al. 1991) dealing with the effective properties of periodic composites; these being computed via finite-element analyses of the pertinent unit-cell, boundary-value problems. An alternative approach advocated by Duva (1984) (see also Duva and Hutchinson 1984), and used more recently by He (1990) and Lee & Mear (1991a,b), is to make use of the solution of a kernel problem, involving an inclusion in an infinite matrix of the nonlinear matrix material, to generate dilute estimates for the nonlinear composites. Generalizations to nondilute concentrations may be accomplished by the use of the differential self-consistent model (McLaughlin, 1977).

This paper deals with the effective constitutive behavior of nonlinear, incompressible composites that are reinforced by either aligned, or randomly oriented, rigid spheroidal inclusions. We concentrate on the effect of the inclusion shape on the effective behavior of the nonlinear composites. The method of analysis is based on Ponte Castañeda's (1991a) variational principle. For the case of aligned spheroidal inclusions with overall transversely isotropic symmetry, the method gives *explicit* expressions for the effective constitutive behavior of the nonlinear composites under *general* loading conditions (i.e., arbitrary combinations of axisymmetric tension, transverse shear and longitudinal shear). For the case of randomly oriented inclusions with overall isotropic symmetry, the method also gives *explicit* expressions for the effective behavior of the composites, but in this case, the composites being incompressible and isotropic, there is essentially only one loading mode.

2. VARIATIONAL CHARACTERIZATION OF EFFECTIVE PROPERTIES

Consider a general specimen of a nonlinear heterogeneous material occupying a domain Ω (of unit volume) with boundary $\partial\Omega$. The material is characterized by a stress potential $U(\mathbf{x}, \boldsymbol{\sigma})$, depending on position $\mathbf{x}$ and the stress field $\boldsymbol{\sigma}(\mathbf{x})$, and is such that the strain field $\boldsymbol{\epsilon}(\mathbf{x})$ is given by

$$\boldsymbol{\epsilon}(\mathbf{x}) = \frac{\partial U(\mathbf{x}, \boldsymbol{\sigma})}{\partial \boldsymbol{\sigma}}. \quad (1)$$

Then, the effective constitutive relation for the heterogeneous material may be defined, following Hill (1963), in terms of

$$\bar{\boldsymbol{\epsilon}} = \frac{\partial \tilde{U}(\bar{\boldsymbol{\sigma}})}{\partial \bar{\boldsymbol{\sigma}}}, \quad (2)$$

where $\bar{\boldsymbol{\epsilon}}$ and $\bar{\boldsymbol{\sigma}}$ are the mean values of the strain and stress fields over Ω , and where $\tilde{U}(\bar{\boldsymbol{\sigma}})$ denotes the effective stress potential of the composite, depending on the uniform stress

boundary condition

$$\mathbf{t}(\mathbf{x}) = \bar{\boldsymbol{\sigma}} \cdot \mathbf{n}, \quad \mathbf{x} \in \partial\Omega. \quad (3)$$

In this last relation, $\mathbf{t}$ is the traction vector and $\mathbf{n}$ is the outward normal to $\partial\Omega$.

The effective potential function $\tilde{U}(\bar{\boldsymbol{\sigma}})$ may be obtained directly from the principle of minimum complementary energy as

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma} \in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U(\mathbf{x}, \boldsymbol{\sigma}) dv, \quad (4)$$

where

$$S(\bar{\boldsymbol{\sigma}}) = \{\boldsymbol{\sigma} | \nabla \cdot \boldsymbol{\sigma} = 0 \text{ in } \Omega, \text{ and } \mathbf{t}(\mathbf{x}) = \bar{\boldsymbol{\sigma}} \cdot \mathbf{n}, \mathbf{x} \in \partial\Omega\} \quad (5)$$

represents the set of statically admissible stress fields satisfying the condition (3).

While the effective behavior of the composite is fully described by $\tilde{U}$ in terms of relation (2), the determination of $\tilde{U}$ from (4) and (5) presents real difficulties in that it requires the solution of a nonlinear boundary value problem with complex structure. Ponte Castañeda (1991a,b) introduced a variational principle that can be used to estimate the effective potential functions of nonlinear composites in terms of optimization problems involving the effective potential functions of appropriate classes of *linear* comparison composites. Thus, results for the effective properties of linear composites may be used to generate corresponding estimates for the effective properties of nonlinear composites. In this paper, we make use of the Hashin-Shtrikman bounds of Walpole (1969) and Willis (1977) for linear composites with spheroidal inclusions to estimate the effective constitutive behavior of ductile-matrix materials reinforced by rigid, spheroidal inclusions. Ponte Castañeda's variational principle is briefly reviewed in the following.

We consider only composites with incompressible, isotropic matrices. Thus, the potential

function for the nonlinear matrix may be written in the form

$$U(\mathbf{x}, \boldsymbol{\sigma}) = \phi(\mathbf{x}, \tau_e), \quad (6)$$

where

$$\tau_e = \sqrt{\frac{1}{2} \boldsymbol{\sigma}' \cdot \boldsymbol{\sigma}'} \quad (7)$$

is the effective shear stress and $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - \frac{\text{tr} \boldsymbol{\sigma}}{3} \mathbf{I}$ is the deviatoric stress tensor.

Then, under certain technical hypotheses (Ponte Castañeda, 1992), satisfied in this paper, we may express the matrix potential function in terms of the following optimization problem, namely,

$$U(\mathbf{x}, \boldsymbol{\sigma}) = \max_{\mu_0 \geq 0} \{U_0(\mathbf{x}, \boldsymbol{\sigma}) - V(\mathbf{x}, \mu_0)\}, \quad (8)$$

where U_0 is the potential function of an incompressible, linear-elastic comparison material with shear modulus $\mu_0(\mathbf{x})$, such that $U_0(\mathbf{x}, \boldsymbol{\sigma}) = \frac{1}{2\mu_0(\mathbf{x})} \tau_e^2$, and where

$$V(\mathbf{x}, \mu_0) = \max_{\boldsymbol{\sigma}} \{U_0(\mathbf{x}, \boldsymbol{\sigma}) - U(\mathbf{x}, \boldsymbol{\sigma})\}. \quad (9)$$

Then, the effective potential function of a given nonlinear composite is obtained by averaging relation (8); the result is (Ponte Castañeda, 1992)

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \max_{\mu_0(\mathbf{x}) \geq 0} \left\{ \tilde{U}_0(\bar{\boldsymbol{\sigma}}) - \int_{\Omega} V(\mathbf{x}, \mu_0(\mathbf{x})) dv \right\}, \quad (10)$$

where $\tilde{U}_0(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma} \in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U_0(\mathbf{x}, \boldsymbol{\sigma}) dv$ is the effective potential of the linear comparison composite.

Even though (10) is an exact expression for the effective potential function of a given nonlinear composite, it involves an infinite-dimensional optimization problem over the set of non-negative functions $\mu_0(\mathbf{x})$. Thus, in general, the variational principle (10) is at least

as difficult to implement as the classical variational principle (4), except for some very special microstructures (e.g., laminated composites; see deBotton & Ponte Castañeda 1992). However, it may be shown that (10) can be utilized in an approximate fashion to generate bounds for the effective properties of classes of nonlinear composites with more general microstructures. This is discussed next.

We begin by specializing the general class of composites considered above to the class of two-phase composites. Each phase is assumed to be incompressible, isotropic and homogeneous, characterized by a convex potential function $U^{(r)}(\boldsymbol{\sigma}) = \phi^{(r)}(\tau_e)$ ($r = 1$ or 2 , for phase 1 or 2). Further, the volume fractions of each phase, denoted by $c^{(r)}$ ($c^{(1)} + c^{(2)} = 1$ for a two-phase composite), are also assumed to be known. Even then, the effective potential function given by (10) requires the solution of an infinite dimensional optimization problem over the set of functions $\mu_0(\mathbf{x})$. However, by restricting our attention to the class of piecewise constant $\mu_0(\mathbf{x})$ (constant in each phase of the nonlinear material), we arrive at the following lower bound approximation for the effective potential function of the nonlinear composite, namely,

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) \geq \max_{\mu_0^{(1)}, \mu_0^{(2)} \geq 0} \left\{ \tilde{U}_0(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^2 c^{(r)} V^{(r)}(\mu_0^{(r)}) \right\}, \quad (11)$$

where $\mu_0^{(1)}$ and $\mu_0^{(2)}$ are the values of $\mu_0(\mathbf{x})$ in phases 1 and 2, respectively. Also, $\tilde{U}_0(\bar{\boldsymbol{\sigma}})$ is now the effective potential function of a linear comparison composite with *precisely* the same microstructure as the nonlinear composite, and

$$V^{(r)}(\mu_0^{(r)}) = \max_{\tau_e} \left\{ \frac{1}{2\mu_0^{(r)}} \tau_e^2 - \phi^{(r)}(\tau_e) \right\}. \quad (12)$$

We note that the inequality in (11) arises because the maximum is now evaluated over a strictly smaller set of functions (i.e., the set of piecewise constant functions).

The above inequality is useful because the optimization problem involved is two-dimensional, and therefore simpler to evaluate, assuming that we have estimates for the effective potential of two-phase, linear composites. In fact, if we have a lower bound for the effective potential of the linear composites $\tilde{U}_0(\bar{\sigma})$, then (11) generates a lower bound for the effective potential of the nonlinear composite. If, on the other hand, we have an upper bound for $\tilde{U}_0(\bar{\sigma})$, (11) generates only an estimate for the upper bound, which is generally lower than the true upper bound.

3. NONLINEAR ESTIMATES FOR COMPOSITES WITH RIGID INCLUSIONS

In this paper, we study composites made up of an incompressible, isotropic matrix material reinforced by either aligned, or randomly oriented, rigid spheroidal inclusions, as depicted in Figure 1. The shape of a typical inclusion is characterized by its aspect ratio $\alpha = b/a$. Thus, oblate spheroids have $\alpha < 1$, prolate spheroids have $\alpha > 1$ and spheres have $\alpha = 1$. The stress potential for the rigid inclusions (phase 2) vanishes identically (i.e., $\phi^{(2)}(\tau_e) = 0$). Then, to be able to make use of (11), we need to obtain estimates for the effective potentials $\tilde{U}_0$ of linear composites with rigid, spheroidal inclusions. Such results may be obtained directly from the work of Walpole (1966a,b, 1969) and Willis (1977) (see also Willis 1982, and Qiu and Weng 1990). Although these authors are concerned with both general bounds of the Hashin-Shtrikman type and self-consistent estimates, here we will make use only of the appropriate (non-trivial) lower bounds for the effective shear modulus. It is known (see, for example, Milton and Kohn 1988) that these lower bounds correspond to the least stiff isotropic composites which may be made with fixed volume fractions of the phases. Further, these extremal composites have particulate microstructures, where the inclusions are made

of the rigid phase. Thus, we may interpret the result of implementing these non-trivial Hashin-Shtrikman bounds into (11) as (upper) estimates for the effective potential functions of the corresponding nonlinear composites. We begin by considering the case of composites with aligned inclusions, and consider the case of randomly oriented inclusions later.

3.1 Aligned spheroidal inclusions

In this section, we consider composite materials with nonlinear, incompressible matrices and aligned rigid spheroidal inclusions. We note that this case was also briefly considered by Talbot and Willis (1992), although explicit results of the type obtained here were only determined for the special case of aligned rigid disks. While each phase of the composite is isotropic, the geometry of the inclusions is such that the composite exhibits overall transversely isotropic symmetry. The direction of inclusion alignment provides the axis of transverse isotropy (see Figure 1(a), where $\mathbf{x}_1$ defines the axis of symmetry).

In this case, an upper bound for the effective stress potential of the linear comparison material $\tilde{U}_0$ may be obtained from the previously mentioned works (see Appendix A). The result may be written

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\mu^{(1)}} \left(\bar{\tau}^{(1)} \right)^2, \quad (13)$$

where

$$\bar{\tau}^{(1)} = \sqrt{\frac{\mu^{(1)}}{\tilde{\mu}_d} \bar{\tau}_d^2 + \frac{\mu^{(1)}}{\tilde{\mu}_p} \bar{\tau}_p^2 + \frac{\mu^{(1)}}{\tilde{\mu}_n} \bar{\tau}_n^2}, \quad (14)$$

Here $\mu^{(1)}$ is the shear modulus of the incompressible, isotropic matrix material, $\frac{\mu^{(1)}}{\tilde{\mu}_d}$, $\frac{\mu^{(1)}}{\tilde{\mu}_p}$ and $\frac{\mu^{(1)}}{\tilde{\mu}_n}$ are given by relations (53) in Appendix A, and $\bar{\tau}_d$, $\bar{\tau}_p$ and $\bar{\tau}_n$ (note that $\bar{\tau}_e = (\bar{\tau}_d^2 + \bar{\tau}_p^2 + \bar{\tau}_n^2)^{1/2}$) are the three transversely isotropic invariants of the applied stress tensor $\bar{\sigma}$, corresponding to the three independent deformation modes for an incompressible, transversely isotropic

material: axisymmetric deformation, transverse shear and longitudinal shear, respectively (see Appendix B).

Using the upper bound (13) for the effective potential of the linear comparison material $\tilde{U}_0$, we can compute, via relation (11), an upper estimate for the effective stress potential of the nonlinear composite $\tilde{U}$. The result is

$$\tilde{U}(\bar{\sigma}) \cong \max_{\mu^{(1)}} \left\{ \frac{1}{2\mu^{(1)}} (\bar{\tau}^{(1)})^2 - c^{(1)} \max_{\tau_e} \left[\frac{1}{2\mu^{(1)}} \tau_e^2 - \phi^{(1)}(\tau_e) \right] \right\}, \quad (15)$$

which can be shown (by interchanging the maxima operations) to reduce to

$$\tilde{U}(\bar{\sigma}) \cong c^{(1)} \phi^{(1)} \left(\frac{\bar{\tau}^{(1)}}{\sqrt{c^{(1)}}} \right). \quad (16)$$

This relation expresses the effective potential function of the nonlinear composite in terms of the stress potential of the nonlinear matrix material $\phi^{(1)}$, the matrix volume fraction $c^{(1)}$, and an estimate for the average stress in the matrix $\bar{\tau}^{(1)}/\sqrt{c^{(1)}}$ (which in turn depends on the appropriate invariants of $\bar{\sigma}$, on the matrix volume fraction and the aspect ratio of the inclusions α). We emphasize that the above result is meant to be interpreted as an *estimate* (not an exact result) for the (weakest) effective behavior of the class of composites under consideration.

Corresponding to the three invariants of the stress tensor $\bar{\sigma}$, we can identify three invariants of the strain tensor $\bar{\epsilon}$: the deviatoric shear strain $\bar{\gamma}_d$, the transverse shear strain $\bar{\gamma}_p$, and the longitudinal shear strain $\bar{\gamma}_n$ (such that $\bar{\epsilon}_e = 1/\sqrt{3}(\bar{\gamma}_d^2 + \bar{\gamma}_p^2 + \bar{\gamma}_n^2)^{1/2}$; see Appendix B). Then, the effective stress-strain relations for the nonlinear composite may be expressed in the form

$$\bar{\gamma}_d = c^{(1)} \frac{\mu^{(1)}}{\tilde{\mu}_d} \frac{\bar{\tau}_d}{\bar{\tau}^{(1)}} \frac{\partial \phi^{(1)}}{\partial \bar{\tau}^{(1)}},$$

$$\begin{aligned}\bar{\gamma}_p &= c^{(1)} \frac{\mu^{(1)}}{\tilde{\mu}_p} \frac{\bar{\tau}_p}{\bar{\tau}^{(1)}} \frac{\partial \phi^{(1)}}{\partial \bar{\tau}^{(1)}} \quad , \\ \bar{\gamma}_n &= c^{(1)} \frac{\mu^{(1)}}{\tilde{\mu}_n} \frac{\bar{\tau}_n}{\bar{\tau}^{(1)}} \frac{\partial \phi^{(1)}}{\partial \bar{\tau}^{(1)}} \quad .\end{aligned}\tag{17}$$

It is seen from these stress-strain relations that, for the nonlinear composite, all three deformation modes are coupled with each other (for example, $\bar{\gamma}_d$ depends on $\bar{\tau}_d, \bar{\tau}_p$ and $\bar{\tau}_n$ through $\partial \phi^{(1)} / \partial \bar{\tau}^{(1)}$). This is in contrast with the corresponding linear composite (see (54) of Appendix A).

So far we have derived effective constitutive relations for a nonlinear matrix material reinforced by aligned rigid spheroidal inclusions in prescribed volume fraction and given aspect ratio of the inclusions. Also, unlike the results of other investigations dealing exclusively with axisymmetric loading conditions (see Bao et al. 1991, and Lee and Mear 1991a,b), these results apply for arbitrary loading conditions. In the next subsections, we specialize these general results to some special cases of theoretical and practical importance. For example, the limit $\alpha \rightarrow \infty$, with finite inclusion concentration, results in a composite with infinitely long fibers (continuous reinforcement). On the other hand, the opposite limit as $\alpha \rightarrow 0$, with finite inclusion concentration, corresponds to a laminated composite material (another case of continuous reinforcement). The result for $\alpha = 1$ corresponds to spherical inclusions (particulate reinforcements). The last two cases have been studied in some detail by Ponte Castañeda (1991a,b) and deBotton and Ponte Castañeda (1992).

3.1.1 Fiber-reinforced composites

In the limit as $\alpha \rightarrow \infty$, for a fixed, finite concentration of inclusions, an asymptotic expansion for $\bar{\tau}^{(1)}$ can be obtained explicitly from (14) and (53). The leading order term is

$$\bar{\tau}^{(1)} = \sqrt{\left(1 - \frac{2c^{(2)}}{1 + c^{(2)}}\right) (\bar{\tau}_p^2 + \bar{\tau}_n^2)}. \quad (18)$$

Correspondingly, the effective potential of the nonlinear composite reduces to

$$\tilde{U}(\bar{\sigma}) = c^{(1)} \phi^{(1)} \left(\sqrt{\frac{1}{c^{(1)}} \left(1 - \frac{2c^{(2)}}{1 + c^{(2)}}\right) (\bar{\tau}_p^2 + \bar{\tau}_n^2)} \right). \quad (19)$$

This equation shows that the response of the nonlinear composite to transverse and longitudinal shear is the same. On the other hand, the composite is effectively rigid under axisymmetric loading. This is physically reasonable since for a finite volume fraction of the inclusions, the inclusions must become infinitely long as $\alpha \rightarrow \infty$. Thus, the fiber-reinforced composite has just two effective deformation modes (instead of three). We note that as the inclusion concentration $c^{(2)} \rightarrow 0$ in (19), the potential function is still dependent only on the shear loading modes.

3.1.2 Needle-reinforced composites

For the fiber-reinforced composite discussed above, the continuity of the rigid fibers severely restricted the modes of deformation possible. We consider next rigid spheroidal inclusions with arbitrarily large aspect ratios but with finite length. These aligned thin needles, each of length $2b$ and radius $a = \varepsilon b$ (with $\varepsilon = 1/\alpha$), are depicted in Figure 1. If the number density per unit volume of these identical needles is n_2 , then (see Willis 1982) their volume fraction may be expressed in the form

$$c^{(2)} = \frac{4}{3} \pi n_2 b^3 \varepsilon^2. \quad (20)$$

We note that, for a composite with fixed number density n_2 , the concentration $c^{(2)} \rightarrow 0$ as $\alpha \rightarrow \infty$ ($\epsilon \rightarrow 0$). The "axial concentration" of these needles is represented by $n_2 b^3$.

Taking a fixed axial concentration $n_2 b^3$, an asymptotic expression for the effective shear stress in the matrix $\bar{\tau}^{(1)}$ is given by

$$\bar{\tau}^{(1)} = \sqrt{\left(1 + \frac{4n_2 b^3 \pi}{9 \ln \epsilon}\right) \bar{\tau}_d^2 + \bar{\tau}_p^2 + \bar{\tau}_n^2}. \quad (21)$$

The corresponding effective stress potential for this nonlinear composite material is then given by stress (21) by

$$\tilde{U}(\bar{\sigma}) = \phi^{(1)} \left(\sqrt{\left(1 + \frac{4n_2 b^3 \pi}{9 \ln \epsilon}\right) \bar{\tau}_d^2 + \bar{\tau}_p^2 + \bar{\tau}_n^2} \right). \quad (22)$$

This effective stress potential is dependent on all three deformation modes—which is consistent with discontinuous reinforcement. Note that in the limit as $\alpha \rightarrow \infty$ ($\epsilon \rightarrow 0$), $\tilde{U}(\bar{\sigma}) \rightarrow \phi^{(1)}(\bar{\tau}_e)$, corresponding to the behavior of the matrix material. This is in sharp contrast with the continuous fiber-reinforced composite, discussed in the previous subsection, which remains rigid with respect to axisymmetric loading even as the concentration $c^{(2)} \rightarrow 0$. This is a consequence of the fact that the limit $\alpha \rightarrow \infty$ is singular (the limit depends on whether $c^{(2)}$ is taken to be finite or zero).

3.1.3 Laminated composites

In the limit as $\alpha \rightarrow 0$, with a fixed, finite inclusion concentration, we have, asymptotically, that

$$\bar{\tau}^{(1)} = \bar{\tau}_n, \quad (23)$$

which results in an effective stress potential

$$\tilde{U}(\bar{\sigma}) = c^{(1)} \phi^{(1)} \left(\frac{\bar{\tau}_n}{\sqrt{c^{(1)}}} \right). \quad (24)$$

This behavior is consistent with the behavior of a two-phase laminated composite with one rigid phase since the only mode of deformation possible in this case is shear along the plane of the layers. Thus, for fixed inclusion concentration, the above result is in agreement with the exact solution for multiple-phase laminated composites, derived by de Botton & Ponte Castañeda(1992).

3.1.4 Disk-strengthened composites

Next, we consider spheroidal inclusions which have radius a and height $2\alpha a$. As $\alpha \rightarrow 0$ the inclusions become flat disks. If the number density per unit volume is n_2 , then the volume fraction of the disks is

$$c^{(2)} = \frac{4}{3}\pi n_2 a^3 \alpha, \quad (25)$$

where $n_2 a^3$ represents the “transverse concentration” of disks.

The effective shear stress $\bar{\tau}^{(1)}$ for thin disks is obtained by substituting (25) into (14) and taking the limit (see Willis 1982) as $\alpha \rightarrow 0$, with n_2 fixed (note that $c^{(2)} \rightarrow 0$). Then, the leading order term in an asymptotic expansion as $\alpha \rightarrow 0$ is given by

$$\bar{\tau}^{(1)} = \sqrt{\left(1 - \frac{16n_2 a^3/9}{1 + 16n_2 a^3/9}\right) \bar{\tau}_d^2 + \left(1 - \frac{32n_2 a^3/9}{1 + 32n_2 a^3/9}\right) \bar{\tau}_p^2 + \bar{\tau}_n^2}. \quad (26)$$

The corresponding effective potential for the composite with thin disks is obtained by substituting (26) into (16), which leads to

$$\tilde{U}(\bar{\sigma}) = \phi^{(1)} \left(\sqrt{\left(1 - \frac{16n_2 a^3/9}{1 + 16n_2 a^3/9}\right) \bar{\tau}_d^2 + \left(1 - \frac{32n_2 a^3/9}{1 + 32n_2 a^3/9}\right) \bar{\tau}_p^2 + \bar{\tau}_n^2} \right). \quad (27)$$

We emphasize that in this case, unlike the case of the needle-reinforced composite, the effect of the ‘flat’ rigid disks on the overall response of the nonlinear composite is nontrivial. We note that an expression analogous to the above relation (in terms of the strain potential instead of the stress potential) has been developed by Talbot and Willis (1992).

3.2 Randomly oriented inclusions

The composite materials considered in this section are made up of a nonlinear, incompressible matrix reinforced by rigid, randomly oriented, spheroidal inclusions, as shown in Figure 1(b). Thus, the composite is macroscopically isotropic and incompressible. Therefore, the effective stress potential $\tilde{U}$ depends only on the mean effective shear stress $\bar{\tau}_e$ (dependence on the third invariant is neglected: see Duva 1984). To obtain an upper estimate for $\tilde{U}$, we observe that $\phi^{(2)} = 0$, and we make use of the variational statement (11) with $\tilde{U}_0$ given by relations (58) in Appendix A. The result is

$$\tilde{U}(\bar{\sigma}) = \max_{\mu^{(1)}} \left\{ \frac{1}{2\tilde{\mu}} \bar{\tau}_e^2 - c^{(1)} \max_{\tau_e} \left[\frac{1}{2\mu^{(1)}} \tau_e^2 - \phi^{(1)}(\tau_e) \right] \right\}, \quad (28)$$

which reduces to

$$\tilde{U}(\bar{\sigma}) = c^{(1)} \phi^{(1)} \left(\sqrt{\frac{\mu^{(1)}}{c^{(1)} \tilde{\mu}}} \bar{\tau}_e \right), \quad (29)$$

where $\frac{\tilde{\mu}}{\mu^{(1)}}$ is given by (56) in Appendix A. This equation expresses the effective stress potential of the nonlinear composite in terms of the stress potential of the matrix material $\phi^{(1)}$, and an estimate for the average stress in the matrix (which in turn depends on the inclusion geometry and concentration). Explicit expressions for the special cases of needle-reinforced and disk-reinforced composites may be obtained in a similar way as for the corresponding special cases for the composites with aligned inclusions. However, in the interest of brevity, these will not be given here.

4. APPLICATION TO COMPOSITES WITH POWER-LAW MATRIX MATERIALS

So far we have derived expressions for the effective properties of composites comprised of a *general* nonlinear matrix material reinforced by rigid spheroidal inclusions, which are

either aligned or randomly oriented. In this section, we specialize the previous results to composites with pure power-law behavior for the matrix. This class of composites has also been considered recently by He (1990) and Lee & Mear (1991), who generalized the calculations of Duva (1984) for spherical rigid particles to spheroidal inclusions. As mentioned earlier, their method considers an isolated rigid spheroidal inclusion embedded in an infinite power-law matrix material, which is subjected to *axisymmetric* remote loading with respect to the spheroidal inclusion. In contrast, our results account approximately for inclusion interaction (without the need of the differential self-consistent scheme) and general remote loading conditions. Duva & Storm (1989) have also obtained dilute concentration results for the plane strain deformation of fiber composites reinforced by rigid fibers. A brief comparison of the results of this paper with the numerical results of these other authors is given in Appendix C. Numerical results for the axisymmetric response of power-law materials reinforced by aligned spheroidal inclusions, with periodic microstructures, have also been given by Bao et al. (1991). Comparison with these results will be given, as far as possible, in the following discussion.

4.1 *Composites with aligned inclusions*

The power-law matrix material is described by the stress potential

$$U^{(1)}(\boldsymbol{\sigma}) = \phi^{(1)}(\tau_e) = \frac{\sigma_0}{n+1} \left(\frac{\tau_e}{\tau_0} \right)^{n+1}, \quad (30)$$

where τ_0 is an appropriate stress normalization factor, which has the interpretation of the yield stress in shear in the limit as $n \rightarrow \infty$, and where $\sigma_0 = \sqrt{3}\tau_0$.

The effective stress potential for the nonlinear composite is obtained by substituting (30)

into (16). The results is

$$\tilde{U}(\bar{\sigma}) = (c^{(1)})^{(1-n)/2} \frac{\sigma_0}{n+1} \left(\frac{\bar{\tau}^{(1)}}{\tau_0} \right)^{n+1}, \quad (31)$$

where $\bar{\tau}^{(1)}$ is given by (14). The corresponding stress-strain relations are

$$\begin{aligned} \bar{\gamma}_d &= \sqrt{3} (c^{(1)})^{(1-n)/2} \frac{\mu^{(1)}}{\tilde{\mu}_d} \left(\frac{\bar{\tau}^{(1)}}{\tau_0} \right)^{n-1} \frac{\bar{\tau}_d}{\tau_0}, \\ \bar{\gamma}_p &= \sqrt{3} (c^{(1)})^{(1-n)/2} \frac{\mu^{(1)}}{\tilde{\mu}_p} \left(\frac{\bar{\tau}^{(1)}}{\tau_0} \right)^{n-1} \frac{\bar{\tau}_p}{\tau_0}, \\ \bar{\gamma}_n &= \sqrt{3} (c^{(1)})^{(1-n)/2} \frac{\mu^{(1)}}{\tilde{\mu}_n} \left(\frac{\bar{\tau}^{(1)}}{\tau_0} \right)^{n-1} \frac{\bar{\tau}_n}{\tau_0}. \end{aligned} \quad (32)$$

These general results will be studied through several specific examples in the following subsections.

4.1.1 *Effective behavior under single-mode loading*

We study each of the three independent deformation modes of the incompressible, transversely isotropic composite first. If only axisymmetric loading $\bar{\tau}_d$ is applied to the composite, then equation (14) reduces to

$$\bar{\tau}^{(1)} = \left(\frac{\mu^{(1)}}{\tilde{\mu}_d} \right)^{1/2} \bar{\tau}_d. \quad (33)$$

Thus, the effective potential of the nonlinear composite $\tilde{U}$ may be written in the form

$$\tilde{U}(\bar{\sigma}) = \frac{(\tilde{\sigma}_0)_d}{n+1} \left(\frac{\bar{\tau}_d}{(\tilde{\tau}_0)_d} \right)^{n+1}, \quad (34)$$

where $(\tilde{\tau}_0)_d$ and $(\tilde{\sigma}_0)_d$ are the stress normalization factors in shear and tension, respectively, for axisymmetric loading, and are given by

$$\frac{\tau_0}{(\tilde{\tau}_0)_d} = \frac{\sigma_0}{(\tilde{\sigma}_0)_d} = (c^{(1)})^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}_d} \right)^{(n+1)/2n}. \quad (35)$$

Here $\frac{\mu^{(1)}}{\tilde{\mu}_d}$ is given by (53). We note that as $n \rightarrow \infty$, $(\tilde{\sigma}_0)_d$ corresponds to the tensile flow stress of the composite under axisymmetric loading.

Similar results may be derived for transverse and longitudinal shear loading with $(\tilde{\tau}_0)_p$ and $(\tilde{\tau}_0)_n$ as the effective stress normalization factors in transverse and longitudinal shear, respectively. These may be expressed by the relations

$$\frac{\tau_0}{(\tilde{\tau}_0)_p} = (c^{(1)})^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}_p} \right)^{(n+1)/2n}, \quad (36)$$

$$\frac{\tau_0}{(\tilde{\tau}_0)_n} = (c^{(1)})^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}_n} \right)^{(n+1)/2n}. \quad (37)$$

In Figures 2 to 4, we show plots of the inverse of these three effective stress normalization factors for a linear ($n = 1$) and nonlinear ($n = 10$) matrix material, respectively, as functions of the inclusion concentration. The composites presented in these figures have inclusions with shapes that vary from oblate ($\alpha < 1$) to prolate ($\alpha > 1$) spheroids. We discuss next the effect of five different inclusion shapes ($\alpha = 0.01, 0.1, 1, 10, 50$) on the effective properties for the three different deformation modes.

Figures 2(a) and 2(b) show the effective behavior of composites with aligned rigid spheroidal inclusions under axisymmetric loading. It can be seen that both prolate and oblate spheroids give rise to stronger axisymmetric responses than spheres. We also observe that the response of prolate spheroids is relatively stiffer than that of oblate spheroids (e.g., needles with $\alpha = 50$ are stiffer than plates with $\alpha = 0.01$). Comparing Figures 2(a) and 2(b), it is evident that the effect of increasing material nonlinearity is to relax the strengthening influence of all particle shapes. For example, a linear composite ($n = 1$) containing a 10% volume fraction of prolate inclusions (with an aspect ratio $\alpha = 10$) is approximately 3.5 times as strong as

the matrix material. Similarly, if the inclusions have an aspect ratio $\alpha = 50$, the composite is approximately 50 times as strong as the matrix material. On the other hand, for a nonlinear composite ($n = 10$), these two inclusion shapes make the material approximately 2 and 6.6 times as strong as the matrix material, respectively. In the limit as $\alpha \rightarrow \infty$, the composite shows a rigid response ($(\tilde{\sigma}_0)_d \rightarrow \infty$) to axisymmetric loading. This is consistent with the behavior of composites reinforced by rigid fibers as discussed in Subsection 3.1.1. In the opposite limit as $\alpha \rightarrow 0$, $(\tilde{\sigma}_0)_d \rightarrow \infty$, which means that the composite also becomes rigid with respect to axisymmetric loading in this limit. This is consistent with the behavior of incompressible laminated materials reinforced by rigid layers as discussed in Subsection 3.1.3.

Figures 3(a) and 3(b) present the effective behavior of linear ($n = 1$) and nonlinear ($n = 10$) composites under transverse shear. It is found that the oblate spheroids ($\alpha = 0.01$) provide far greater strengthening than the prolate spheroids ($\alpha = 50$). This is because oblate spheroids have larger dimensions in the $\mathbf{x}_2 - \mathbf{x}_3$ plane (see Figure 1), and therefore resist this deformation mode more effectively. A nonlinear composite ($n = 10$) reinforced by 10% volume fraction of prolate inclusions (with $\alpha = 50$) is only 1.07 times as stiff as its matrix material, while, if it is reinforced by oblate inclusions (with $\alpha = 0.01$), it is 3.6 times as stiff as the matrix material. Also note that, a composite containing prolate inclusions with an aspect ratio $\alpha = 10$ has almost the same strength as a composite reinforced by prolate inclusions of aspect ratio $\alpha = 50$. As $\alpha \rightarrow \infty$, $(\tilde{\tau}_0)_p$ reaches a limit which almost coincides with its value at $\alpha = 50$. This limit inclusion shape provides a finite strengthening of the composite material, which is consistent with the behavior of rigid fiber-reinforced materials. In fact, prolate spheroids provide less stiffening than do spheres under transverse shearing.

In the opposite limit, as the inclusions become plate-like ($\alpha \rightarrow 0$), we obtain $(\tilde{\tau}_0)_p \rightarrow \infty$. The composite is thus rigid with respect to the transverse shear loading mode, which corresponds to the behavior of laminated materials reinforced by rigid layers. Finally, we note that under applied transverse shear loading, the fiber material can be treated as a plane strain problem. Duva & Storm (1989) have studied this plane strain problem numerically for the case of one rigid fiber in an infinite matrix. Comparing their dilute results with our results (as $\alpha \rightarrow \infty$), we find that there is little difference between the two sets of results. For example, if we compare the effective stress potential calculated by Duva & Storm with that of our explicit estimation, the difference is less than 5% for a nonlinear matrix (with $n = 10$), as discussed in some detail in Appendix C.

Figures 4(a) and 4(b) show the effect of rigid inclusions on the effective strength under longitudinal shear (τ_n) for a linear ($n = 1$) and a highly nonlinear matrix ($n = 10$). For this type of loading, we find that spherical inclusions provide the largest strengthening (of these five inclusion shapes) while oblate spheroids provide the least. It is interesting to note, that in the limit as $\alpha \rightarrow \infty$, the composite has the same strength under transverse shear loading (discussed in the previous paragraph) as under longitudinal shear ($(\tilde{\tau}_0)_n$ and $(\tilde{\tau}_0)_p$ reach the same limit). Since, for oblate inclusions, the longitudinal shear mode corresponds to a shearing deformation in a plane normal to their axis of symmetry, the strengthening effect of these rigid oblate spheroidal inclusions is significantly less for this deformation mode than for the other two deformation modes discussed above. In the limit of $\alpha \rightarrow 0$, the oblate inclusions become flat layers and the shearing deformation is concentrated along the matrix material between these rigid flat layers. This corresponds precisely to the response of laminated materials (with one phase rigid) which have only the longitudinal shear mode

available, as discussed in Section 3.1.3. Thus, of the five inclusion shapes studied here, oblate and prolate inclusion shapes provide less resistance to the longitudinal shear than spherical inclusions.

Figure 5(a) gives results for all three deformation modes (axisymmetric loading, transverse shear and longitudinal shear) in the perfectly-plastic limit ($n \rightarrow \infty$). For each loading mode, the ratio of the matrix flow stress to the effective flow stress ($\tau_0/(\tilde{\tau}_0)_d$, $\tau_0/(\tilde{\tau}_0)_p$, $\tau_0/(\tilde{\tau}_0)_n$) is plotted as a function of α , with a fixed inclusion concentration $c^{(2)} = 0.2$. For the axisymmetric deformation mode, prolate spheroids are stiffer than oblate spheroids if we compare a prolate shape with aspect ratio α with the oblate shape which has a reciprocal aspect ratio $1/\alpha$. The weakest reinforcement is given by the oblate shape with $\alpha \approx 0.64$, which is even less effective in increasing the axisymmetric strength than the spherical shape ($\alpha = 1$). On the other hand, for the transverse deformation mode, the largest reinforcement is given by the oblate shape with aspect ratio $\alpha = 0$. As α increases from 0 to ∞ , the strengthening effect decreases continuously. In the limit as $\alpha \rightarrow \infty$, the strength of the composite approaches the transverse shearing strength of fiber-reinforced composites. The reinforcement provided by rigid inclusions to the longitudinal shear mode is (generally) weaker than that for the other two deformation modes. As $\alpha \rightarrow 0$, the inclusions have no reinforcing effect on the flow strength of the composite. In the opposite limit as $\alpha \rightarrow \infty$, the longitudinal shear flow strength approaches that of the fiber-reinforced materials. It is noted that the strongest reinforcement for this deformation mode is given by a prolate shape with $\alpha \approx 1.4$.

Thus, for composites with aligned rigid spheroidal inclusions, there is no inclusion shape which provides optimal strengthening for all of the three independent deformation modes.

Needles give a stronger response (strongest overall) for one mode (axisymmetric mode), whereas disks provide significant reinforcement for two modes (axisymmetric mode and transverse shear mode). Thus, the optimal choice of particle shape in a given application depends on the particular service load to which the composite will be subjected.

Bao et al. (1991) have carried out a study of the tensile flow stress of a two-phase composite with periodic microstructures using unit-cell, finite-element calculations. While the matrix material, inclusion shape and loading mode utilized by these authors are identical to those utilized in this work, the distribution of inclusions in their work is periodic. With this difference in mind, we compare the effect of spheroidal inclusion shape on the axisymmetric tensile flow stress of composites with inclusion concentration $c^{(2)} = 0.1$ (our estimates may be obtained from Figure 5(b)). For rigid spherical inclusions in a perfectly plastic matrix ($n \rightarrow \infty$), the effective tensile flow stress predicted by our method is $\tilde{\sigma}_0 / \sigma_0 = 1.0695$, while the corresponding result of Bao et al. (for a periodic microstructure) is $\tilde{\sigma}_0 / \sigma_0 = 1.025$ (from the curves in Figure 6 of their paper). For an aspect ratio $\alpha = 0.1$ (or 10), the tensile flow stresses shown in Figure 5(b) is $\tilde{\sigma}_0 / \sigma_0 = 1.2$ (or 1.742). The corresponding result of Bao et al. is $\tilde{\sigma}_0 / \sigma_0 = 1.36$ (or 1.62) (from the curves in Figure 6 of their paper). Thus the predictions of both methods are in qualitative agreement for these three inclusion shapes.

Finally, we note, for completeness, that for the special case of disk-reinforced composites we obtain the following simple expressions for the effective stress normalization factors in the three different modes:

$$\frac{\tau_0}{(\tilde{\tau}_0)_d} = \left(\frac{1}{1 + 16n_2 a^3 / 9} \right)^{(n+1)/2n}, \quad (38)$$

$$\frac{\tau_0}{(\tilde{\tau}_0)_p} = \left(\frac{1}{1 + 32n_2 a^3 / 9} \right)^{(n+1)/2n}, \quad (39)$$

$$\frac{\tau_0}{(\tilde{\tau}_0)_n} = 1. \quad (40)$$

We observe that the longitudinal shear stress normalization factor (40) is unity indicating that the longitudinal shear mode is not affected by the presence of thin disk-like inclusions. On the other hand, the axisymmetric and transverse shear modes (38 and 39) are both sensitive to the presence of thin disks through the parameter $n_2 a^3$. If this parameter $n_2 a^3 \rightarrow \infty$, the material becomes effectively rigid under these two modes of deformation, as expected. Corresponding results can also be easily written down for needle-reinforced composites.

4.1.2 *Effective behavior under multiple-mode loading*

An interesting difference between the effective behavior of linear and nonlinear composite materials is depicted in Figures 6 and 7, where the coupling between different deformation modes is analyzed for the nonlinear composites. As is well-known, the effective response of linear, incompressible fiber-reinforced composites with transversely isotropic symmetry is such that there is no coupling between the three independent modes of deformation available for such materials (see (52)). However, this is not the case for nonlinear materials. For example, if we preload a nonlinear composite (matrix with $n = 10$ and aligned prolate spheroids with $\alpha = 10$ and $c^{(2)} = 0.2$) with axisymmetric stresses $\bar{\tau}_d/\tau_0 = 0.1, 2.8$ and 3.5 , the deformation of the nonlinear composite is correspondingly characterized by strains $\bar{\gamma}_d$ as given by (32) with (33). If we now increase the transverse shear stress $\bar{\tau}_p$, we observe (Figure 6) a significant further increase in the axisymmetric strain $\bar{\gamma}_d$. By comparison, the corresponding linear composite would not show further increase of the axisymmetric strain $\bar{\gamma}_d$ under increasing transverse shear stress $\bar{\tau}_p$. This example clearly illustrates the significant coupling between the different loading modes for nonlinear composites.

Another example is given in Figure 7, which shows plots the axisymmetric stress $\bar{\tau}_d$ versus

the axisymmetric shear strain $\bar{\gamma}_d$ for the same composite discussed above. In this figure, however, the composite has been preloaded with three different values of the transverse shear stress ($\bar{\tau}_p/\tau_0 = 0.0, 1.0, 1.5$). We observe that the effect of the transverse shear preload is to significantly reduce the axisymmetric load carrying capability of the composite.

4.2 Composites with randomly oriented inclusions

In the previous section, we observed that, for aligned spheroidal inclusions, there is no aligned rigid spheroidal inclusion shape which provides optimal strengthening for all deformations. In this section, we study the shape effect for randomly oriented inclusions. By substituting the general power-law material description (30) into the effective potential for nonlinear materials reinforced with randomly oriented rigid spheroids (29), this effective stress potential may be written in the form

$$\tilde{U}(\bar{\sigma}) = \frac{\tilde{\sigma}_0}{n+1} \left(\frac{\bar{\sigma}_e}{\tilde{\sigma}_0} \right)^{n+1}, \quad (41)$$

where $\bar{\sigma}_e (= \sqrt{3}\bar{\tau}_e)$ is the effective average stress, and where the effective stress normalization factor $\tilde{\sigma}_0$ is given by

$$\frac{\sigma_0}{\tilde{\sigma}_0} = \left(c^{(1)} \right)^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}} \right)^{(n+1)/2n}, \quad (42)$$

with $\tilde{\mu}/\mu^{(1)}$ given in turn by (56).

Figures 8 show plots $\sigma_0/\tilde{\sigma}_0$ versus inclusion concentration $c^{(2)}$ for a linear ($n = 1$) and a strongly nonlinear composites ($n = 10$) with randomly oriented rigid spheroidal inclusions. It can be seen that prolate spheroids with $\alpha = 50$ are close to oblate spheroids with $\alpha = 0.01$ (the prolate spheroids are slightly stiffer). This similarity in behavior is in marked contrast with the very different behaviors displayed by aligned prolate and oblate spheroids under different loading modes. Also, while the spherical inclusions result in the least strengthening

for a given concentration in these isotropic materials, there are loading modes (see Figure 4) for which spherical inclusions produce stronger effective behavior than aligned prolate or oblate spheroids (at a given concentration).

A study on the tensile flow stress of a composite reinforced by rigid, randomly oriented, packet morphologies has been carried out by Bao et al. (1991) using finite element calculations. While it is not possible to make direct comparisons of our results with theirs, there are qualitative similarities in some of the findings. For example, we find, just like Bao et al., that the inclusion shape with $\alpha = 10$ provides stronger reinforcement than the inclusion shape with $\alpha = 0.1$.

It is interesting to examine the limits as $\alpha \rightarrow 0$ and $\alpha \rightarrow \infty$, with a fixed, *finite* inclusion concentration $c^{(2)}$. From (42), we obtain, respectively, the following effective stress normalization factors

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim (c^{(1)})^{1/n} \left(\frac{3\pi\alpha}{4c^{(2)}} \right)^{(n+1)/2n}, \quad \text{as } \alpha \rightarrow 0; \quad (43)$$

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim (c^{(1)})^{1/n} \left(\frac{15 \ln(2\alpha)}{c^{(2)}\alpha^2} \right)^{(n+1)/2n}, \quad \text{as } \alpha \rightarrow \infty. \quad (44)$$

We observe that, in both of these limits, the stress normalization factor $\tilde{\sigma}_0$ becomes unbounded. As discussed in Section 3, these two limits correspond to rigid fibers and rigid flat layers, respectively. Thus, in these limits, it is clear that an orientational average will lead to perfectly rigid behavior for the composite. A more sensible approach is to consider the limits as $\alpha \rightarrow 0$ and $\alpha \rightarrow \infty$, with fixed density of the inclusions (n_2). These limits correspond to the cases of randomly oriented disks and needles, respectively. Thus the effective stress normalization factor for the case of thin needles (with aspect ratio α) is given by

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim \left[1 - \frac{2\pi n_2 b^3/3}{5(1 + 6 \ln(2\alpha)) + 2\pi n_2 b^3/3} \right]^{(n+1)/2n}, \quad (45)$$

and the corresponding factor for disks is given by

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim \left(1 - \frac{16n_2a^3}{9 + 16n_2a^3}\right)^{(n+1)/2n}. \quad (46)$$

We note that infinitely thin needles provide no reinforcement, whereas infinitely thin disks do provide some finite reinforcement (depending on the density of disks).

5. CONCLUSIONS

In this paper, we have studied the effective constitutive behavior of composites made up of incompressible, ductile materials reinforced by either aligned, or randomly oriented, rigid spheroidal inclusions. The method of solution is based on the variational principle of Ponte Castañeda(1991a) for nonlinear inhomogeneous media. When the rigid inclusions are aligned, the resulting composites are transversely isotropic (and incompressible), and characterized by three independent modes of deformation – axisymmetric tension, transverse shear and longitudinal shear. When the rigid inclusions are randomly oriented, the resulting composites are isotropic (and incompressible), and are (approximately) characterized by one deformation mode (e.g., tensile loading).

For the aligned-spheroid composites, the axisymmetric loading mode has been studied extensively in the recent past. However, the transverse shear and longitudinal shear modes of deformation for these composites have hardly received any attention. In this work, we have obtained *explicit* expressions for the constitutive response of these composites under *general* loading conditions (including transverse and longitudinal shear loading), and for finite (nondilute) concentrations of the inclusions. In addition, simple expressions have been obtained for the special cases of aligned rigid needles and disks. The results show that the

optimal choice for the shape of the particle reinforcement, within this class of composites, depends strongly on the service load to which the composite will be subjected. We also note that the nonlinear aligned-spheroid composites exhibit strong coupling between the three possible modes of deformation for these composites. By comparison, such coupling is not present for the corresponding linear composites. For composites reinforced by randomly oriented spheroidal inclusions, we find that both the prolate and oblate spheroidal inclusions provide significant reinforcement. We conclude by noting that more complicated microstructures than the ones considered in this work may eventually lead to metal-matrix composites with superior effective properties. We hope that the method developed in this work will serve as a useful tool in the optimal design of such microstructures.

Acknowledgements

This research has been supported by the Air Force Office of Scientific Research under Grant No. 91-0161. The first author would like to acknowledge valuable discussions with K. Sarkar.

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APPENDIX A: LINEAR COMPOSITES WITH RIGID SPHEROIDAL INCLUSIONS

A.1 Composites with aligned inclusions

The results given here are based on the works of Hill (1965), Walpole (1966a,b, 1969) and Willis (1977) (see also Willis 1982 and Qiu and Weng 1990) on linear, anisotropic composite materials. Consider a two-phase composite material, as depicted in Figure 1, in which the spheroidal inclusions are either aligned in x_1 direction or randomly oriented. The matrix and the stiffer inclusion materials are labeled phase 1 and 2, respectively. Also, the volume fraction of phase 1 and phase 2 are denoted by $c^{(1)}$ and $c^{(2)}$ ($c^{(1)} + c^{(2)} = 1$). In the following discussion, fourth-order symmetric tensors will be denoted by upper-case italicized Roman letters, while second order tensors will be denoted by bold-face, Greek letters.

The mechanical properties of each phase will be characterized by fourth-order compliance tensors, which, using Hill's convention, may be written in the form

$$M^{(r)} = \left(\frac{1}{3k^{(r)}}, \frac{1}{2\mu^{(r)}} \right), \quad (r = 1, 2), \quad (47)$$

where $k^{(r)}$ and $\mu^{(r)}$ are the bulk modulus and shear modulus, respectively, of the r th-phase material.

Then, the effective compliance tensor $\tilde{M}$ of the two-phase, linear composite may be expressed in the form

$$\tilde{M} = \sum_{r=1}^2 c^{(r)} M^{(r)} B^{(r)} \left\{ \sum_{s=1}^2 c^{(s)} B^{(s)} \right\}^{-1}, \quad (48)$$

where $B^{(r)}$ is the stress concentration factor tensor (Hill, 1965), given by

$$B^{(r)} = \left[I + \left(M^{(0)} \right)^{-1} (I - S^{(0)}) (M^{(r)} - M^{(0)}) \right]^{-1}, \quad (49)$$

In this last relation, $M^{(0)}$ is the compliance tensor of a comparison material, I is the fourth-order identity tensor, and $S^{(0)}$ is the Eshelby's tensor corresponding to a matrix made of the comparison material. For a transversely isotropic material, $S^{(0)}$ may be expressed, in Walpole's notation, as

$$S^{(0)} = (S_{2222}^{(0)} + S_{2233}^{(0)}, S_{1111}^{(0)}, 2S_{2323}^{(0)}, 2S_{1212}^{(0)}, S_{1122}^{(0)}, S_{2211}^{(0)}), \quad (50)$$

where the relevant components are given explicitly in Appendix B.

In order to obtain the required upper bound for the effective compliance (a lower bound for the stiffness) of the linear composite, the compliance of the comparison material $M^{(0)}$ in (48) and () must be chosen to be the less stiff phase (i.e., $M^{(0)} = M^{(1)}$). Then, after some algebra, (48) may be rewritten in the form

$$\tilde{M} = M^{(1)} + c^{(2)} \left[\left(M^{(2)} (M^{(1)})^{-1} - I \right)^{-1} + c^{(1)} (I - S^{(1)}) \right]^{-1} M^{(1)}, \quad (51)$$

where $S^{(1)}$ is the Eshelby tensor with phase-1 as the matrix material. Finally, since the inclusions are rigid (i.e., $k^{(2)} \rightarrow \infty$, and $\mu^{(2)} \rightarrow \infty$, so that $M^{(2)} = (0,0)$), and the matrix material is incompressible (i.e., $k^{(1)} \rightarrow \infty$), the transversely isotropic tensor, $\tilde{M}$, reduces to

$$\tilde{M} = \left(\frac{1}{2\tilde{\mu}_d}, \frac{1}{\tilde{\mu}_d}, \frac{1}{2\tilde{\mu}_p}, \frac{1}{2\tilde{\mu}_n}, -\frac{1}{2\tilde{\mu}_d}, -\frac{1}{2\tilde{\mu}_d} \right), \quad (52)$$

where

$$\begin{aligned} \frac{\mu^{(1)}}{\tilde{\mu}_d} &= 1 - \frac{c^{(2)}}{1 - c^{(1)} f(\alpha)}, \\ \frac{\mu^{(1)}}{\tilde{\mu}_p} &= 1 - \frac{c^{(2)}}{c^{(2)} + 2c^{(1)} S_{2323}^{(1)}}, \\ \frac{\mu^{(1)}}{\tilde{\mu}_n} &= 1 - \frac{c^{(2)}}{c^{(2)} + 2c^{(1)} S_{1212}^{(1)}}. \end{aligned} \quad (53)$$

Here f is a function of the inclusion aspect ratio α , and is given in Appendix B. Thus, the effective stress potential $\tilde{U}_0$ of the linear composite has the form (see Walpole 1969; deBotton & Ponte Castañeda 1992)

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\tilde{\mu}_d}\bar{\tau}_d^2 + \frac{1}{2\tilde{\mu}_p}\bar{\tau}_p^2 + \frac{1}{2\tilde{\mu}_n}\bar{\tau}_n^2, \quad (54)$$

where $\bar{\tau}_d, \bar{\tau}_p$ and $\bar{\tau}_n$ are the three transversely isotropic invariants of the applied stress tensor $\bar{\sigma}$, corresponding to the three independent deformation modes for an incompressible, transversely isotropic material: axisymmetric stress, transverse shear stress and longitudinal shear stress, respectively (see Appendix B).

A.2 Composites with randomly oriented inclusions

When the inclusions in the composite are randomly oriented, the expression for the effective compliance of the composite (48) must be replaced by (this is based on the orientation average of strain field, as in Wu (1966) and Walpole (1966b))

$$\tilde{M} = \left(c^{(1)}M^{(1)} + c^{(2)}M^{(2)}\{B^{(2)}\} \right) \left(c^{(1)}I + c^{(2)}\{B^{(2)}\} \right)^{-1}. \quad (55)$$

where $\{B^{(2)}\}$ denotes the orientational average of tensor $B^{(2)}$. We note that on account of the incompressibility and isotropy of the composite, we may write $\tilde{M} = \left(0, \frac{1}{2\tilde{\mu}} \right)$. Some complicated algebra leads to the following result for the effective shear modulus of the composite, appropriately normalized, namely,

$$\frac{\mu^{(1)}}{\tilde{\mu}} = \frac{5c^{(1)}(1 - e_s)(1 - f_s)[3 + 2(g_s + h_s) - (2d_s + c_s)]}{A_s(g_s + h_s) + B_se_sf_s + C_s(e_s + f_s) + D_s} \quad (56)$$

where

$$c_s = 1 - (S_{2222}^{(1)} + S_{2233}^{(1)}), \quad d_s = 1 - S_{1111}^{(1)},$$

$$\begin{aligned}
e_s &= 1 - 2S_{2323}^{(1)}, & f_s &= 1 - 2S_{1212}^{(1)}, \\
g_s &= -S_{1122}^{(1)}, & h_s &= -S_{2211}^{(1)}. \\
A_s &= 10c^{(1)}e_sf_s - 2(5 - 3c^{(2)})(e_s + f_s) + 2(5 - c^{(2)}), \\
B_s &= 3(5 - 4c^{(2)}) - 5c^{(1)}(2d_s + c_s), \\
C_s &= (5 - 3c^{(2)})(2d_s + c_s) - 3(5 - 2c^{(2)}), \\
D_s &= -(5 - c^{(2)})(2d_s + c_s) + 15.
\end{aligned} \tag{57}$$

Then, the corresponding potential function for the isotropic composite is

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\tilde{\mu}}\bar{\tau}_e^2. \tag{58}$$

APPENDIX B

Letting the $\mathbf{x}_1$ coordinate axis define the axis of transverse isotropy (see Figure 1(c)), the three transversely isotropic invariants of the stress tensor $\boldsymbol{\sigma}$ are given by

$$\begin{aligned}\tau_d^2 &= \frac{1}{3} \left[\frac{1}{2}(\sigma_{33} + \sigma_{22}) - \sigma_{11} \right]^2, \\ \tau_p^2 &= \sigma_{23}^2 + \frac{1}{4}(\sigma_{33} - \sigma_{22})^2, \\ \tau_n^2 &= \sigma_{12}^2 + \sigma_{13}^2.\end{aligned}\tag{59}$$

Correspondingly, the three transversely isotropic invariants of the strain tensor *strain* are given by

$$\begin{aligned}\gamma_d^2 &= \frac{4}{3} \left[\frac{1}{2}(\varepsilon_{33} + \varepsilon_{22}) - \varepsilon_{11} \right]^2, \\ \gamma_p^2 &= 4 \left[\varepsilon_{23}^2 + \frac{1}{4}(\varepsilon_{33} - \varepsilon_{22})^2 \right], \\ \gamma_n^2 &= 4(\varepsilon_{12}^2 + \varepsilon_{13}^2).\end{aligned}\tag{60}$$

Also, for a spheroidal inclusion, aligned with the $\mathbf{x}_1$ axis, the components of the Eshelby's tensor are (Eshelby, 1957)

$$\begin{aligned}S_{1111}^{(0)} &= \frac{1}{2(1-\nu^{(0)})} \left\{ 1 - 2\nu^{(0)} + \frac{3\alpha^2 - 1}{\alpha^2 - 1} - \left[1 - 2\nu^{(0)} + \frac{3\alpha^2}{\alpha^2 - 1} \right] g \right\}, \\ S_{2222}^{(0)} = S_{3333}^{(0)} &= \frac{3}{8(1-\nu^{(0)})} \frac{\alpha^2}{\alpha^2 - 1} + \frac{1}{4(1-\nu^{(0)})} \left[1 - 2\nu^{(0)} - \frac{9}{4(\alpha^2 - 1)} \right] g, \\ S_{2233}^{(0)} = S_{3322}^{(0)} &= \frac{1}{4(1-\nu^{(0)})} \left\{ \frac{\alpha^2}{2(\alpha^2 - 1)} - \left[1 - 2\nu^{(0)} + \frac{3}{4(\alpha^2 - 1)} \right] g \right\}, \\ S_{2211}^{(0)} = S_{3311}^{(0)} &= -\frac{1}{2(1-\nu^{(0)})} \frac{\alpha^2}{\alpha^2 - 1} + \frac{1}{4(1-\nu^{(0)})} \left\{ \frac{3\alpha^2}{\alpha^2 - 1} - (1 - 2\nu^{(0)}) \right\} g, \\ S_{1122}^{(0)} = S_{1133}^{(0)} &= \frac{1}{2(1-\nu^{(0)})} \left\{ -1 + 2\nu^{(0)} - \frac{1}{\alpha^2 - 1} + \left[1 - 2\nu^{(0)} + \frac{3}{2(\alpha^2 - 1)} \right] g \right\},\end{aligned}\tag{61}$$

$$S_{1212}^{(0)} = S_{1313}^{(0)} = \frac{1}{4(1 - \nu^{(0)})} \left\{ 1 - 2\nu^{(0)} - \frac{\alpha^2 + 1}{\alpha^2 - 1} - \frac{1}{2} \left[1 - 2\nu^{(0)} - \frac{3(\alpha^2 + 1)}{\alpha^2 - 1} \right] g \right\},$$

$$S_{2323}^{(0)} = \frac{S_{2222}^{(0)} - S_{2233}^{(0)}}{2},$$

where $\nu^{(0)}$ is the Poisson's ratio of the matrix material, $\alpha = b/a$ is the aspect ratio of the spheroidal inclusion in Figure 1(c), and g is given by

$$g = \frac{\alpha}{(\alpha^2 - 1)^{3/2}} \left[\alpha(\alpha^2 - 1)^{1/2} - \cosh^{-1} \alpha \right], \quad \alpha > 1, \quad \text{prolate};$$

$$\frac{\alpha}{(1 - \alpha^2)^{3/2}} \left[\cos^{-1} \alpha - \alpha(1 - \alpha^2)^{1/2} \right], \quad \alpha < 1, \quad \text{oblate.} \quad (62)$$

Finally, the function $f(\alpha)$ in equation (53) is given by

$$f(\alpha) = \frac{2\alpha^2 + 1}{\alpha^2 - 1} \left[\frac{3}{2} g(\alpha) - 1 \right], \quad (63)$$

where g is the function of α given by (62).

APPENDIX C: COMPARISON WITH AVAILABLE DILUTE RESULTS

For a composite comprised of an incompressible, pure power-law material reinforced by aligned, rigid spheroidal inclusions, we have seen that any external loading state may be represented in terms of three independent loading modes (Section 4). For any one of these loading modes ($\bar{\tau} = \bar{\tau}_d, \bar{\tau}_p$ or $\bar{\tau}_n$), the effective nonlinear energy function may be expressed in the form (see, for example, (34) for the axisymmetric mode, $\bar{\tau} = \bar{\tau}_d$)

$$\tilde{U}(\bar{\sigma}) = F(c^{(2)}; n, \alpha) \frac{\sigma_0}{n+1} \left(\frac{\bar{\tau}}{\tau_0} \right)^{n+1}, \quad (64)$$

where the function F reflects the reinforcing effect of the rigid inclusions.

For dilute concentrations of inclusions, a formal expansion in the concentration $c^{(2)}$ leads to the relation

$$F \approx 1 - c^{(2)} g(n, \alpha), \quad (65)$$

where g is a function of the nonlinearity n and inclusion aspect ratio α . Such an expansion is valid when the product of $c^{(2)}$ and g is much smaller than unity. Since it can be shown that the function g becomes unbounded in the limits as α approaches zero or infinity, it is useful to note that the range of validity (in $c^{(2)}$) of the dilute expansion (65) approaches zero as α approaches zero or infinity. This is in agreement with our previous comment that the limit as $c^{(2)} \rightarrow 0$ and $\alpha \rightarrow 0, \infty$ is singular (see Section 3.1.2). One must also be careful with the limit as $n \rightarrow \infty$ in the above expansion.

Next we compare our analytical estimates for F , in the case of axisymmetric loading, with corresponding dilute numerical results of He (1990) and Lee & Mear (1991a). These numerical results are based on the computation of the effective energy function $\tilde{U}$ of a composite made of one inclusion embedded in an infinite power-law matrix material.

Figure 9 shows a comparison, for a strongly nonlinear composite ($n = 10$), between our analytical estimates of F (they appear as curved lines) and the dilute numerical estimates of F , based on expansion (65), using the results of Lee & Mear (1991a) for the function g (they appear as straight lines). We observe that our results initially (for small $c^{(2)}$) lie slightly below the dilute estimates; however, the two curves intersect at a certain critical value of $c^{(2)}$, after which the two curves quickly diverge. The following observations are in order.

The intersection point, which serves as a measure of the range of validity of the dilute approximation, occurs for smaller and smaller values of $c^{(2)}$ as the aspect ratio of the inclusion increases, or decreases, from unity. This is in agreement with the discussion following equation (65).

Although the initial slopes of the dilute and nondilute curves are different, and this difference becomes larger as the aspect ratio of the inclusions increases, or decreases, from unity, we observe that the maximum difference between the actual values of the two curves (before they intersect) is not very large. Since the limit as $c^{(2)} \rightarrow 0$ and $\alpha \rightarrow 0, \infty$ is singular, it makes more sense to compare this difference in the actual values of the estimates for the function F (a direct measure of the reinforcing effect of the inclusions), and not the differences in the slopes. This is because, although the difference in the slopes may be large, the regions over which the dilute approximation holds are small. This leads to relatively small differences in the actual values of the functions. In Figure 10, we depict plots of this maximum relative difference, denoted by Δ_{rel} , as functions of the aspect ratio of the inclusions α , for the dilute results of both He (1990) and Lee & Mear (1991a). Thus we find that for values of α between 0.1 and 10, the maximum relative difference varies roughly

between 10 and 20 percent. For values of α outside this range, this type of comparison is not very useful because, at finite inclusion volume fractions $c^{(2)}$, the inclusions become so long (or wide) that the limits of continuous reinforcement are approached (fiber-reinforced and laminated composites). Thus, for these extreme limits, expressions of the type of (22) and (27), for fixed density (zero volume fraction) of thin needles and disks, respectively, are more appropriate.

An alternative comparison that may be made is with the dilute results of Duva & Storm (1989) for composites reinforced by rigid fibers. In this case, the axisymmetric mode is rigid, and these authors considered the transverse shear mode $\bar{\tau}_p$ (plane strain deformation perpendicular to the fibers). Thus, they were able to compute the function $g(n, \alpha)$ appearing in (65) for the transverse shear mode of fiber-reinforced composites. The corresponding relative difference Δ_{rel} between their numerical results and our analytical results (19) for fibers is shown in Table 1, as a function of the nonlinearity parameter n . We find that the maximum difference, occurring for $n = 10$, is less than five percent, which corresponds to excellent agreement between the two sets of results.

In summary, we find relatively good agreement between our analytical results and available numerical results for dilute concentrations of inclusions, provided that we stay away from singular limits (such as the limit of long fibers for the axisymmetric mode), in which case the differences can become large (although not very relevant, physically). The strength of our results, however, lies in their generality and relative simplicity.

Table 1. Maximum relative difference between our analytical results and the dilute numerical results of Duva & Storm (1989) for the transverse mode in fiber-reinforced composites.

n	1	2	3	4	5	6	7	8	9	10
Δ_{rel}	0	.0053	.0154	.0198	.0268	.0315	.0357	.0377	.0414	.0458

List of captions

Figure 1. Composite materials reinforced by spheroidal inclusions. (a) A composite with aligned spheroidal inclusions; (b) a composite with randomly oriented spheroidal inclusions; (c) a description of prolate and oblate spheroids.

Figure 2. The effect of inclusion shape on the effective linear and nonlinear stress normalization factors for aligned-inclusion composites subjected to axisymmetric loading. Note that, since the reinforcing phase is rigid, the reciprocal of strength $1/(\tilde{\sigma}_0)_d$ is plotted for convenience. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 3. The effect of inclusion shape on the effective linear and nonlinear stress normalization factors for aligned-inclusion composites subjected to transverse shear loading. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 4. The effect of inclusion shape on the effective linear and nonlinear stress normalization factors for aligned-inclusion composites subjected to longitudinal shear loading. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 5. A comparison of the effective flow stresses of the three independent loading modes for perfectly plastic composites reinforced by aligned spheroidal inclusions: (a) effective flow stresses versus aspect ratio (for $c^{(2)} = 0.2$); (b) effective flow stresses versus inclusion

concentration $c^{(2)}$ (for $\alpha = 0.1, 1$ and 10).

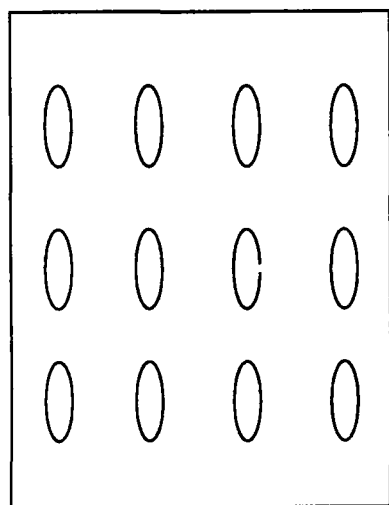
Figure 6. Plot of the axisymmetric strain $\bar{\gamma}_d$ versus the transverse shear stress $\bar{\tau}_p$ for an aligned-inclusion composite with three different axisymmetric preloadings, namely, $\bar{\tau}_d/\tau_0 = 0.1, 2.8$ and 3.5 .

Figure 7. Plot of the axisymmetric strain $\bar{\gamma}_d$ versus the axisymmetric stress $\bar{\tau}_d$ an aligned-inclusion composite with three different transverse shear preloadings, namely, $\bar{\tau}_p/\tau_0 = 0, 1.0$ and 1.5 .

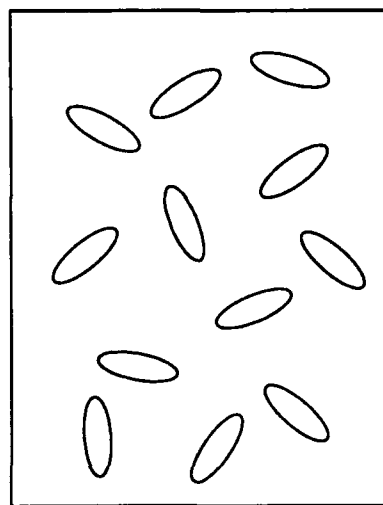
Figure 8. Plots of the reciprocal of the effective stress normalization factors for composites reinforced by randomly oriented spheroidal inclusions versus inclusion concentration $c^{(2)}$. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 9. A comparison of our analytical prediction for the function F with the dilute calculations of Lee & Mear (1991a). The later results appear as straight lines; the former as curved lines.

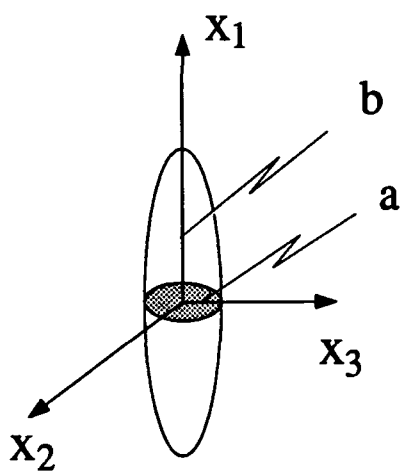
Figure 10. Plots of the largest relative difference Δ_{rel} between our results and the dilute results of He (1990) and Lee & Mear (1991a), respectively, versus aspect ratio α .



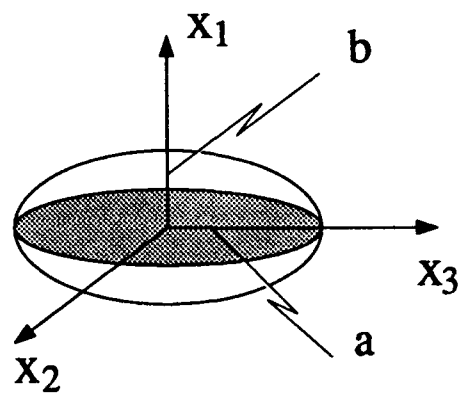
(a)



(b)



Prolate



Oblate

(c)

Fig. 1

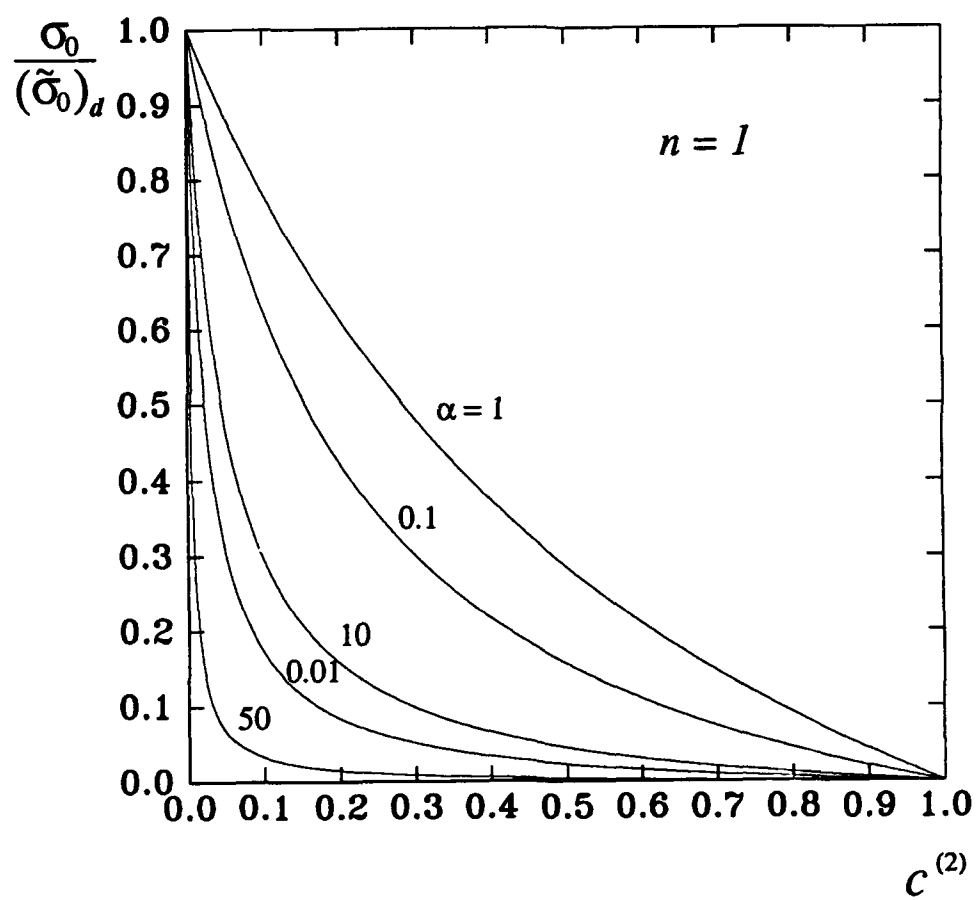


Fig. 2(a)

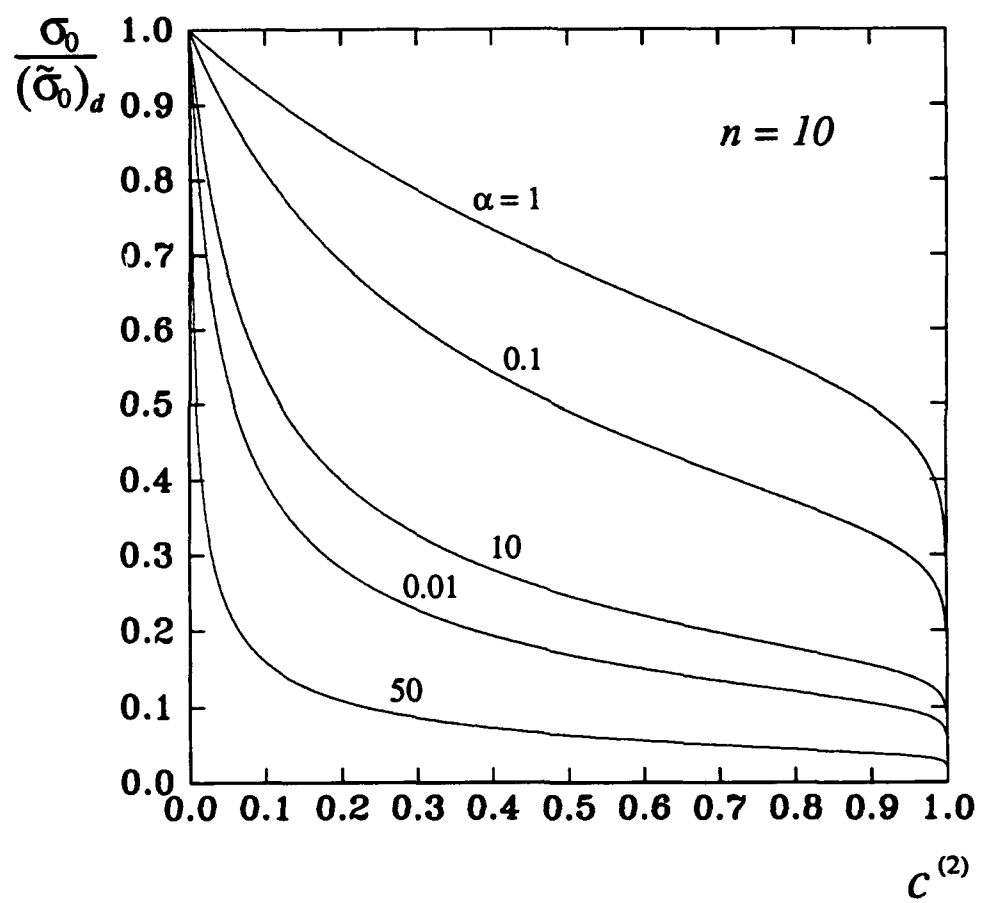


Fig. 2(b)

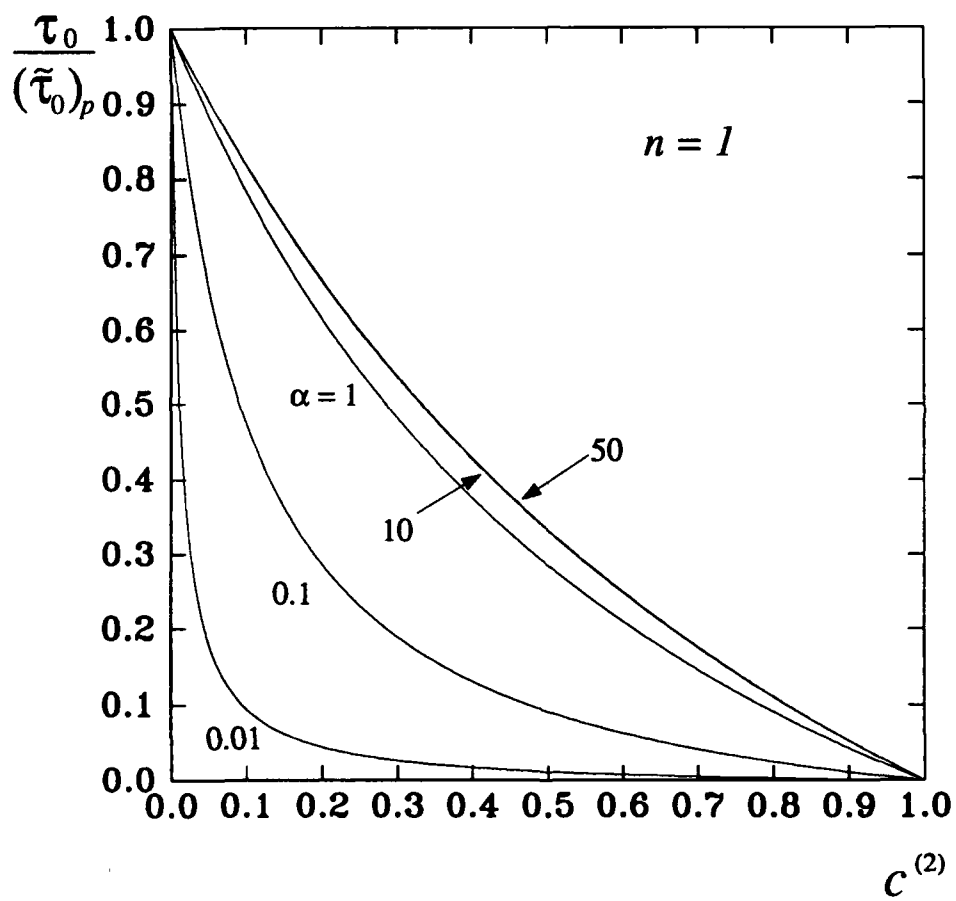


Fig. 3(a)

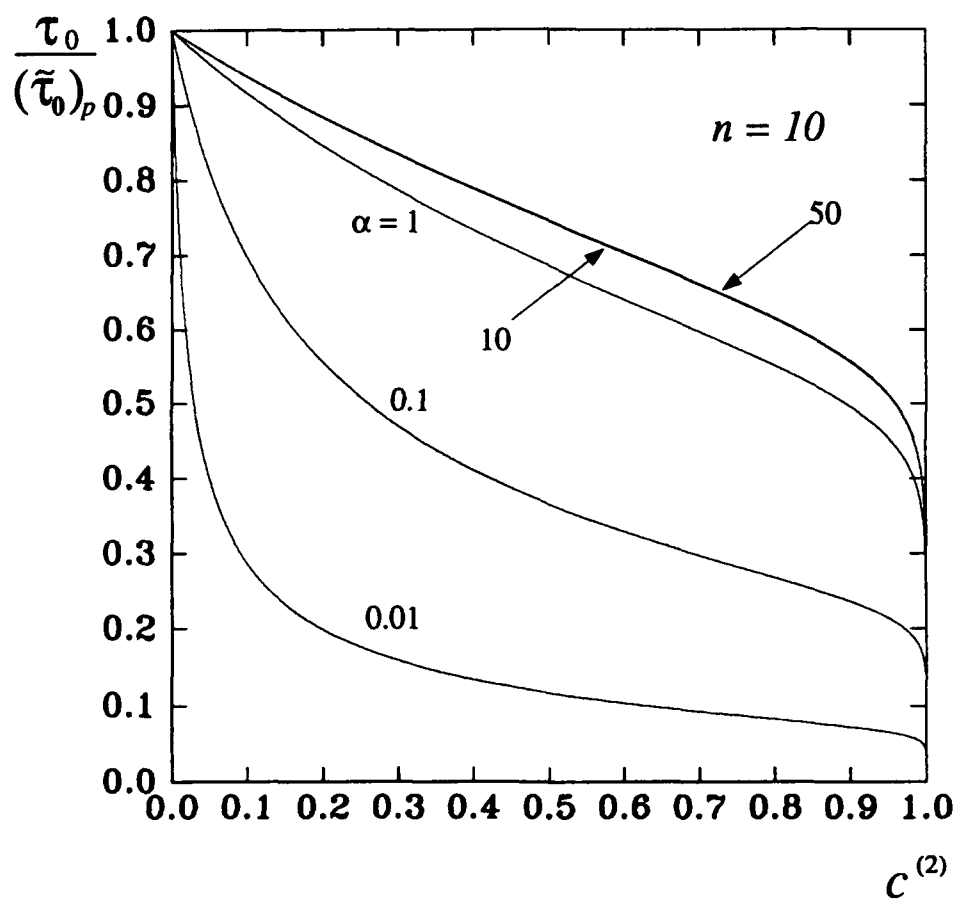


Fig. 3(b)

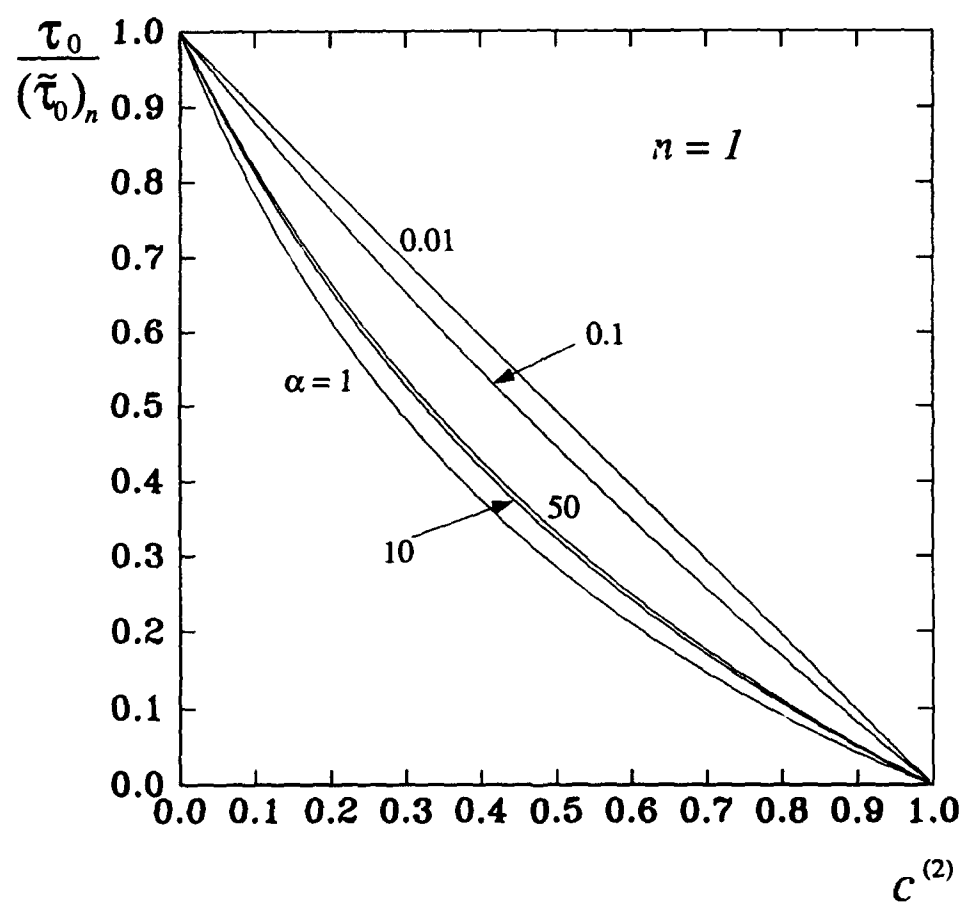


Fig. 4(a)

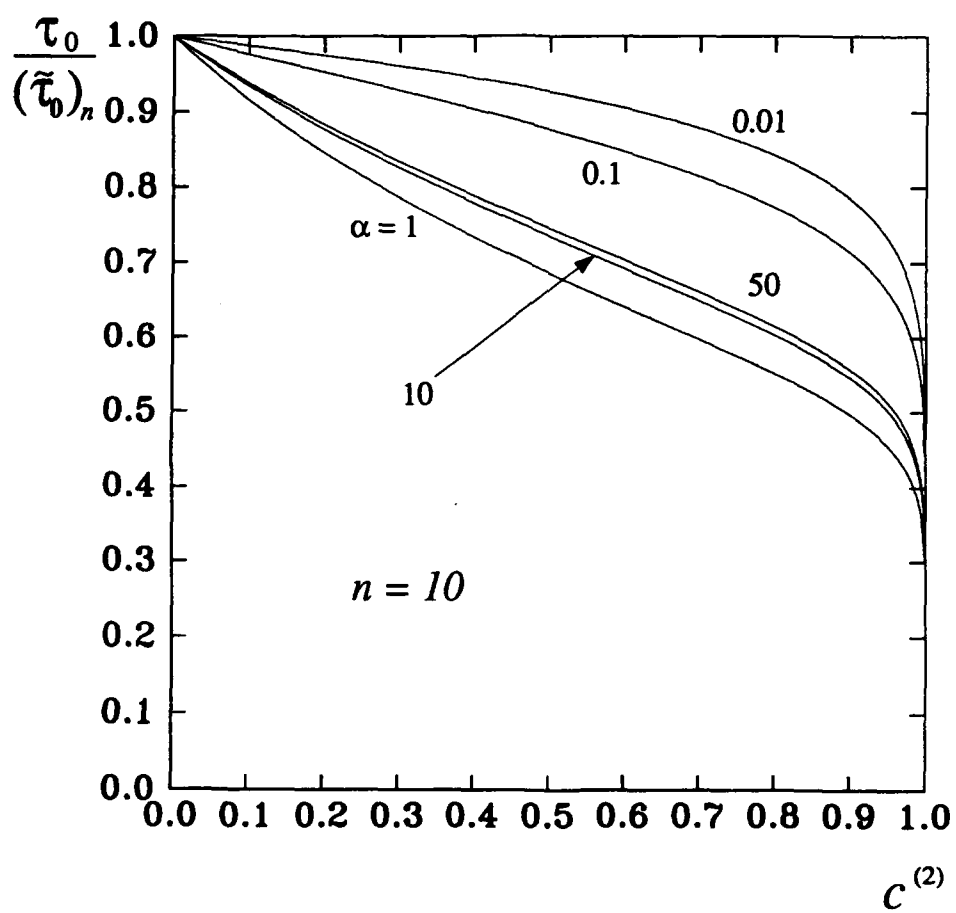


Fig. 4(b)

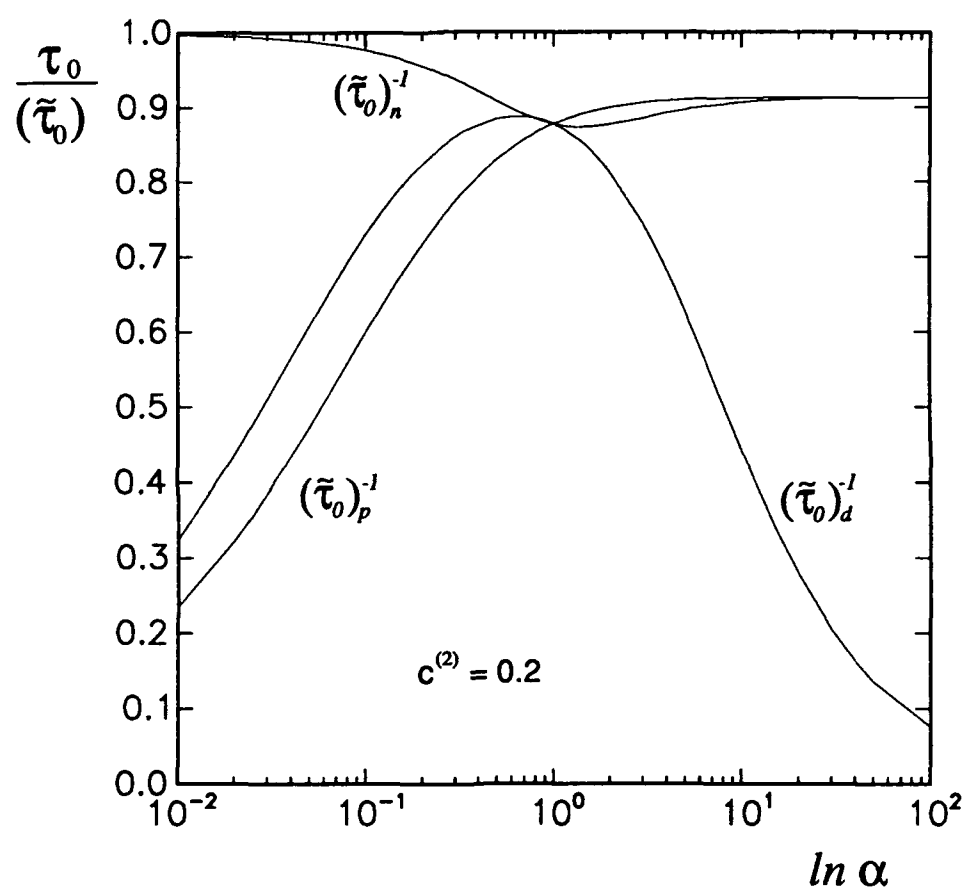


Fig. 5(a)

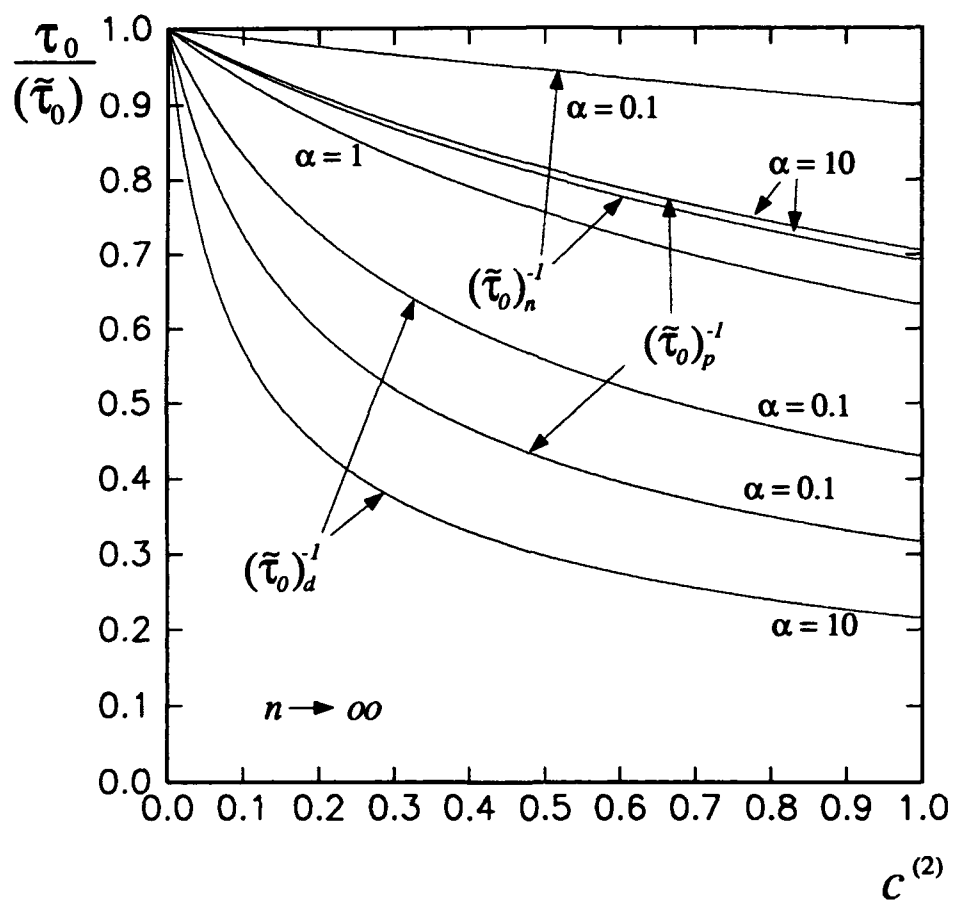


Fig. 5(b)

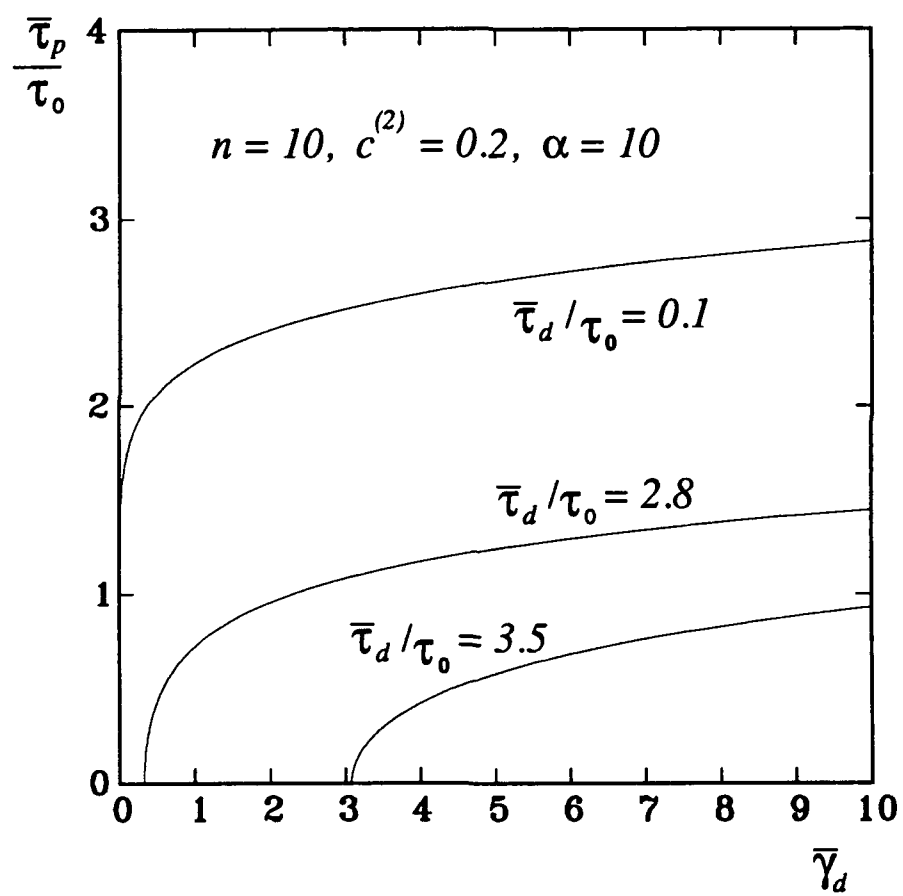


Fig. 6

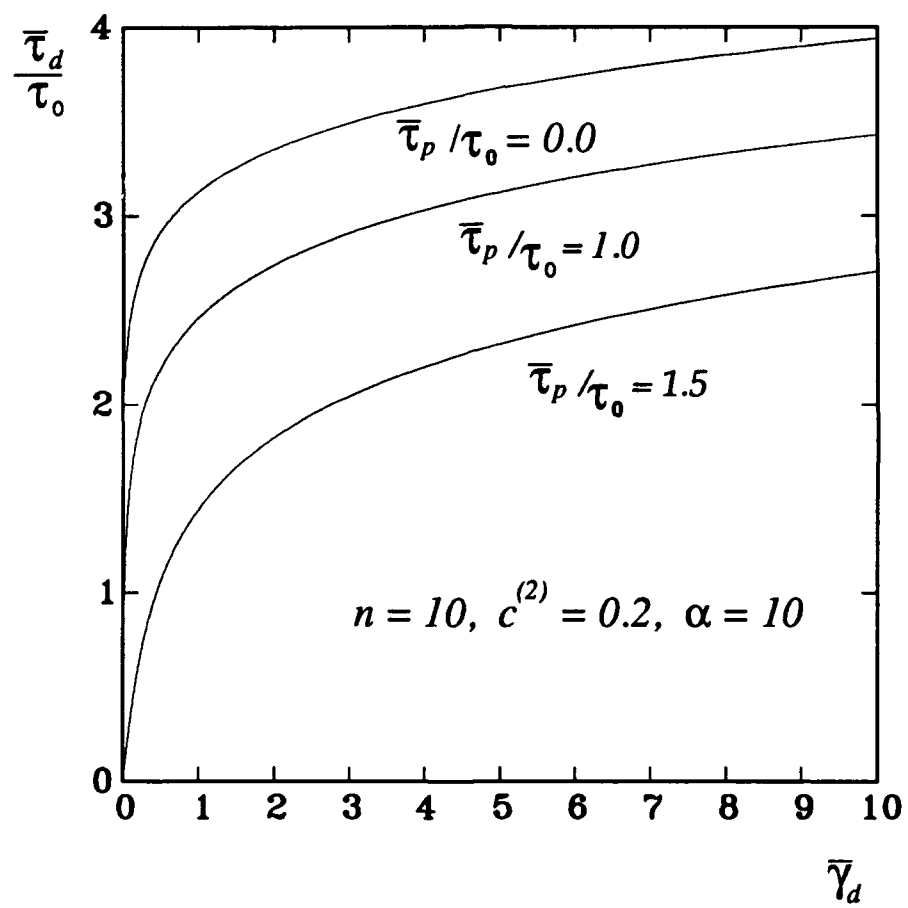


Fig. 7

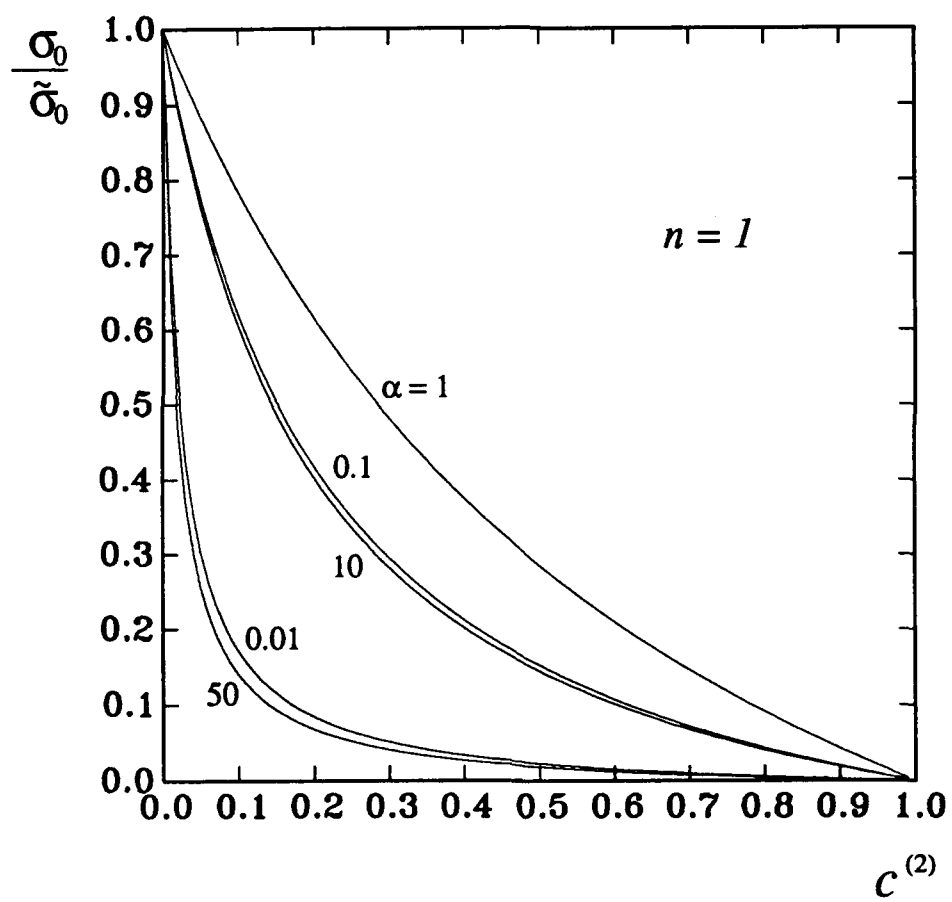


Fig. 8(a)

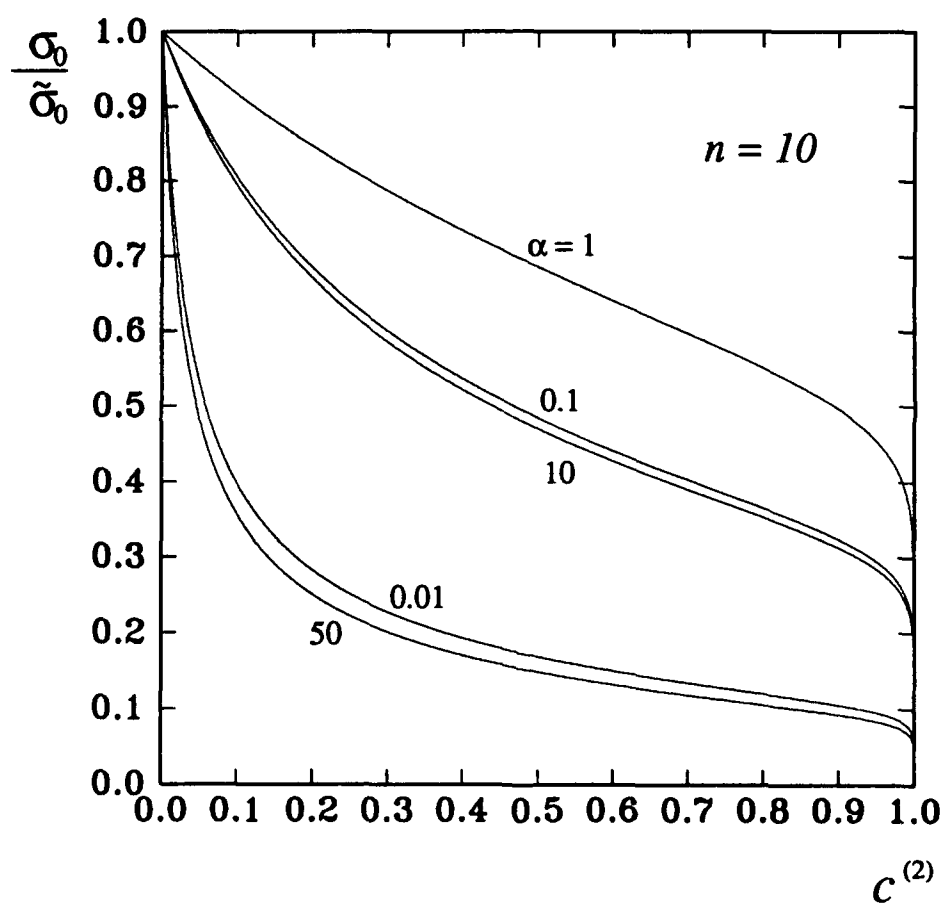


Fig. 8(b)

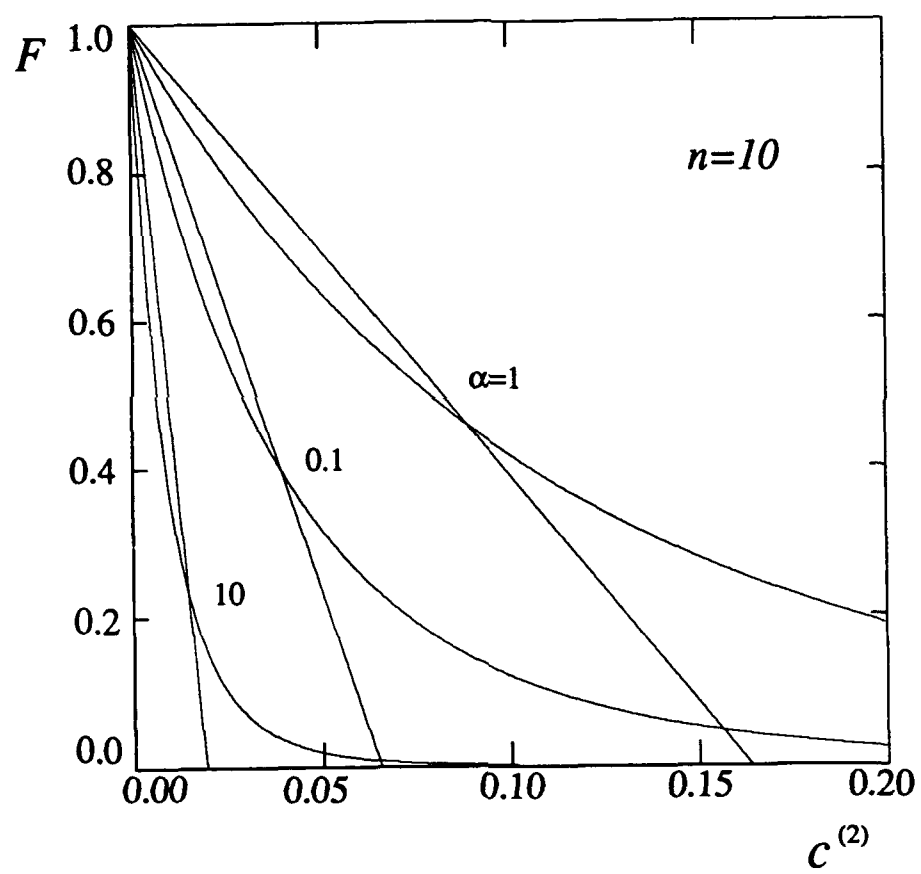


Fig. 9

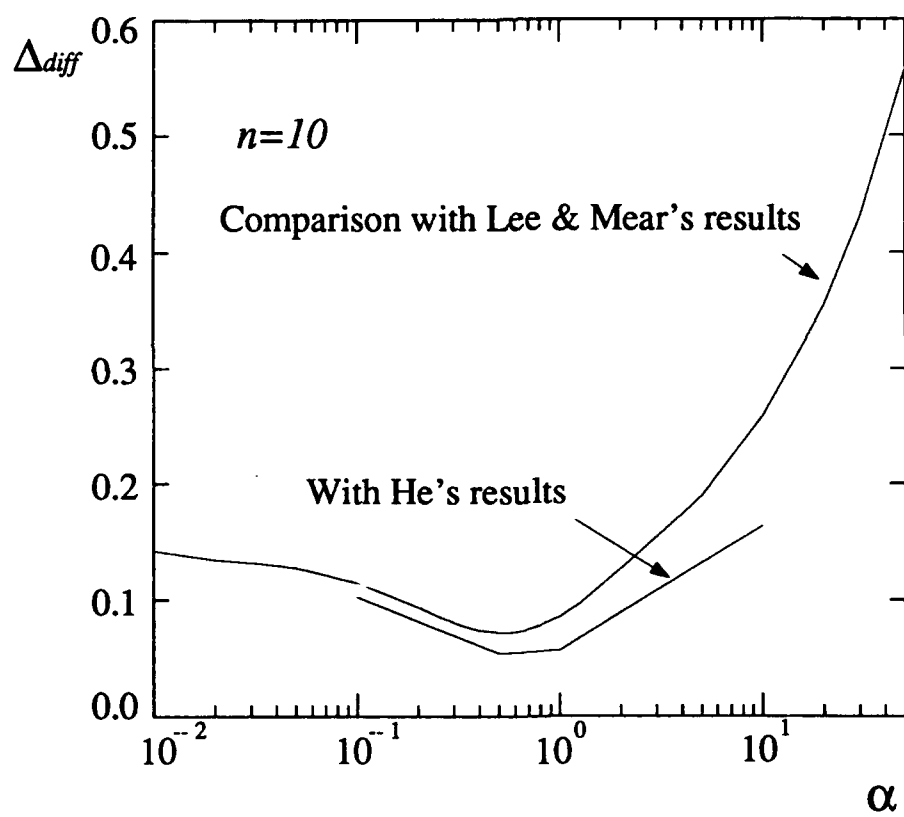


Fig. 10

Reference [16]

CONSTITUTIVE MODELS FOR DUCTILE SOLIDS REINFORCED BY RIGID SPHEROIDAL INCLUSIONS

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Abstract— We study the effective constitutive response of composite materials made of rigid spheroidal inclusions dispersed in a ductile matrix phase. Given a general convex potential characterizing the plastic (in the context of J_2 -deformation theory) behavior of the matrix material, we derive expressions for the corresponding effective potentials of the rigidly reinforced composites, under *general* loading conditions. The derivation of the effective potentials for the nonlinear composites is based on a variational procedure developed recently by Ponte Castañeda (1991a). We consider two classes of composites. In the first class, the spheroidal inclusions are aligned, resulting in overall transversely isotropic symmetry for the composite. In the second class, the inclusions are randomly oriented, and thus the composite is macroscopically isotropic. The effective response of composites with aligned inclusions depends on both the orientation of the loading and on the inclusion concentration and shape. Comparing the strengthening effects of rigid oblate and prolate spheroids, we find that pro-

late spheroids give rise to stiffer effective response under axisymmetric (relative to the axis of transverse isotropy) deformations, while oblate spheroids provide greater reinforcement for materials loaded in transverse shear (relative to the axial direction). On the other hand, nearly spherical (slightly prolate) spheroids are most effective in strengthening the composite under longitudinal shear deformations. Thus, the optimal shape for strengthening composites with aligned inclusions depends strongly on the deformation mode. Alternatively, the properties of composites with randomly oriented spheroidal inclusions, being isotropic, depend only on the concentration and shape of the inclusions. We find that both oblate and prolate inclusions lead to significant strengthening for this class of composites.

1. INTRODUCTION

In this work, we seek to estimate the effective, or overall, constitutive response of composite materials comprised of an incompressible, ductile matrix phase reinforced by rigid, spheroidal inclusions. The matrix and inclusions are assumed to be perfectly bonded, and the inclusions are further assumed to be small compared to the overall dimension of the specimen under consideration. Here, we are interested in predicting the effects of the volume fraction, aspect ratio and orientation of the inclusions on the effective properties of the composite. A potential application of this study would be in the optimal design of materials in which a nonlinear ductile phase is reinforced by relatively stiff inclusions. These materials include some metal-matrix composites and intermetallics reinforced by stiff particles.

Composites with linear constitutive behavior for both the matrix and inclusion phases have been studied extensively. In this connection, Eshelby's (1957) celebrated solution for the constant stress and strain fields within an ellipsoidal inclusion in an infinite matrix, subjected

to uniform remote tractions, was instrumental in many developments, including the determination of the effective moduli of elastic composites with dilute concentrations of ellipsoidal inclusions. This fundamental result can also be used to compute the effective properties of multi-phase elastic composites with nondilute concentrations of the phases. Thus, for this purpose, Budiansky (1965) and Hill (1965) proposed the self-consistent method and obtained explicit expressions for the effective moduli of composites with isotropic distributions of spherical inclusions. Wu (1966) extended the use of the self-consistent method to predict the effective behavior of isotropic composites with randomly oriented ellipsoidal inclusions. Another remarkable achievement in the field of linear composite materials was the developments of new variational principles by Hashin and Shtrikman (1962). These variational principles allowed the derivation, by Hashin and Shtrikman (1963), of upper and lower bounds for the effective moduli of isotropic composite materials with prescribed volume fractions of the phases, but otherwise arbitrary microstructure. Extensions of this class of bounds for fiber-reinforced composites with overall transversely isotropic symmetry were first given by Hill (1964) and Hashin (1965). Further generalization of the Hashin-Shtrikman bounds and self-consistent estimates for anisotropic linear elastic composites have been given by Walpole (1966a,b, 1969) and Willis (1977). An alternative approach in the study of composite materials is to assume periodic microstructures and to compute the effective properties of the composite from a boundary-value problem defined on the unit cell with periodic boundary conditions. This approach has been pursued by Nemat-Nasser and Taya (1981) and Nemat-Nasser *et al.* (1982). For a more complete review of the field of linear composites, the reader is referred to the articles by Hashin (1983) and Willis (1982) and to the monograph by Sanchez-Palencia (1980).

Nonlinear composite materials have recently been the subject of increased attention. Talbot and Willis (1987) and Ponte Castañeda and Willis (1988) (see also Willis 1991) developed bounds and self-consistent estimates for the effective behavior of nonlinear composites via an extension of the Hashin-Shtrikman variational principles to nonlinear materials, proposed by Talbot and Willis (1985). More recently, Ponte Castañeda (1991a) proposed a new, conceptually simple, variational procedure which leads to more general bounds and estimates for the properties of nonlinear composites. The procedure makes use of arbitrary bounds and estimates for classes of *linear* comparison composites to generate bounds and estimates for the corresponding classes of nonlinear composites. This variational procedure has been used by Ponte Castañeda (1991a,b, 1992) and deBotton and Ponte Castañeda (1992) to obtain estimates for the properties of isotropic particulate composites and anisotropic laminated and fiber-reinforced composites. Other contributions in the field of nonlinear composites include the work of Qiu and Weng (1991) dealing with the shape effect on the overall properties of two-phase elastic-plastic composite materials. They make use of an appropriate adaptation of the method of Mori and Tanaka (1973). In addition, a number of works have appeared recently (see, for example, Christman et al. 1989 and Bao et al. 1991) dealing with the effective properties of periodic composites; these being computed via finite-element analyses of the pertinent unit-cell, boundary-value problems. An alternative approach advocated by Duva (1984) (see also Duva and Hutchinson 1984), and used more recently by He (1990) and Lee & Mear (1991a,b), is to make use of the solution of a kernel problem, involving an inclusion in an infinite matrix of the nonlinear matrix material, to generate dilute estimates for the nonlinear composites. Generalizations to nondilute concentrations may be accomplished by the use of the differential self-consistent model (McLaughlin, 1977).

This paper deals with the effective constitutive behavior of nonlinear, incompressible composites that are reinforced by either aligned, or randomly oriented, rigid spheroidal inclusions. We concentrate on the effect of the inclusion shape on the effective behavior of the nonlinear composites. The method of analysis is based on Ponte Castañeda's (1991a) variational principle. For the case of aligned spheroidal inclusions with overall transversely isotropic symmetry, the method gives *explicit* expressions for the effective constitutive behavior of the nonlinear composites under *general* loading conditions (i.e., arbitrary combinations of axisymmetric tension, transverse shear and longitudinal shear). For the case of randomly oriented inclusions with overall isotropic symmetry, the method also gives *explicit* expressions for the effective behavior of the composites, but in this case, the composites being incompressible and isotropic, there is essentially only one loading mode.

2. VARIATIONAL CHARACTERIZATION OF EFFECTIVE PROPERTIES

Consider a general specimen of a nonlinear heterogeneous material occupying a domain Ω (of unit volume) with boundary $\partial\Omega$. The material is characterized by a stress potential $U(\mathbf{x}, \boldsymbol{\sigma})$, depending on position $\mathbf{x}$ and the stress field $\boldsymbol{\sigma}(\mathbf{x})$, and is such that the strain field $\boldsymbol{\epsilon}(\mathbf{x})$ is given by

$$\boldsymbol{\epsilon}(\mathbf{x}) = \frac{\partial U(\mathbf{x}, \boldsymbol{\sigma})}{\partial \boldsymbol{\sigma}}. \quad (1)$$

Then, the effective constitutive relation for the heterogeneous material may be defined, following Hill (1963), in terms of

$$\bar{\boldsymbol{\epsilon}} = \frac{\partial \tilde{U}(\bar{\boldsymbol{\sigma}})}{\partial \bar{\boldsymbol{\sigma}}}, \quad (2)$$

where $\bar{\boldsymbol{\epsilon}}$ and $\bar{\boldsymbol{\sigma}}$ are the mean values of the strain and stress fields over Ω , and where $\tilde{U}(\bar{\boldsymbol{\sigma}})$ denotes the effective stress potential of the composite, depending on the uniform stress

boundary condition

$$\mathbf{t}(\mathbf{x}) = \bar{\boldsymbol{\sigma}} \cdot \mathbf{n}, \quad \mathbf{x} \in \partial\Omega. \quad (3)$$

In this last relation, $\mathbf{t}$ is the traction vector and $\mathbf{n}$ is the outward normal to $\partial\Omega$.

The effective potential function $\tilde{U}(\bar{\boldsymbol{\sigma}})$ may be obtained directly from the principle of minimum complementary energy as

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma} \in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U(\mathbf{x}, \boldsymbol{\sigma}) dv, \quad (4)$$

where

$$S(\bar{\boldsymbol{\sigma}}) = \{\boldsymbol{\sigma} | \nabla \cdot \boldsymbol{\sigma} = 0 \text{ in } \Omega, \text{ and } \mathbf{t}(\mathbf{x}) = \bar{\boldsymbol{\sigma}} \cdot \mathbf{n}, \mathbf{x} \in \partial\Omega\} \quad (5)$$

represents the set of statically admissible stress fields satisfying the condition (3).

While the effective behavior of the composite is fully described by $\tilde{U}$ in terms of relation (2), the determination of $\tilde{U}$ from (4) and (5) presents real difficulties in that it requires the solution of a nonlinear boundary value problem with complex structure. Ponte Castañeda (1991a,b) introduced a variational principle that can be used to estimate the effective potential functions of nonlinear composites in terms of optimization problems involving the effective potential functions of appropriate classes of *linear* comparison composites. Thus, results for the effective properties of linear composites may be used to generate corresponding estimates for the effective properties of nonlinear composites. In this paper, we make use of the Hashin-Shtrikman bounds of Walpole (1969) and Willis (1977) for linear composites with spheroidal inclusions to estimate the effective constitutive behavior of ductile-matrix materials reinforced by rigid, spheroidal inclusions. Ponte Castañeda's variational principle is briefly reviewed in the following.

We consider only composites with incompressible, isotropic matrices. Thus, the potential

function for the nonlinear matrix may be written in the form

$$U(\mathbf{x}, \boldsymbol{\sigma}) = \phi(\mathbf{x}, \tau_e), \quad (6)$$

where

$$\tau_e = \sqrt{\frac{1}{2} \boldsymbol{\sigma}' \cdot \boldsymbol{\sigma}'} \quad (7)$$

is the effective shear stress and $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - \frac{\text{tr} \boldsymbol{\sigma}}{3} \mathbf{I}$ is the deviatoric stress tensor.

Then, under certain technical hypotheses (Ponte Castañeda, 1992), satisfied in this paper, we may express the matrix potential function in terms of the following optimization problem, namely,

$$U(\mathbf{x}, \boldsymbol{\sigma}) = \max_{\mu_0 \geq 0} \{U_0(\mathbf{x}, \boldsymbol{\sigma}) - V(\mathbf{x}, \mu_0)\}, \quad (8)$$

where U_0 is the potential function of an incompressible, linear-elastic comparison material with shear modulus $\mu_0(\mathbf{x})$, such that $U_0(\mathbf{x}, \boldsymbol{\sigma}) = \frac{1}{2\mu_0(\mathbf{x})} \tau_e^2$, and where

$$V(\mathbf{x}, \mu_0) = \max_{\boldsymbol{\sigma}} \{U_0(\mathbf{x}, \boldsymbol{\sigma}) - U(\mathbf{x}, \boldsymbol{\sigma})\}. \quad (9)$$

Then, the effective potential function of a given nonlinear composite is obtained by averaging relation (8); the result is (Ponte Castañeda, 1992)

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \max_{\mu_0(\mathbf{x}) \geq 0} \left\{ \tilde{U}_0(\bar{\boldsymbol{\sigma}}) - \int_{\Omega} V(\mathbf{x}, \mu_0(\mathbf{x})) dv \right\}, \quad (10)$$

where $\tilde{U}_0(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma} \in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U_0(\mathbf{x}, \boldsymbol{\sigma}) dv$ is the effective potential of the linear comparison composite.

Even though (10) is an exact expression for the effective potential function of a given nonlinear composite, it involves an infinite-dimensional optimization problem over the set of non-negative functions $\mu_0(\mathbf{x})$. Thus, in general, the variational principle (10) is at least

as difficult to implement as the classical variational principle (4), except for some very special microstructures (e.g., laminated composites; see deBotton & Ponte Castañeda 1992). However, it may be shown that (10) can be utilized in an approximate fashion to generate bounds for the effective properties of classes of nonlinear composites with more general microstructures. This is discussed next.

We begin by specializing the general class of composites considered above to the class of two-phase composites. Each phase is assumed to be incompressible, isotropic and homogeneous, characterized by a convex potential function $U^{(r)}(\boldsymbol{\sigma}) = \phi^{(r)}(\tau_e)$ ($r = 1$ or 2 , for phase 1 or 2). Further, the volume fractions of each phase, denoted by $c^{(r)}$ ($c^{(1)} + c^{(2)} = 1$ for a two-phase composite), are also assumed to be known. Even then, the effective potential function given by (10) requires the solution of an infinite dimensional optimization problem over the set of functions $\mu_0(\mathbf{x})$. However, by restricting our attention to the class of piecewise constant $\mu_0(\mathbf{x})$ (constant in each phase of the nonlinear material), we arrive at the following lower bound approximation for the effective potential function of the nonlinear composite, namely,

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) \geq \max_{\mu_0^{(1)}, \mu_0^{(2)} \geq 0} \left\{ \tilde{U}_0(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^2 c^{(r)} V^{(r)}(\mu_0^{(r)}) \right\}, \quad (11)$$

where $\mu_0^{(1)}$ and $\mu_0^{(2)}$ are the values of $\mu_0(\mathbf{x})$ in phases 1 and 2, respectively. Also, $\tilde{U}_0(\bar{\boldsymbol{\sigma}})$ is now the effective potential function of a linear comparison composite with *precisely* the same microstructure as the nonlinear composite, and

$$V^{(r)}(\mu_0^{(r)}) = \max_{\tau_e} \left\{ \frac{1}{2\mu_0^{(r)}} \tau_e^2 - \phi^{(r)}(\tau_e) \right\}. \quad (12)$$

We note that the inequality in (11) arises because the maximum is now evaluated over a strictly smaller set of functions (i.e., the set of piecewise constant functions).

The above inequality is useful because the optimization problem involved is two-dimensional, and therefore simpler to evaluate, assuming that we have estimates for the effective potential of two-phase, linear composites. In fact, if we have a lower bound for the effective potential of the linear composites $\tilde{U}_0(\bar{\sigma})$, then (11) generates a lower bound for the effective potential of the nonlinear composite. If, on the other hand, we have an upper bound for $\tilde{U}_0(\bar{\sigma})$, (11) generates only an estimate for the upper bound, which is generally lower than the true upper bound.

3. NONLINEAR ESTIMATES FOR COMPOSITES WITH RIGID INCLUSIONS

In this paper, we study composites made up of an incompressible, isotropic matrix material reinforced by either aligned, or randomly oriented, rigid spheroidal inclusions, as depicted in Figure 1. The shape of a typical inclusion is characterized by its aspect ratio $\alpha = b/a$. Thus, oblate spheroids have $\alpha < 1$, prolate spheroids have $\alpha > 1$ and spheres have $\alpha = 1$. The stress potential for the rigid inclusions (phase 2) vanishes identically (i.e., $\phi^{(2)}(\tau_e) = 0$). Then, to be able to make use of (11), we need to obtain estimates for the effective potentials $\tilde{U}_0$ of linear composites with rigid, spheroidal inclusions. Such results may be obtained directly from the work of Walpole (1966a,b, 1969) and Willis (1977) (see also Willis 1982, and Qiu and Weng 1990). Although these authors are concerned with both general bounds of the Hashin-Shtrikman type and self-consistent estimates, here we will make use only of the appropriate (non-trivial) lower bounds for the effective shear modulus. It is known (see, for example, Milton and Kohn 1988) that these lower bounds correspond to the least stiff isotropic composites which may be made with fixed volume fractions of the phases. Further, these extremal composites have particulate microstructures, where the inclusions are made

of the rigid phase. Thus, we may interpret the result of implementing these non-trivial Hashin-Shtrikman bounds into (11) as (upper) estimates for the effective potential functions of the corresponding nonlinear composites. We begin by considering the case of composites with aligned inclusions, and consider the case of randomly oriented inclusions later.

3.1 Aligned spheroidal inclusions

In this section, we consider composite materials with nonlinear, incompressible matrices and aligned rigid spheroidal inclusions. We note that this case was also briefly considered by Talbot and Willis (1992), although explicit results of the type obtained here were only determined for the special case of aligned rigid disks. While each phase of the composite is isotropic, the geometry of the inclusions is such that the composite exhibits overall transversely isotropic symmetry. The direction of inclusion alignment provides the axis of transverse isotropy (see Figure 1(a), where $\mathbf{x}_1$ defines the axis of symmetry).

In this case, an upper bound for the effective stress potential of the linear comparison material $\tilde{U}_0$ may be obtained from the previously mentioned works (see Appendix A). The result may be written

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\mu^{(1)}} \left(\bar{\tau}^{(1)} \right)^2, \quad (13)$$

where

$$\bar{\tau}^{(1)} = \sqrt{\frac{\mu^{(1)}}{\tilde{\mu}_d} \bar{\tau}_d^2 + \frac{\mu^{(1)}}{\tilde{\mu}_p} \bar{\tau}_p^2 + \frac{\mu^{(1)}}{\tilde{\mu}_n} \bar{\tau}_n^2}, \quad (14)$$

Here $\mu^{(1)}$ is the shear modulus of the incompressible, isotropic matrix material, $\frac{\mu^{(1)}}{\tilde{\mu}_d}$, $\frac{\mu^{(1)}}{\tilde{\mu}_p}$ and $\frac{\mu^{(1)}}{\tilde{\mu}_n}$ are given by relations (53) in Appendix A, and $\bar{\tau}_d, \bar{\tau}_p$ and $\bar{\tau}_n$ (note that $\bar{\tau}_e = (\bar{\tau}_d^2 + \bar{\tau}_p^2 + \bar{\tau}_n^2)^{1/2}$) are the three transversely isotropic invariants of the applied stress tensor $\bar{\sigma}$, corresponding to the three independent deformation modes for an incompressible, transversely isotropic

material: axisymmetric deformation, transverse shear and longitudinal shear, respectively (see Appendix B).

Using the upper bound (13) for the effective potential of the linear comparison material $\tilde{U}_0$, we can compute, via relation (11), an upper estimate for the effective stress potential of the nonlinear composite $\tilde{U}$. The result is

$$\tilde{U}(\bar{\sigma}) \cong \max_{\mu^{(1)}} \left\{ \frac{1}{2\mu^{(1)}} (\bar{\tau}^{(1)})^2 - c^{(1)} \max_{\tau_e} \left[\frac{1}{2\mu^{(1)}} \tau_e^2 - \phi^{(1)}(\tau_e) \right] \right\}, \quad (15)$$

which can be shown (by interchanging the maxima operations) to reduce to

$$\tilde{U}(\bar{\sigma}) \cong c^{(1)} \phi^{(1)} \left(\frac{\bar{\tau}^{(1)}}{\sqrt{c^{(1)}}} \right). \quad (16)$$

This relation expresses the effective potential function of the nonlinear composite in terms of the stress potential of the nonlinear matrix material $\phi^{(1)}$, the matrix volume fraction $c^{(1)}$, and an estimate for the average stress in the matrix $\bar{\tau}^{(1)}/\sqrt{c^{(1)}}$ (which in turn depends on the appropriate invariants of $\bar{\sigma}$, on the matrix volume fraction and the aspect ratio of the inclusions α). We emphasize that the above result is meant to be interpreted as an *estimate* (not an exact result) for the (weakest) effective behavior of the class of composites under consideration.

Corresponding to the three invariants of the stress tensor $\bar{\sigma}$, we can identify three invariants of the strain tensor $\bar{\epsilon}$: the deviatoric shear strain $\bar{\gamma}_d$, the transverse shear strain $\bar{\gamma}_p$ and the longitudinal shear strain $\bar{\gamma}_n$ (such that $\bar{\epsilon}_e = 1/\sqrt{3}(\bar{\gamma}_d^2 + \bar{\gamma}_p^2 + \bar{\gamma}_n^2)^{1/2}$; see Appendix B). Then, the effective stress-strain relations for the nonlinear composite may be expressed in the form

$$\bar{\gamma}_d = c^{(1)} \frac{\mu^{(1)}}{\tilde{\mu}_d} \frac{\bar{\tau}_d}{\bar{\tau}^{(1)}} \frac{\partial \phi^{(1)}}{\partial \bar{\tau}^{(1)}} \quad ,$$

$$\begin{aligned}\bar{\gamma}_p &= c^{(1)} \frac{\mu^{(1)}}{\tilde{\mu}_p} \frac{\bar{\tau}_p}{\bar{\tau}^{(1)}} \frac{\partial \phi^{(1)}}{\partial \bar{\tau}^{(1)}} \quad , \\ \bar{\gamma}_n &= c^{(1)} \frac{\mu^{(1)}}{\tilde{\mu}_n} \frac{\bar{\tau}_n}{\bar{\tau}^{(1)}} \frac{\partial \phi^{(1)}}{\partial \bar{\tau}^{(1)}} \quad .\end{aligned}\tag{17}$$

It is seen from these stress-strain relations that, for the nonlinear composite, all three deformation modes are coupled with each other (for example, $\bar{\gamma}_d$ depends on $\bar{\tau}_d, \bar{\tau}_p$ and $\bar{\tau}_n$ through $\partial \phi^{(1)} / \partial \bar{\tau}^{(1)}$). This is in contrast with the corresponding linear composite (see (54) of Appendix A).

So far we have derived effective constitutive relations for a nonlinear matrix material reinforced by aligned rigid spheroidal inclusions in prescribed volume fraction and given aspect ratio of the inclusions. Also, unlike the results of other investigations dealing exclusively with axisymmetric loading conditions (see Bao et al. 1991, and Lee and Mear 1991a,b), these results apply for arbitrary loading conditions. In the next subsections, we specialize these general results to some special cases of theoretical and practical importance. For example, the limit $\alpha \rightarrow \infty$, with finite inclusion concentration, results in a composite with infinitely long fibers (continuous reinforcement). On the other hand, the opposite limit as $\alpha \rightarrow 0$, with finite inclusion concentration, corresponds to a laminated composite material (another case of continuous reinforcement). The result for $\alpha = 1$ corresponds to spherical inclusions (particulate reinforcements). The last two cases have been studied in some detail by Ponte Castañeda (1991a,b) and deBotton and Ponte Castañeda (1992).

3.1.1 Fiber-reinforced composites

In the limit as $\alpha \rightarrow \infty$, for a fixed, finite concentration of inclusions, an asymptotic expansion for $\bar{\tau}^{(1)}$ can be obtained explicitly from (14) and (53). The leading order term is

$$\bar{\tau}^{(1)} = \sqrt{\left(1 - \frac{2c^{(2)}}{1 + c^{(2)}}\right) (\bar{\tau}_p^2 + \bar{\tau}_n^2)}. \quad (18)$$

Correspondingly, the effective potential of the nonlinear composite reduces to

$$\tilde{U}(\bar{\sigma}) = c^{(1)} \phi^{(1)} \left(\sqrt{\frac{1}{c^{(1)}} \left(1 - \frac{2c^{(2)}}{1 + c^{(2)}}\right) (\bar{\tau}_p^2 + \bar{\tau}_n^2)} \right). \quad (19)$$

This equation shows that the response of the nonlinear composite to transverse and longitudinal shear is the same. On the other hand, the composite is effectively rigid under axisymmetric loading. This is physically reasonable since for a finite volume fraction of the inclusions, the inclusions must become infinitely long as $\alpha \rightarrow \infty$. Thus, the fiber-reinforced composite has just two effective deformation modes (instead of three). We note that as the inclusion concentration $c^{(2)} \rightarrow 0$ in (19), the potential function is still dependent only on the shear loading modes.

3.1.2 Needle-reinforced composites

For the fiber-reinforced composite discussed above, the continuity of the rigid fibers severely restricted the modes of deformation possible. We consider next rigid spheroidal inclusions with arbitrarily large aspect ratios but with finite length. These aligned thin needles, each of length $2b$ and radius $a = \epsilon b$ (with $\epsilon = 1/\alpha$), are depicted in Figure 1. If the number density per unit volume of these identical needles is n_2 , then (see Willis 1982) their volume fraction may be expressed in the form

$$c^{(2)} = \frac{4}{3} \pi n_2 b^3 \epsilon^2. \quad (20)$$

We note that, for a composite with fixed number density n_2 , the concentration $c^{(2)} \rightarrow 0$ as $\alpha \rightarrow \infty$ ($\varepsilon \rightarrow 0$). The "axial concentration" of these needles is represented by $n_2 b^3$.

Taking a fixed axial concentration $n_2 b^3$, an asymptotic expression for the effective shear stress in the matrix $\bar{\tau}^{(1)}$ is given by

$$\bar{\tau}^{(1)} = \sqrt{\left(1 + \frac{4n_2 b^3 \pi}{9 \ln \varepsilon}\right) \bar{\tau}_d^2 + \bar{\tau}_p^2 + \bar{\tau}_n^2}. \quad (21)$$

The corresponding effective stress potential for this nonlinear composite material is then given by stress (21) by

$$\tilde{U}(\bar{\sigma}) = \phi^{(1)} \left(\sqrt{\left(1 + \frac{4n_2 b^3 \pi}{9 \ln \varepsilon}\right) \bar{\tau}_d^2 + \bar{\tau}_p^2 + \bar{\tau}_n^2} \right). \quad (22)$$

This effective stress potential is dependent on all three deformation modes—which is consistent with discontinuous reinforcement. Note that in the limit as $\alpha \rightarrow \infty$ ($\varepsilon \rightarrow 0$), $\tilde{U}(\bar{\sigma}) \rightarrow \phi^{(1)}(\bar{\tau}_e)$, corresponding to the behavior of the matrix material. This is in sharp contrast with the continuous fiber-reinforced composite, discussed in the previous subsection, which remains rigid with respect to axisymmetric loading even as the concentration $c^{(2)} \rightarrow 0$. This is a consequence of the fact that the limit $\alpha \rightarrow \infty$ is singular (the limit depends on whether $c^{(2)}$ is taken to be finite or zero).

3.1.3 Laminated composites

In the limit as $\alpha \rightarrow 0$, with a fixed, finite inclusion concentration, we have, asymptotically, that

$$\bar{\tau}^{(1)} = \bar{\tau}_n, \quad (23)$$

which results in an effective stress potential

$$\tilde{U}(\bar{\sigma}) = c^{(1)} \phi^{(1)} \left(\frac{\bar{\tau}_n}{\sqrt{c^{(1)}}} \right). \quad (24)$$

This behavior is consistent with the behavior of a two-phase laminated composite with one rigid phase since the only mode of deformation possible in this case is shear along the plane of the layers. Thus, for fixed inclusion concentration, the above result is in agreement with the exact solution for multiple-phase laminated composites, derived by de Botton & Ponte Castañeda(1992).

3.1.4 Disk-strengthened composites

Next, we consider spheroidal inclusions which have radius a and height $2\alpha a$. As $\alpha \rightarrow 0$ the inclusions become flat disks. If the number density per unit volume is n_2 , then the volume fraction of the disks is

$$c^{(2)} = \frac{4}{3}\pi n_2 a^3 \alpha, \quad (25)$$

where $n_2 a^3$ represents the “transverse concentration” of disks.

The effective shear stress $\bar{\tau}^{(1)}$ for thin disks is obtained by substituting (25) into (14) and taking the limit (see Willis 1982) as $\alpha \rightarrow 0$, with n_2 fixed (note that $c^{(2)} \rightarrow 0$). Then, the leading order term in an asymptotic expansion as $\alpha \rightarrow 0$ is given by

$$\bar{\tau}^{(1)} = \sqrt{\left(1 - \frac{16n_2 a^3/9}{1 + 16n_2 a^3/9}\right) \bar{\tau}_d^2 + \left(1 - \frac{32n_2 a^3/9}{1 + 32n_2 a^3/9}\right) \bar{\tau}_p^2 + \bar{\tau}_n^2}. \quad (26)$$

The corresponding effective potential for the composite with thin disks is obtained by substituting (26) into (16), which leads to

$$\tilde{U}(\bar{\sigma}) = \phi^{(1)} \left(\sqrt{\left(1 - \frac{16n_2 a^3/9}{1 + 16n_2 a^3/9}\right) \bar{\tau}_d^2 + \left(1 - \frac{32n_2 a^3/9}{1 + 32n_2 a^3/9}\right) \bar{\tau}_p^2 + \bar{\tau}_n^2} \right). \quad (27)$$

We emphasize that in this case, unlike the case of the needle-reinforced composite, the effect of the ‘flat’ rigid disks on the overall response of the nonlinear composite is nontrivial. We note that an expression analogous to the above relation (in terms of the strain potential instead of the stress potential) has been developed by Talbot and Willis (1992).

3.2 Randomly oriented inclusions

The composite materials considered in this section are made up of a nonlinear, incompressible matrix reinforced by rigid, randomly oriented, spheroidal inclusions, as shown in Figure 1(b). Thus, the composite is macroscopically isotropic and incompressible. Therefore, the effective stress potential $\tilde{U}$ depends only on the mean effective shear stress $\bar{\tau}_e$ (dependence on the third invariant is neglected: see Duva 1984). To obtain an upper estimate for $\tilde{U}$, we observe that $\phi^{(2)} = 0$, and we make use of the variational statement (11) with $\tilde{U}_0$ given by relations (58) in Appendix A. The result is

$$\tilde{U}(\bar{\sigma}) = \max_{\mu^{(1)}} \left\{ \frac{1}{2\tilde{\mu}} \bar{\tau}_e^2 - c^{(1)} \max_{\tau_e} \left[\frac{1}{2\mu^{(1)}} \tau_e^2 - \phi^{(1)}(\tau_e) \right] \right\}, \quad (28)$$

which reduces to

$$\tilde{U}(\bar{\sigma}) = c^{(1)} \phi^{(1)} \left(\sqrt{\frac{\mu^{(1)}}{c^{(1)} \tilde{\mu}}} \bar{\tau}_e \right), \quad (29)$$

where $\frac{\tilde{\mu}}{\mu^{(1)}}$ is given by (56) in Appendix A. This equation expresses the effective stress potential of the nonlinear composite in terms of the stress potential of the matrix material $\phi^{(1)}$, and an estimate for the average stress in the matrix (which in turn depends on the inclusion geometry and concentration). Explicit expressions for the special cases of needle-reinforced and disk-reinforced composites may be obtained in a similar way as for the corresponding special cases for the composites with aligned inclusions. However, in the interest of brevity, these will not be given here.

4. APPLICATION TO COMPOSITES WITH POWER-LAW MATRIX MATERIALS

So far we have derived expressions for the effective properties of composites comprised of a *general* nonlinear matrix material reinforced by rigid spheroidal inclusions, which are

either aligned or randomly oriented. In this section, we specialize the previous results to composites with pure power-law behavior for the matrix. This class of composites has also been considered recently by He (1990) and Lee & Mear (1991), who generalized the calculations of Duva (1984) for spherical rigid particles to spheroidal inclusions. As mentioned earlier, their method considers an isolated rigid spheroidal inclusion embedded in an infinite power-law matrix material, which is subjected to *axisymmetric* remote loading with respect to the spheroidal inclusion. In contrast, our results account approximately for inclusion interaction (without the need of the differential self-consistent scheme) and general remote loading conditions. Duva & Storm (1989) have also obtained dilute concentration results for the plane strain deformation of fiber composites reinforced by rigid fibers. A brief comparison of the results of this paper with the numerical results of these other authors is given in Appendix C. Numerical results for the axisymmetric response of power-law materials reinforced by aligned spheroidal inclusions, with periodic microstructures, have also been given by Bao et al. (1991). Comparison with these results will be given, as far as possible, in the following discussion.

4.1 *Composites with aligned inclusions*

The power-law matrix material is described by the stress potential

$$U^{(1)}(\boldsymbol{\sigma}) = \phi^{(1)}(\tau_e) = \frac{\sigma_0}{n+1} \left(\frac{\tau_e}{\tau_0} \right)^{n+1}, \quad (30)$$

where τ_0 is an appropriate stress normalization factor, which has the interpretation of the yield stress in shear in the limit as $n \rightarrow \infty$, and where $\sigma_0 = \sqrt{3}\tau_0$.

The effective stress potential for the nonlinear composite is obtained by substituting (30)

into (16). The results is

$$\tilde{U}(\bar{\sigma}) = \left(c^{(1)}\right)^{(1-n)/2} \frac{\sigma_0}{n+1} \left(\frac{\bar{\tau}^{(1)}}{\tau_0}\right)^{n+1}, \quad (31)$$

where $\bar{\tau}^{(1)}$ is given by (14). The corresponding stress-strain relations are

$$\begin{aligned} \bar{\gamma}_d &= \sqrt{3} \left(c^{(1)}\right)^{(1-n)/2} \frac{\mu^{(1)}}{\tilde{\mu}_d} \left(\frac{\bar{\tau}^{(1)}}{\tau_0}\right)^{n-1} \frac{\bar{\tau}_d}{\tau_0}, \\ \bar{\gamma}_p &= \sqrt{3} \left(c^{(1)}\right)^{(1-n)/2} \frac{\mu^{(1)}}{\tilde{\mu}_p} \left(\frac{\bar{\tau}^{(1)}}{\tau_0}\right)^{n-1} \frac{\bar{\tau}_p}{\tau_0}, \\ \bar{\gamma}_n &= \sqrt{3} \left(c^{(1)}\right)^{(1-n)/2} \frac{\mu^{(1)}}{\tilde{\mu}_n} \left(\frac{\bar{\tau}^{(1)}}{\tau_0}\right)^{n-1} \frac{\bar{\tau}_n}{\tau_0}. \end{aligned} \quad (32)$$

These general results will be studied through several specific examples in the following sub-sections.

4.1.1 *Effective behavior under single-mode loading*

We study each of the three independent deformation modes of the incompressible, transversely isotropic composite first. If only axisymmetric loading $\bar{\tau}_d$ is applied to the composite, then equation (14) reduces to

$$\bar{\tau}^{(1)} = \left(\frac{\mu^{(1)}}{\tilde{\mu}_d}\right)^{1/2} \bar{\tau}_d. \quad (33)$$

Thus, the effective potential of the nonlinear composite $\tilde{U}$ may be written in the form

$$\tilde{U}(\bar{\sigma}) = \frac{(\tilde{\sigma}_0)_d}{n+1} \left(\frac{\bar{\tau}_d}{(\tilde{\tau}_0)_d}\right)^{n+1}, \quad (34)$$

where $(\tilde{\tau}_0)_d$ and $(\tilde{\sigma}_0)_d$ are the stress normalization factors in shear and tension, respectively, for axisymmetric loading, and are given by

$$\frac{\tau_0}{(\tilde{\tau}_0)_d} = \frac{\sigma_0}{(\tilde{\sigma}_0)_d} = \left(c^{(1)}\right)^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}_d}\right)^{(n+1)/2n}. \quad (35)$$

Here $\frac{\mu^{(1)}}{\tilde{\mu}_d}$ is given by (53). We note that as $n \rightarrow \infty$, $(\tilde{\sigma}_0)_d$ corresponds to the tensile flow stress of the composite under axisymmetric loading.

Similar results may be derived for transverse and longitudinal shear loading with $(\tilde{\tau}_0)_p$ and $(\tilde{\tau}_0)_n$ as the effective stress normalization factors in transverse and longitudinal shear, respectively. These may be expressed by the relations

$$\frac{\tau_0}{(\tilde{\tau}_0)_p} = (c^{(1)})^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}_p} \right)^{(n+1)/2n}, \quad (36)$$

$$\frac{\tau_0}{(\tilde{\tau}_0)_n} = (c^{(1)})^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}_n} \right)^{(n+1)/2n}. \quad (37)$$

In Figures 2 to 4, we show plots of the inverse of these three effective stress normalization factors for a linear ($n = 1$) and nonlinear ($n = 10$) matrix material, respectively, as functions of the inclusion concentration. The composites presented in these figures have inclusions with shapes that vary from oblate ($\alpha < 1$) to prolate ($\alpha > 1$) spheroids. We discuss next the effect of five different inclusion shapes ($\alpha = 0.01, 0.1, 1, 10, 50$) on the effective properties for the three different deformation modes.

Figures 2(a) and 2(b) show the effective behavior of composites with aligned rigid spheroidal inclusions under axisymmetric loading. It can be seen that both prolate and oblate spheroids give rise to stronger axisymmetric responses than spheres. We also observe that the response of prolate spheroids is relatively stiffer than that of oblate spheroids (e.g., needles with $\alpha = 50$ are stiffer than plates with $\alpha = 0.01$). Comparing Figures 2(a) and 2(b), it is evident that the effect of increasing material nonlinearity is to relax the strengthening influence of all particle shapes. For example, a linear composite ($n = 1$) containing a 10% volume fraction of prolate inclusions (with an aspect ratio $\alpha = 10$) is approximately 3.5 times as strong as

the matrix material. Similarly, if the inclusions have an aspect ratio $\alpha = 50$, the composite is approximately 50 times as strong as the matrix material. On the other hand, for a nonlinear composite ($n = 10$), these two inclusion shapes make the material approximately 2 and 6.6 times as strong as the matrix material, respectively. In the limit as $\alpha \rightarrow \infty$, the composite shows a rigid response ($(\tilde{\sigma}_0)_d \rightarrow \infty$) to axisymmetric loading. This is consistent with the behavior of composites reinforced by rigid fibers as discussed in Subsection 3.1.1. In the opposite limit as $\alpha \rightarrow 0$, $(\tilde{\sigma}_0)_d \rightarrow \infty$, which means that the composite also becomes rigid with respect to axisymmetric loading in this limit. This is consistent with the behavior of incompressible laminated materials reinforced by rigid layers as discussed in Subsection 3.1.3.

Figures 3(a) and 3(b) present the effective behavior of linear ($n = 1$) and nonlinear ($n = 10$) composites under transverse shear. It is found that the oblate spheroids ($\alpha = 0.01$) provide far greater strengthening than the prolate spheroids ($\alpha = 50$). This is because oblate spheroids have larger dimensions in the $\mathbf{x}_2 - \mathbf{x}_3$ plane (see Figure 1), and therefore resist this deformation mode more effectively. A nonlinear composite ($n = 10$) reinforced by 10% volume fraction of prolate inclusions (with $\alpha = 50$) is only 1.07 times as stiff as its matrix material, while, if it is reinforced by oblate inclusions (with $\alpha = 0.01$), it is 3.6 times as stiff as the matrix material. Also note that, a composite containing prolate inclusions with an aspect ratio $\alpha = 10$ has almost the same strength as a composite reinforced by prolate inclusions of aspect ratio $\alpha = 50$. As $\alpha \rightarrow \infty$, $(\tilde{\tau}_0)_p$ reaches a limit which almost coincides with its value at $\alpha = 50$. This limit inclusion shape provides a finite strengthening of the composite material, which is consistent with the behavior of rigid fiber-reinforced materials. In fact, prolate spheroids provide less stiffening than do spheres under transverse shearing.

In the opposite limit, as the inclusions become plate-like ($\alpha \rightarrow 0$), we obtain $(\tilde{\tau}_0)_p \rightarrow \infty$. The composite is thus rigid with respect to the transverse shear loading mode, which corresponds to the behavior of laminated materials reinforced by rigid layers. Finally, we note that under applied transverse shear loading, the fiber material can be treated as a plane strain problem. Duva & Storm (1989) have studied this plane strain problem numerically for the case of one rigid fiber in an infinite matrix. Comparing their dilute results with our results (as $\alpha \rightarrow \infty$), we find that there is little difference between the two sets of results. For example, if we compare the effective stress potential calculated by Duva & Storm with that of our explicit estimation, the difference is less than 5% for a nonlinear matrix (with $n = 10$), as discussed in some detail in Appendix C.

Figures 4(a) and 4(b) show the effect of rigid inclusions on the effective strength under longitudinal shear ($\bar{\tau}_n$) for a linear ($n = 1$) and a highly nonlinear matrix ($n = 10$). For this type of loading, we find that spherical inclusions provide the largest strengthening (of these five inclusion shapes) while oblate spheroids provide the least. It is interesting to note, that in the limit as $\alpha \rightarrow \infty$, the composite has the same strength under transverse shear loading (discussed in the previous paragraph) as under longitudinal shear ($(\tilde{\tau}_0)_n$ and $(\tilde{\tau}_0)_p$ reach the same limit). Since, for oblate inclusions, the longitudinal shear mode corresponds to a shearing deformation in a plane normal to their axis of symmetry, the strengthening effect of these rigid oblate spheroidal inclusions is significantly less for this deformation mode than for the other two deformation modes discussed above. In the limit of $\alpha \rightarrow 0$, the oblate inclusions become flat layers and the shearing deformation is concentrated along the matrix material between these rigid flat layers. This corresponds precisely to the response of laminated materials (with one phase rigid) which have only the longitudinal shear mode

available, as discussed in Section 3.1.3. Thus, of the five inclusion shapes studied here, oblate and prolate inclusion shapes provide less resistance to the longitudinal shear than spherical inclusions.

Figure 5(a) gives results for all three deformation modes (axisymmetric loading, transverse shear and longitudinal shear) in the perfectly-plastic limit ($n \rightarrow \infty$). For each loading mode, the ratio of the matrix flow stress to the effective flow stress ($\tau_0/(\tilde{\tau}_0)_d$, $\tau_0/(\tilde{\tau}_0)_p$, $\tau_0/(\tilde{\tau}_0)_n$) is plotted as a function of α , with a fixed inclusion concentration $c^{(2)} = 0.2$. For the axisymmetric deformation mode, prolate spheroids are stiffer than oblate spheroids if we compare a prolate shape with aspect ratio α with the oblate shape which has a reciprocal aspect ratio $1/\alpha$. The weakest reinforcement is given by the oblate shape with $\alpha \approx 0.64$, which is even less effective in increasing the axisymmetric strength than the spherical shape ($\alpha = 1$). On the other hand, for the transverse deformation mode, the largest reinforcement is given by the oblate shape with aspect ratio $\alpha = 0$. As α increases from 0 to ∞ , the strengthening effect decreases continuously. In the limit as $\alpha \rightarrow \infty$, the strength of the composite approaches the transverse shearing strength of fiber-reinforced composites. The reinforcement provided by rigid inclusions to the longitudinal shear mode is (generally) weaker than that for the other two deformation modes. As $\alpha \rightarrow 0$, the inclusions have no reinforcing effect on the flow strength of the composite. In the opposite limit as $\alpha \rightarrow \infty$, the longitudinal shear flow strength approaches that of the fiber-reinforced materials. It is noted that the strongest reinforcement for this deformation mode is given by a prolate shape with $\alpha \approx 1.4$.

Thus, for composites with aligned rigid spheroidal inclusions, there is no inclusion shape which provides optimal strengthening for all of the three independent deformation modes.

Needles give a stronger response (strongest overall) for one mode (axisymmetric mode), whereas disks provide significant reinforcement for two modes (axisymmetric mode and transverse shear mode). Thus, the optimal choice of particle shape in a given application depends on the particular service load to which the composite will be subjected.

Bao et al. (1991) have carried out a study of the tensile flow stress of a two-phase composite with periodic microstructures using unit-cell, finite-element calculations. While the matrix material, inclusion shape and loading mode utilized by these authors are identical to those utilized in this work, the distribution of inclusions in their work is periodic. With this difference in mind, we compare the effect of spheroidal inclusion shape on the axisymmetric tensile flow stress of composites with inclusion concentration $c^{(2)} = 0.1$ (our estimates may be obtained from Figure 5(b)). For rigid spherical inclusions in a perfectly plastic matrix ($n \rightarrow \infty$), the effective tensile flow stress predicted by our method is $\tilde{\sigma}_0 / \sigma_0 = 1.0695$, while the corresponding result of Bao et al. (for a periodic microstructure) is $\tilde{\sigma}_0 / \sigma_0 = 1.025$ (from the curves in Figure 6 of their paper). For an aspect ratio $\alpha = 0.1$ (or 10), the tensile flow stresses shown in Figure 5(b) is $\tilde{\sigma}_0 / \sigma_0 = 1.2$ (or 1.742). The corresponding result of Bao et al. is $\tilde{\sigma}_0 / \sigma_0 = 1.36$ (or 1.62) (from the curves in Figure 6 of their paper). Thus the predictions of both methods are in qualitative agreement for these three inclusion shapes.

Finally, we note, for completeness, that for the special case of disk-reinforced composites we obtain the following simple expressions for the effective stress normalization factors in the three different modes:

$$\frac{\tau_0}{(\tilde{\tau}_0)_d} = \left(\frac{1}{1 + 16n_2 a^3 / 9} \right)^{(n+1)/2n}, \quad (38)$$

$$\frac{\tau_0}{(\tilde{\tau}_0)_p} = \left(\frac{1}{1 + 32n_2 a^3 / 9} \right)^{(n+1)/2n}, \quad (39)$$

$$\frac{\tau_0}{(\tilde{\tau}_0)_n} = 1. \quad (40)$$

We observe that the longitudinal shear stress normalization factor (40) is unity indicating that the longitudinal shear mode is not affected by the presence of thin disk-like inclusions. On the other hand, the axisymmetric and transverse shear modes (38 and 39) are both sensitive to the presence of thin disks through the parameter $n_2 a^3$. If this parameter $n_2 a^3 \rightarrow \infty$, the material becomes effectively rigid under these two modes of deformation, as expected. Corresponding results can also be easily written down for needle-reinforced composites.

4.1.2 *Effective behavior under multiple-mode loading*

An interesting difference between the effective behavior of linear and nonlinear composite materials is depicted in Figures 6 and 7, where the coupling between different deformation modes is analyzed for the nonlinear composites. As is well-known, the effective response of linear, incompressible fiber-reinforced composites with transversely isotropic symmetry is such that there is no coupling between the three independent modes of deformation available for such materials (see (52)). However, this is not the case for nonlinear materials. For example, if we preload a nonlinear composite (matrix with $n = 10$ and aligned prolate spheroids with $\alpha = 10$ and $c^{(2)} = 0.2$) with axisymmetric stresses $\bar{\tau}_d/\tau_0 = 0.1, 2.8$ and 3.5 , the deformation of the nonlinear composite is correspondingly characterized by strains $\bar{\gamma}_d$ as given by (32) with (33). If we now increase the transverse shear stress $\bar{\tau}_p$, we observe (Figure 6) a significant further increase in the axisymmetric strain $\bar{\gamma}_d$. By comparison, the corresponding linear composite would not show further increase of the axisymmetric strain $\bar{\gamma}_d$ under increasing transverse shear stress $\bar{\tau}_p$. This example clearly illustrates the significant coupling between the different loading modes for nonlinear composites.

Another example is given in Figure 7, which shows plots the axisymmetric stress $\bar{\tau}_d$ versus

the axisymmetric shear strain $\bar{\gamma}_d$ for the same composite discussed above. In this figure, however, the composite has been preloaded with three different values of the transverse shear stress ($\bar{\tau}_p/\tau_0 = 0.0, 1.0, 1.5$). We observe that the effect of the transverse shear preload is to significantly reduce the axisymmetric load carrying capability of the composite.

4.2 Composites with randomly oriented inclusions

In the previous section, we observed that, for aligned spheroidal inclusions, there is no aligned rigid spheroidal inclusion shape which provides optimal strengthening for all deformations. In this section, we study the shape effect for randomly oriented inclusions. By substituting the general power-law material description (30) into the effective potential for nonlinear materials reinforced with randomly oriented rigid spheroids (29), this effective stress potential may be written in the form

$$\tilde{U}(\bar{\sigma}) = \frac{\tilde{\sigma}_0}{n+1} \left(\frac{\bar{\sigma}_e}{\tilde{\sigma}_0} \right)^{n+1}, \quad (41)$$

where $\bar{\sigma}_e (= \sqrt{3}\bar{\tau}_e)$ is the effective average stress, and where the effective stress normalization factor $\tilde{\sigma}_0$ is given by

$$\frac{\sigma_0}{\tilde{\sigma}_0} = \left(c^{(1)} \right)^{(1-n)/2n} \left(\frac{\mu^{(1)}}{\tilde{\mu}} \right)^{(n+1)/2n}, \quad (42)$$

with $\tilde{\mu}/\mu^{(1)}$ given in turn by (56).

Figures 8 show plots $\sigma_0/\tilde{\sigma}_0$ versus inclusion concentration $c^{(2)}$ for a linear ($n = 1$) and a strongly nonlinear composites ($n = 10$) with randomly oriented rigid spheroidal inclusions. It can be seen that prolate spheroids with $\alpha = 50$ are close to oblate spheroids with $\alpha = 0.01$ (the prolate spheroids are slightly stiffer). This similarity in behavior is in marked contrast with the very different behaviors displayed by aligned prolate and oblate spheroids under different loading modes. Also, while the spherical inclusions result in the least strengthening

for a given concentration in these isotropic materials, there are loading modes (see Figure 4) for which spherical inclusions produce stronger effective behavior than aligned prolate or oblate spheroids (at a given concentration).

A study on the tensile flow stress of a composite reinforced by rigid, randomly oriented, packet morphologies has been carried out by Bao et al. (1991) using finite element calculations. While it is not possible to make direct comparisons of our results with theirs, there are qualitative similarities in some of the findings. For example, we find, just like Bao et al., that the inclusion shape with $\alpha = 10$ provides stronger reinforcement than the inclusion shape with $\alpha = 0.1$.

It is interesting to examine the limits as $\alpha \rightarrow 0$ and $\alpha \rightarrow \infty$, with a fixed, *finite* inclusion concentration $c^{(2)}$. From (42), we obtain, respectively, the following effective stress normalization factors

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim (c^{(1)})^{1/n} \left(\frac{3\pi\alpha}{4c^{(2)}} \right)^{(n+1)/2n}, \quad \text{as } \alpha \rightarrow 0; \quad (43)$$

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim (c^{(1)})^{1/n} \left(\frac{15 \ln(2\alpha)}{c^{(2)}\alpha^2} \right)^{(n+1)/2n}, \quad \text{as } \alpha \rightarrow \infty. \quad (44)$$

We observe that, in both of these limits, the stress normalization factor $\tilde{\sigma}_0$ becomes unbounded. As discussed in Section 3, these two limits correspond to rigid fibers and rigid flat layers, respectively. Thus, in these limits, it is clear that an orientational average will lead to perfectly rigid behavior for the composite. A more sensible approach is to consider the limits as $\alpha \rightarrow 0$ and $\alpha \rightarrow \infty$, with fixed density of the inclusions (n_2). These limits correspond to the cases of randomly oriented disks and needles, respectively. Thus the effective stress normalization factor for the case of thin needles (with aspect ratio α) is given by

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim \left[1 - \frac{2\pi n_2 b^3/3}{5(1 + 6 \ln(2\alpha)) + 2\pi n_2 b^3/3} \right]^{(n+1)/2n}, \quad (45)$$

and the corresponding factor for disks is given by

$$\frac{\sigma_0}{\tilde{\sigma}_0} \sim \left(1 - \frac{16n_2a^3}{9 + 16n_2a^3}\right)^{(n+1)/2n}. \quad (46)$$

We note that infinitely thin needles provide no reinforcement, whereas infinitely thin disks do provide some finite reinforcement (depending on the density of disks).

5. CONCLUSIONS

In this paper, we have studied the effective constitutive behavior of composites made up of incompressible, ductile materials reinforced by either aligned, or randomly oriented, rigid spheroidal inclusions. The method of solution is based on the variational principle of Ponte Castañeda(1991a) for nonlinear inhomogeneous media. When the rigid inclusions are aligned, the resulting composites are transversely isotropic (and incompressible), and characterized by three independent modes of deformation – axisymmetric tension, transverse shear and longitudinal shear. When the rigid inclusions are randomly oriented, the resulting composites are isotropic (and incompressible), and are (approximately) characterized by one deformation mode (e.g., tensile loading).

For the aligned-spheroid composites, the axisymmetric loading mode has been studied extensively in the recent past. However, the transverse shear and longitudinal shear modes of deformation for these composites have hardly received any attention. In this work, we have obtained *explicit* expressions for the constitutive response of these composites under *general* loading conditions (including transverse and longitudinal shear loading), and for finite (nondilute) concentrations of the inclusions. In addition, simple expressions have been obtained for the special cases of aligned rigid needles and disks. The results show that the

optimal choice for the shape of the particle reinforcement, within this class of composites, depends strongly on the service load to which the composite will be subjected. We also note that the nonlinear aligned-spheroid composites exhibit strong coupling between the three possible modes of deformation for these composites. By comparison, such coupling is not present for the corresponding linear composites. For composites reinforced by randomly oriented spheroidal inclusions, we find that both the prolate and oblate spheroidal inclusions provide significant reinforcement. We conclude by noting that more complicated microstructures that the ones considered in this work may eventually lead to metal-matrix composites with superior effective properties. We hope that the method developed in this work will serve as a useful tool in the optimal design of such microstructures.

Acknowledgements

This research has been supported by the Air Force Office of Scientific Research under Grant No. 91-0161. The first author would like to acknowledge valuable discussions with K. Sarkar.

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APPENDIX A: LINEAR COMPOSITES WITH RIGID SPHEROIDAL INCLUSIONS

A.1 Composites with aligned inclusions

The results given here are based on the works of Hill (1965), Walpole (1966a,b, 1969) and Willis (1977) (see also Willis 1982 and Qiu and Weng 1990) on linear, anisotropic composite materials. Consider a two-phase composite material, as depicted in Figure 1, in which the spheroidal inclusions are either aligned in $\mathbf{x}_1$ direction or randomly oriented. The matrix and the stiffer inclusion materials are labeled phase 1 and 2, respectively. Also, the volume fraction of phase 1 and phase 2 are denoted by $c^{(1)}$ and $c^{(2)}$ ($c^{(1)} + c^{(2)} = 1$). In the following discussion, fourth-order symmetric tensors will be denoted by upper-case italicized Roman letters, while second order tensors will be denoted by bold-face, Greek letters.

The mechanical properties of each phase will be characterized by fourth-order compliance tensors, which, using Hill's convention, may be written in the form

$$M^{(r)} = \left(\frac{1}{3k^{(r)}}, \frac{1}{2\mu^{(r)}} \right), \quad (r = 1, 2), \quad (47)$$

where $k^{(r)}$ and $\mu^{(r)}$ are the bulk modulus and shear modulus, respectively, of the r th-phase material.

Then, the effective compliance tensor $\tilde{M}$ of the two-phase, linear composite may be expressed in the form

$$\tilde{M} = \sum_{r=1}^2 c^{(r)} M^{(r)} B^{(r)} \left\{ \sum_{s=1}^2 c^{(s)} B^{(s)} \right\}^{-1}, \quad (48)$$

where $B^{(r)}$ is the stress concentration factor tensor (Hill, 1965), given by

$$B^{(r)} = \left[I + (M^{(0)})^{-1} (I - S^{(0)}) (M^{(r)} - M^{(0)}) \right]^{-1}, \quad (49)$$

In this last relation, $M^{(0)}$ is the compliance tensor of a comparison material, I is the fourth-order identity tensor, and $S^{(0)}$ is the Eshelby's tensor corresponding to a matrix made of the comparison material. For a transversely isotropic material, $S^{(0)}$ may be expressed, in Walpole's notation, as

$$S^{(0)} = (S_{2222}^{(0)} + S_{2233}^{(0)}, S_{1111}^{(0)}, 2S_{2323}^{(0)}, 2S_{1212}^{(0)}, S_{1122}^{(0)}, S_{2211}^{(0)}), \quad (50)$$

where the relevant components are given explicitly in Appendix B.

In order to obtain the required upper bound for the effective compliance (a lower bound for the stiffness) of the linear composite, the compliance of the comparison material $M^{(0)}$ in (48) and () must be chosen to be the less stiff phase (i.e., $M^{(0)} = M^{(1)}$). Then, after some algebra, (48) may be rewritten in the form

$$\tilde{M} = M^{(1)} + c^{(2)} \left[\left(M^{(2)} (M^{(1)})^{-1} - I \right)^{-1} + c^{(1)} (I - S^{(1)}) \right]^{-1} M^{(1)}, \quad (51)$$

where $S^{(1)}$ is the Eshelby tensor with phase-1 as the matrix material. Finally, since the inclusions are rigid (i.e., $k^{(2)} \rightarrow \infty$, and $\mu^{(2)} \rightarrow \infty$, so that $M^{(2)} = (0, 0)$), and the matrix material is incompressible (i.e., $k^{(1)} \rightarrow \infty$), the transversely isotropic tensor, $\tilde{M}$, reduces to

$$\tilde{M} = \left(\frac{1}{2\tilde{\mu}_d}, \frac{1}{\tilde{\mu}_d}, \frac{1}{2\tilde{\mu}_p}, \frac{1}{2\tilde{\mu}_n}, -\frac{1}{2\tilde{\mu}_d}, -\frac{1}{2\tilde{\mu}_d} \right), \quad (52)$$

where

$$\begin{aligned} \frac{\mu^{(1)}}{\tilde{\mu}_d} &= 1 - \frac{c^{(2)}}{1 - c^{(1)} f(\alpha)}, \\ \frac{\mu^{(1)}}{\tilde{\mu}_p} &= 1 - \frac{c^{(2)}}{c^{(2)} + 2c^{(1)} S_{2323}^{(1)}}, \\ \frac{\mu^{(1)}}{\tilde{\mu}_n} &= 1 - \frac{c^{(2)}}{c^{(2)} + 2c^{(1)} S_{1212}^{(1)}}. \end{aligned} \quad (53)$$

Here f is a function of the inclusion aspect ratio α , and is given in Appendix B. Thus, the effective stress potential $\tilde{U}_0$ of the linear composite has the form (see Walpole 1969; deBotton & Ponte Castañeda 1992)

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\tilde{\mu}_d}\bar{\tau}_d^2 + \frac{1}{2\tilde{\mu}_p}\bar{\tau}_p^2 + \frac{1}{2\tilde{\mu}_n}\bar{\tau}_n^2, \quad (54)$$

where $\bar{\tau}_d, \bar{\tau}_p$ and $\bar{\tau}_n$ are the three transversely isotropic invariants of the applied stress tensor $\bar{\sigma}$, corresponding to the three independent deformation modes for an incompressible, transversely isotropic material: axisymmetric stress, transverse shear stress and longitudinal shear stress, respectively (see Appendix B).

A.2 Composites with randomly oriented inclusions

When the inclusions in the composite are randomly oriented, the expression for the effective compliance of the composite (48) must be replaced by (this is based on the orientation average of strain field, as in Wu (1966) and Walpole (1966b))

$$\tilde{M} = \left(c^{(1)}M^{(1)} + c^{(2)}M^{(2)} \{B^{(2)}\} \right) \left(c^{(1)}I + c^{(2)}\{B^{(2)}\} \right)^{-1}. \quad (55)$$

where $\{B^{(2)}\}$ denotes the orientational average of tensor $B^{(2)}$. We note that on account of the incompressibility and isotropy of the composite, we may write $\tilde{M} = \left(0, \frac{1}{2\tilde{\mu}} \right)$. Some complicated algebra leads to the following result for the effective shear modulus of the composite, appropriately normalized, namely,

$$\frac{\mu^{(1)}}{\tilde{\mu}} = \frac{5c^{(1)}(1 - e_s)(1 - f_s)[3 + 2(g_s + h_s) - (2d_s + c_s)]}{A_s(g_s + h_s) + B_se_sf_s + C_s(e_s + f_s) + D_s} \quad (56)$$

where

$$c_s = 1 - (S_{2222}^{(1)} + S_{2233}^{(1)}), \quad d_s = 1 - S_{1111}^{(1)},$$

$$\begin{aligned}
e_s &= 1 - 2S_{2323}^{(1)}, & f_s &= 1 - 2S_{1212}^{(1)}, \\
g_s &= -S_{1122}^{(1)}, & h_s &= -S_{2211}^{(1)}. \\
A_s &= 10c^{(1)}e_sf_s - 2(5 - 3c^{(2)})(e_s + f_s) + 2(5 - c^{(2)}), \\
B_s &= 3(5 - 4c^{(2)}) - 5c^{(1)}(2d_s + c_s), \\
C_s &= (5 - 3c^{(2)})(2d_s + c_s) - 3(5 - 2c^{(2)}), \\
D_s &= -(5 - c^{(2)})(2d_s + c_s) + 15.
\end{aligned} \tag{57}$$

Then, the corresponding potential function for the isotropic composite is

$$\tilde{U}_0(\bar{\sigma}) = \frac{1}{2\tilde{\mu}}\bar{\tau}_e^2. \tag{58}$$

APPENDIX B

Letting the x_1 coordinate axis define the axis of transverse isotropy (see Figure 1(c)), the three transversely isotropic invariants of the stress tensor σ are given by

$$\begin{aligned}\tau_d^2 &= \frac{1}{3} \left[\frac{1}{2}(\sigma_{33} + \sigma_{22}) - \sigma_{11} \right]^2, \\ \tau_p^2 &= \sigma_{23}^2 + \frac{1}{4}(\sigma_{33} - \sigma_{22})^2, \\ \tau_n^2 &= \sigma_{12}^2 + \sigma_{13}^2.\end{aligned}\tag{59}$$

Correspondingly, the three transversely isotropic invariants of the strain tensor *strain* are given by

$$\begin{aligned}\gamma_d^2 &= \frac{4}{3} \left[\frac{1}{2}(\epsilon_{33} + \epsilon_{22}) - \epsilon_{11} \right]^2, \\ \gamma_p^2 &= 4 \left[\epsilon_{23}^2 + \frac{1}{4}(\epsilon_{33} - \epsilon_{22})^2 \right], \\ \gamma_n^2 &= 4(\epsilon_{12}^2 + \epsilon_{13}^2).\end{aligned}\tag{60}$$

Also, for a spheroidal inclusion, aligned with the x_1 axis, the components of the Eshelby's tensor are (Eshelby, 1957)

$$\begin{aligned}S_{1111}^{(0)} &= \frac{1}{2(1-\nu^{(0)})} \left\{ 1 - 2\nu^{(0)} + \frac{3\alpha^2 - 1}{\alpha^2 - 1} - \left[1 - 2\nu^{(0)} + \frac{3\alpha^2}{\alpha^2 - 1} \right] g \right\}, \\ S_{2222}^{(0)} = S_{3333}^{(0)} &= \frac{3}{8(1-\nu^{(0)})} \frac{\alpha^2}{\alpha^2 - 1} + \frac{1}{4(1-\nu^{(0)})} \left[1 - 2\nu^{(0)} - \frac{9}{4(\alpha^2 - 1)} \right] g, \\ S_{2233}^{(0)} = S_{3322}^{(0)} &= \frac{1}{4(1-\nu^{(0)})} \left\{ \frac{\alpha^2}{2(\alpha^2 - 1)} - \left[1 - 2\nu^{(0)} + \frac{3}{4(\alpha^2 - 1)} \right] g \right\}, \\ S_{2211}^{(0)} = S_{3311}^{(0)} &= -\frac{1}{2(1-\nu^{(0)})} \frac{\alpha^2}{\alpha^2 - 1} + \frac{1}{4(1-\nu^{(0)})} \left\{ \frac{3\alpha^2}{\alpha^2 - 1} - (1 - 2\nu^{(0)}) \right\} g, \\ S_{1122}^{(0)} = S_{1133}^{(0)} &= \frac{1}{2(1-\nu^{(0)})} \left\{ -1 + 2\nu^{(0)} - \frac{1}{\alpha^2 - 1} + \left[1 - 2\nu^{(0)} + \frac{3}{2(\alpha^2 - 1)} \right] g \right\},\end{aligned}\tag{61}$$

$$S_{1212}^{(0)} = S_{1313}^{(0)} = \frac{1}{4(1 - \nu^{(0)})} \left\{ 1 - 2\nu^{(0)} - \frac{\alpha^2 + 1}{\alpha^2 - 1} - \frac{1}{2} \left[1 - 2\nu^{(0)} - \frac{3(\alpha^2 + 1)}{\alpha^2 - 1} \right] g \right\},$$

$$S_{2323}^{(0)} = \frac{S_{2222}^{(0)} - S_{2233}^{(0)}}{2},$$

where $\nu^{(0)}$ is the Poisson's ratio of the matrix material, $\alpha = b/a$ is the aspect ratio of the spheroidal inclusion in Figure 1(c), and g is given by

$$g = \frac{\alpha}{(\alpha^2 - 1)^{3/2}} \left[\alpha(\alpha^2 - 1)^{1/2} - \cosh^{-1} \alpha \right], \quad \alpha > 1, \quad \text{prolate};$$

$$\frac{\alpha}{(1 - \alpha^2)^{3/2}} \left[\cos^{-1} \alpha - \alpha(1 - \alpha^2)^{1/2} \right], \quad \alpha < 1, \quad \text{oblate.} \quad (62)$$

Finally, the function $f(\alpha)$ in equation (53) is given by

$$f(\alpha) = \frac{2\alpha^2 + 1}{\alpha^2 - 1} \left[\frac{3}{2}g(\alpha) - 1 \right], \quad (63)$$

where g is the function of α given by (62).

APPENDIX C: COMPARISON WITH AVAILABLE DILUTE RESULTS

For a composite comprised of an incompressible, pure power-law material reinforced by aligned, rigid spheroidal inclusions, we have seen that any external loading state may be represented in terms of three independent loading modes (Section 4). For any one of these loading modes ($\bar{\tau} = \bar{\tau}_d, \bar{\tau}_p$ or $\bar{\tau}_n$), the effective nonlinear energy function may be expressed in the form (see, for example, (34) for the axisymmetric mode, $\bar{\tau} = \bar{\tau}_d$)

$$\tilde{U}(\bar{\sigma}) = F(c^{(2)}; n, \alpha) \frac{\sigma_0}{n+1} \left(\frac{\bar{\tau}}{\tau_0} \right)^{n+1}, \quad (64)$$

where the function F reflects the reinforcing effect of the rigid inclusions.

For dilute concentrations of inclusions, a formal expansion in the concentration $c^{(2)}$ leads to the relation

$$F \approx 1 - c^{(2)} g(n, \alpha), \quad (65)$$

where g is a function of the nonlinearity n and inclusion aspect ratio α . Such an expansion is valid when the product of $c^{(2)}$ and g is much smaller than unity. Since it can be shown that the function g becomes unbounded in the limits as α approaches zero or infinity, it is useful to note that the range of validity (in $c^{(2)}$) of the dilute expansion (65) approaches zero as α approaches zero or infinity. This is in agreement with our previous comment that the limit as $c^{(2)} \rightarrow 0$ and $\alpha \rightarrow 0, \infty$ is singular (see Section 3.1.2). One must also be careful with the limit as $n \rightarrow \infty$ in the above expansion.

Next we compare our analytical estimates for F , in the case of axisymmetric loading, with corresponding dilute numerical results of He (1990) and Lee & Mear (1991a). These numerical results are based on the computation of the effective energy function $\tilde{U}$ of a composite made of one inclusion embedded in an infinite power-law matrix material.

Figure 9 shows a comparison, for a strongly nonlinear composite ($n = 10$), between our analytical estimates of F (they appear as curved lines) and the dilute numerical estimates of F , based on expansion (65), using the results of Lee & Mear (1991a) for the function g (they appear as straight lines). We observe that our results initially (for small $c^{(2)}$) lie slightly below the dilute estimates; however, the two curves intersect at a certain critical value of $c^{(2)}$, after which the two curves quickly diverge. The following observations are in order.

The intersection point, which serves as a measure of the range of validity of the dilute approximation, occurs for smaller and smaller values of $c^{(2)}$ as the aspect ratio of the inclusion increases, or decreases, from unity. This is in agreement with the discussion following equation (65).

Although the initial slopes of the dilute and nondilute curves are different, and this difference becomes larger as the aspect ratio of the inclusions increases, or decreases, from unity, we observe that the maximum difference between the actual values of the two curves (before they intersect) is not very large. Since the limit as $c^{(2)} \rightarrow 0$ and $\alpha \rightarrow 0, \infty$ is singular, it makes more sense to compare this difference in the actual values of the estimates for the function F (a direct measure of the reinforcing effect of the inclusions), and not the differences in the slopes. This is because, although the difference in the slopes may be large, the regions over which the dilute approximation holds are small. This leads to relatively small differences in the actual values of the functions. In Figure 10, we depict plots of this maximum relative difference, denoted by Δ_{rel} , as functions of the aspect ratio of the inclusions α , for the dilute results of both He (1990) and Lee & Mear (1991a). Thus we find that for values of α between 0.1 and 10, the maximum relative difference varies roughly

between 10 and 20 percent. For values of α outside this range, this type of comparison is not very useful because, at finite inclusion volume fractions $c^{(2)}$, the inclusions become so long (or wide) that the limits of continuous reinforcement are approached (fiber-reinforced and laminated composites). Thus, for these extreme limits, expressions of the type of (22) and (27), for fixed density (zero volume fraction) of thin needles and disks, respectively, are more appropriate.

An alternative comparison that may be made is with the dilute results of Duva & Storm (1989) for composites reinforced by rigid fibers. In this case, the axisymmetric mode is rigid, and these authors considered the transverse shear mode $\bar{\tau}_p$ (plane strain deformation perpendicular to the fibers). Thus, they were able to compute the function $g(n, \alpha)$ appearing in (65) for the transverse shear mode of fiber-reinforced composites. The corresponding relative difference Δ_{rel} between their numerical results and our analytical results (19) for fibers is shown in Table 1, as a function of the nonlinearity parameter n . We find that the maximum difference, occurring for $n = 10$, is less than five percent, which corresponds to excellent agreement between the two sets of results.

In summary, we find relatively good agreement between our analytical results and available numerical results for dilute concentrations of inclusions, provided that we stay away from singular limits (such as the limit of long fibers for the axisymmetric mode), in which case the differences can become large (although not very relevant, physically). The strength of our results, however, lies in their generality and relative simplicity.

Table 1. Maximum relative difference between our analytical results and the dilute numerical results of Duva & Storm (1989) for the transverse mode in fiber-reinforced composites.

n	1	2	3	4	5	6	7	8	9	10
Δ_{rel}	0	.0053	.0154	.0198	.0268	.0315	.0357	.0377	.0414	.0458

List of captions

Figure 1. Composite materials reinforced by spheroidal inclusions. (a) A composite with aligned spheroidal inclusions; (b) a composite with randomly oriented spheroidal inclusions; (c) a description of prolate and oblate spheroids.

Figure 2. The effect of inclusion shape on the effective linear and nonlinear stress normalization factors for aligned-inclusion composites subjected to axisymmetric loading. Note that, since the reinforcing phase is rigid, the reciprocal of strength $1/(\tilde{\sigma}_0)_d$ is plotted for convenience. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 3. The effect of inclusion shape on the effective linear and nonlinear stress normalization factors for aligned-inclusion composites subjected to transverse shear loading. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 4. The effect of inclusion shape on the effective linear and nonlinear stress normalization factors for aligned-inclusion composites subjected to longitudinal shear loading. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 5. A comparison of the effective flow stresses of the three independent loading modes for perfectly plastic composites reinforced by aligned spheroidal inclusions: (a) effective flow stresses versus aspect ratio (for $c^{(2)} = 0.2$); (b) effective flow stresses versus inclusion

concentration $c^{(2)}$ (for $\alpha = 0.1, 1$ and 10).

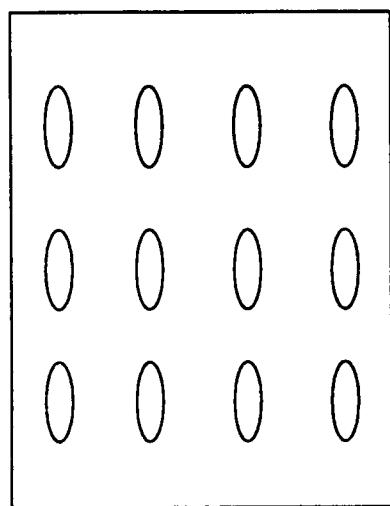
Figure 6. Plot of the axisymmetric strain $\bar{\gamma}_d$ versus the transverse shear stress $\bar{\tau}_p$ for an aligned-inclusion composite with three different axisymmetric preloadings, namely, $\bar{\tau}_d/\tau_0 = 0.1, 2.8$ and 3.5 .

Figure 7. Plot of the axisymmetric strain $\bar{\gamma}_d$ versus the axisymmetric stress $\bar{\tau}_d$ an aligned-inclusion composite with three different transverse shear preloadings, namely, $\bar{\tau}_p/\tau_0 = 0, 1.0$ and 1.5 .

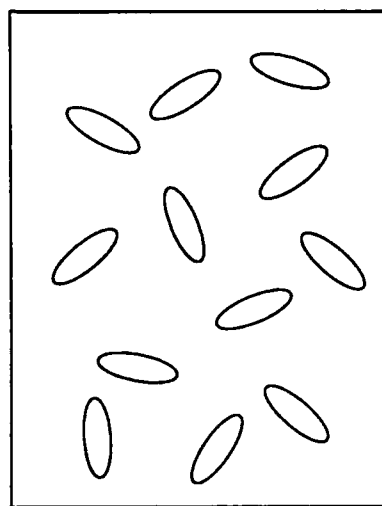
Figure 8. Plots of the reciprocal of the effective stress normalization factors for composites reinforced by randomly oriented spheroidal inclusions versus inclusion concentration $c^{(2)}$. (a) Composites with a linear matrix ($n = 1$); (b) composites with a nonlinear matrix ($n = 10$).

Figure 9. A comparison of our analytical prediction for the function F with the dilute calculations of Lee & Mear (1991a). The later results appear as straight lines; the former as curved lines.

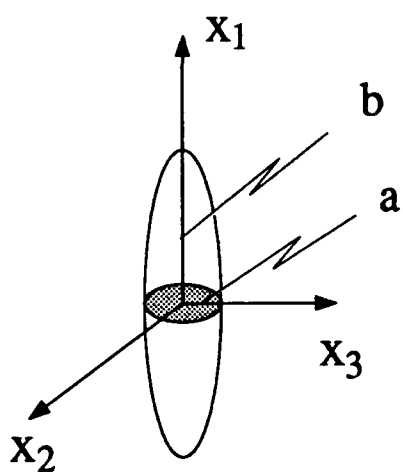
Figure 10. Plots of the largest relative difference Δ_{rel} between our results and the dilute results of He (1990) and Lee & Mear (1991a), respectively, versus aspect ratio α .



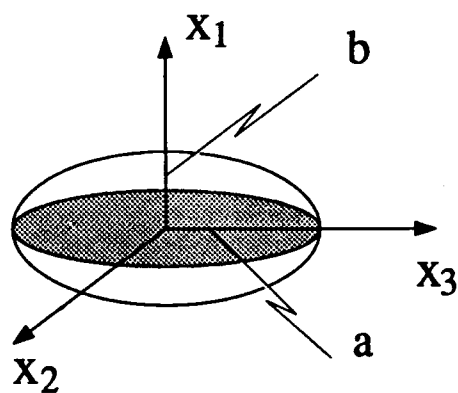
(a)



(b)



Prolate



Oblate

(c)

Fig. 1

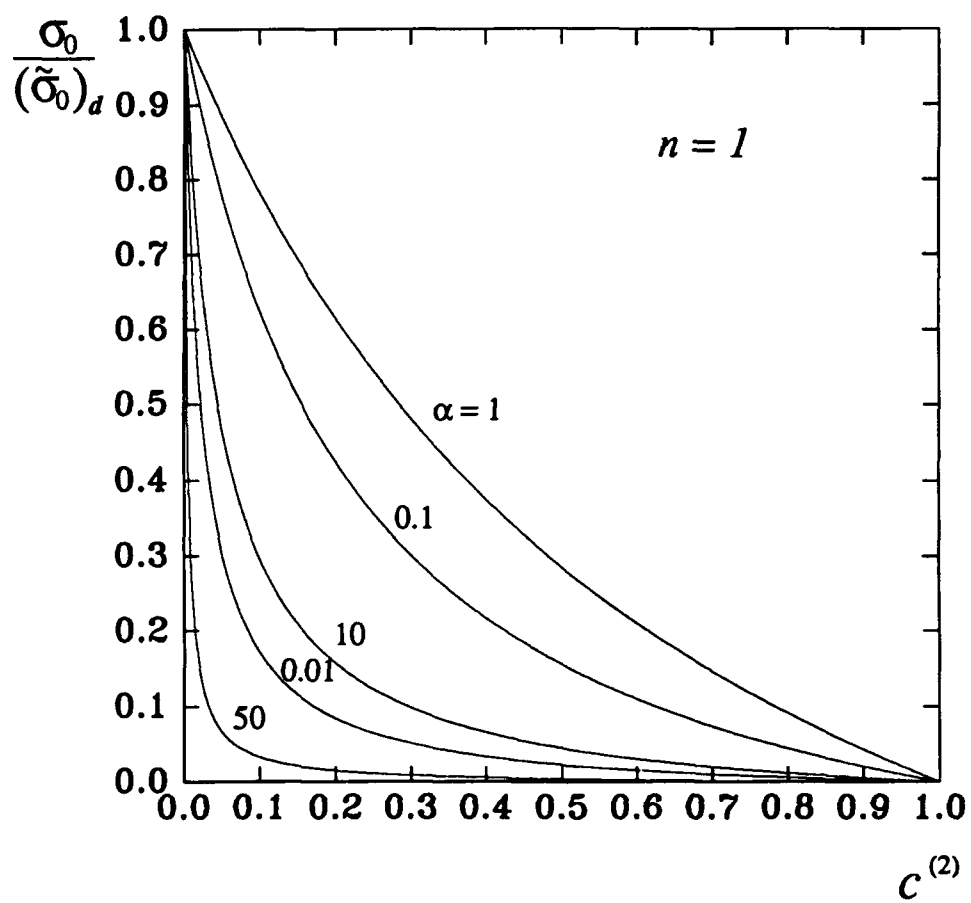


Fig. 2(a)

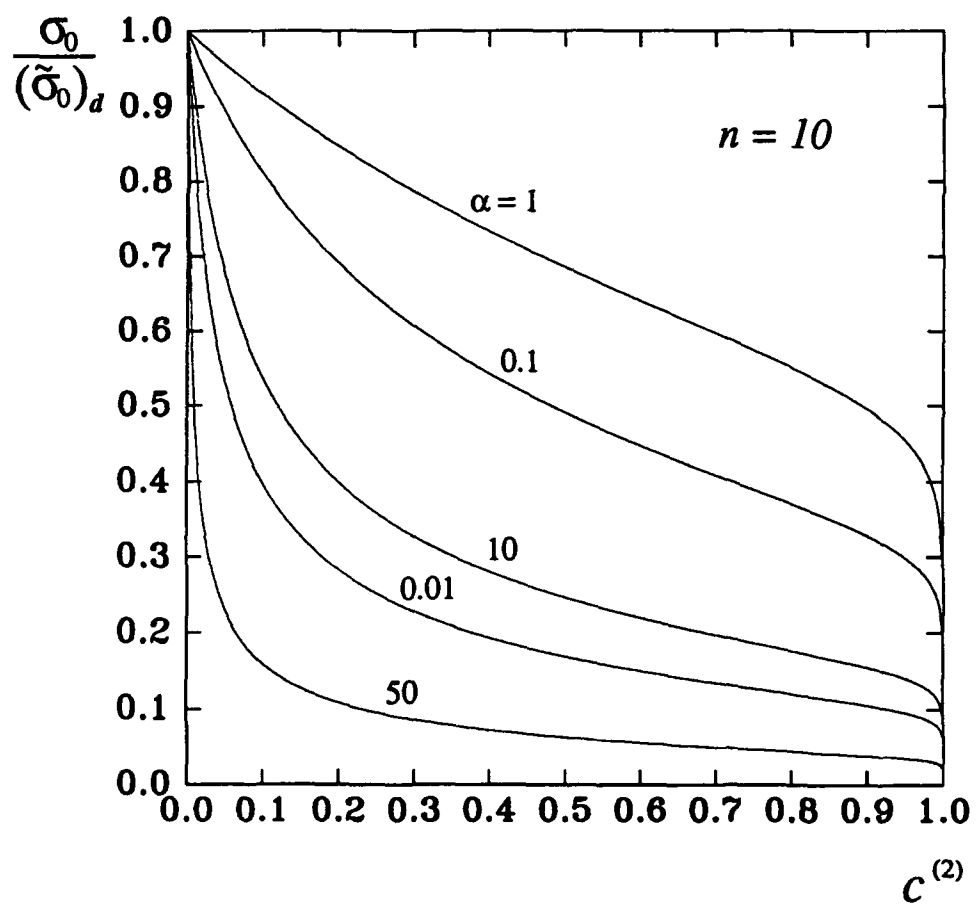


Fig. 2(b)

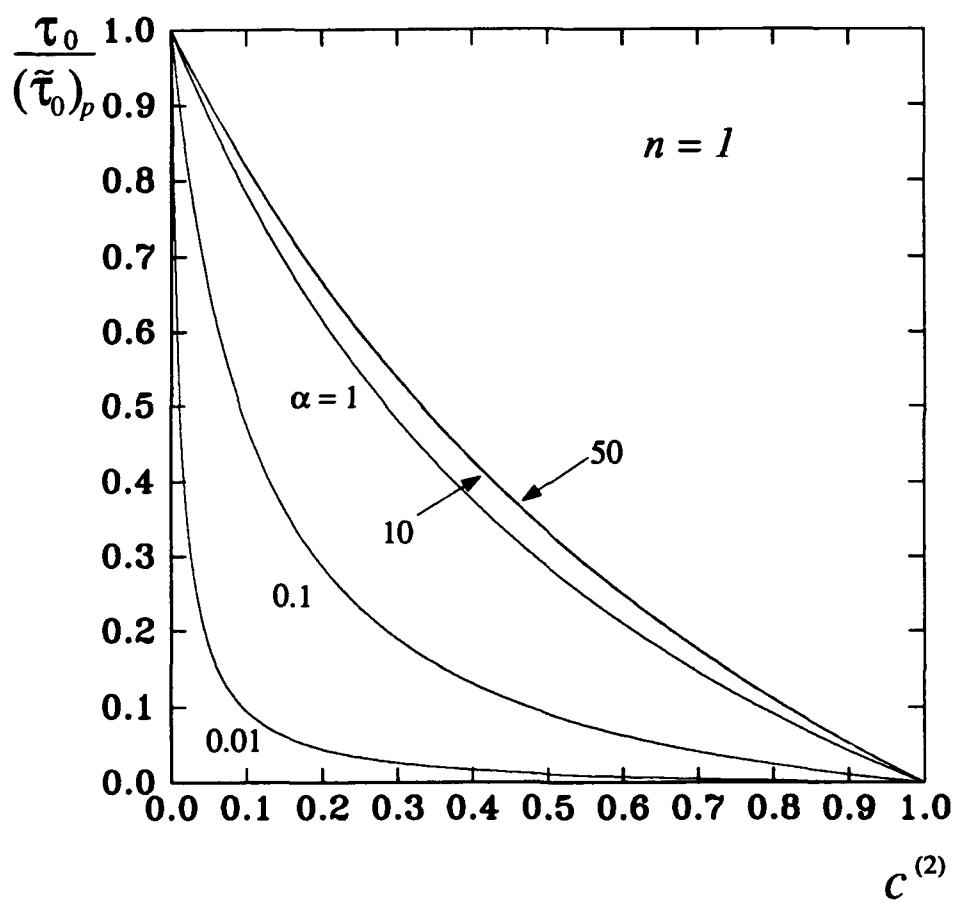


Fig. 3(a)

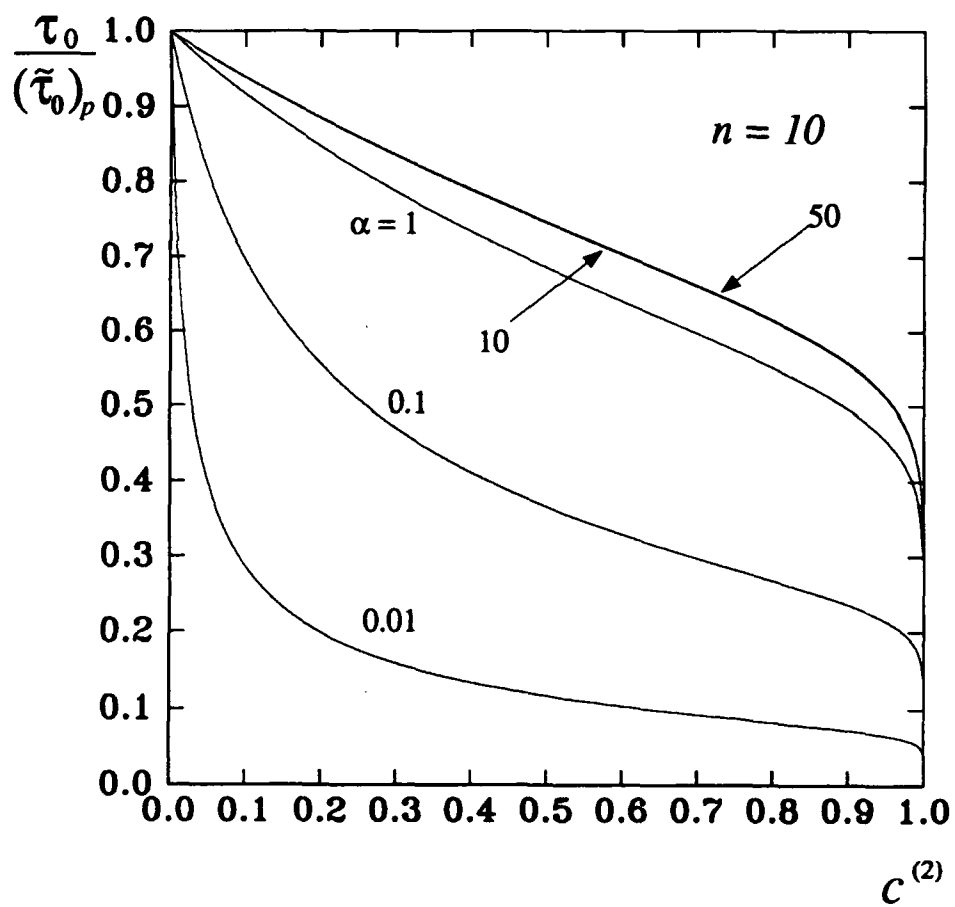


Fig. 3(b)

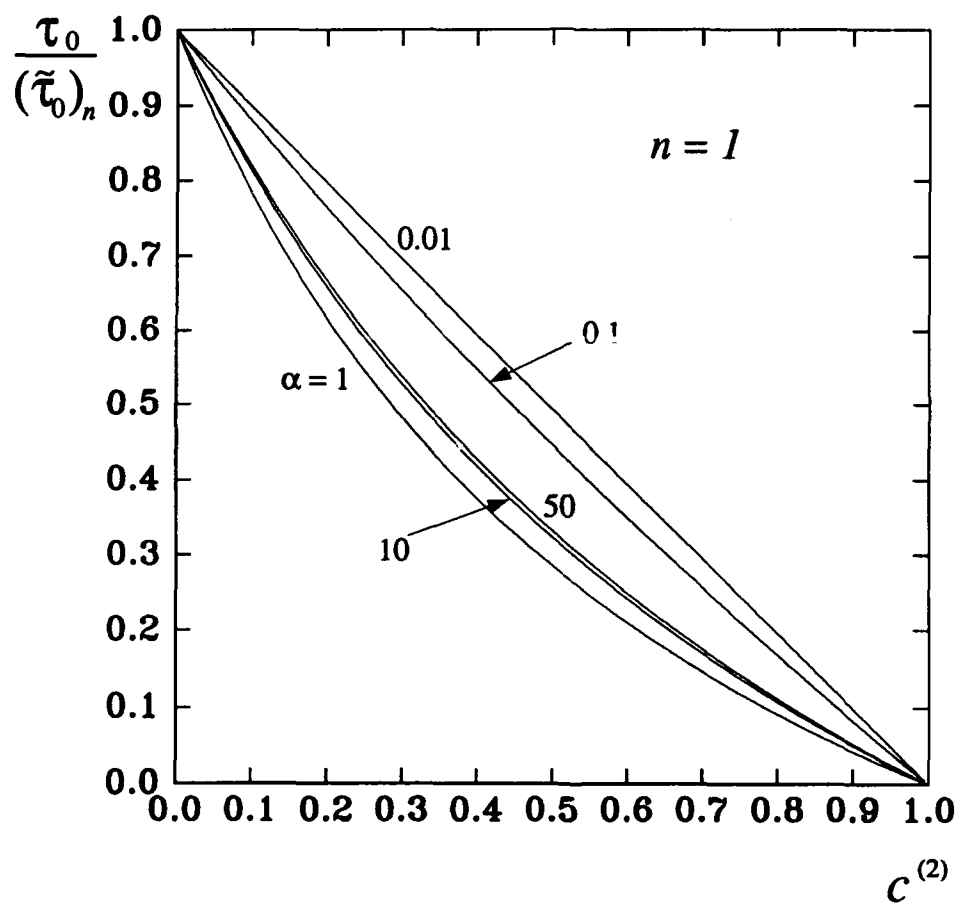


Fig. 4(a)

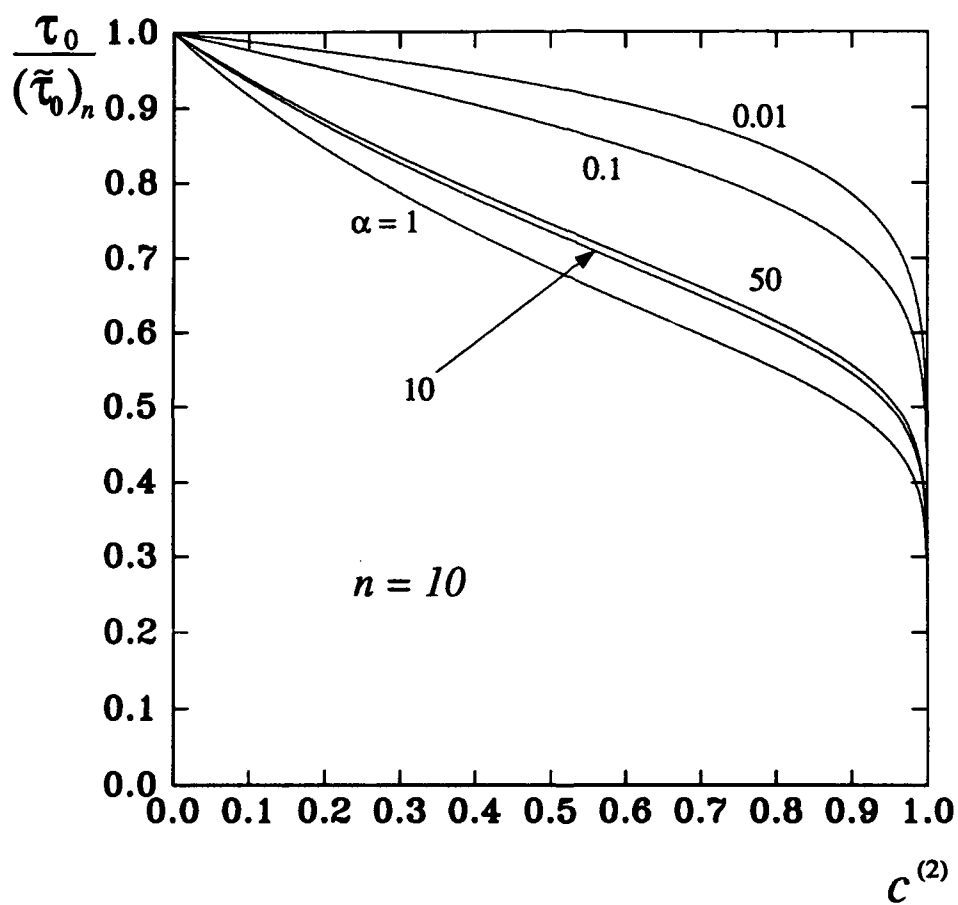


Fig. 4(b)

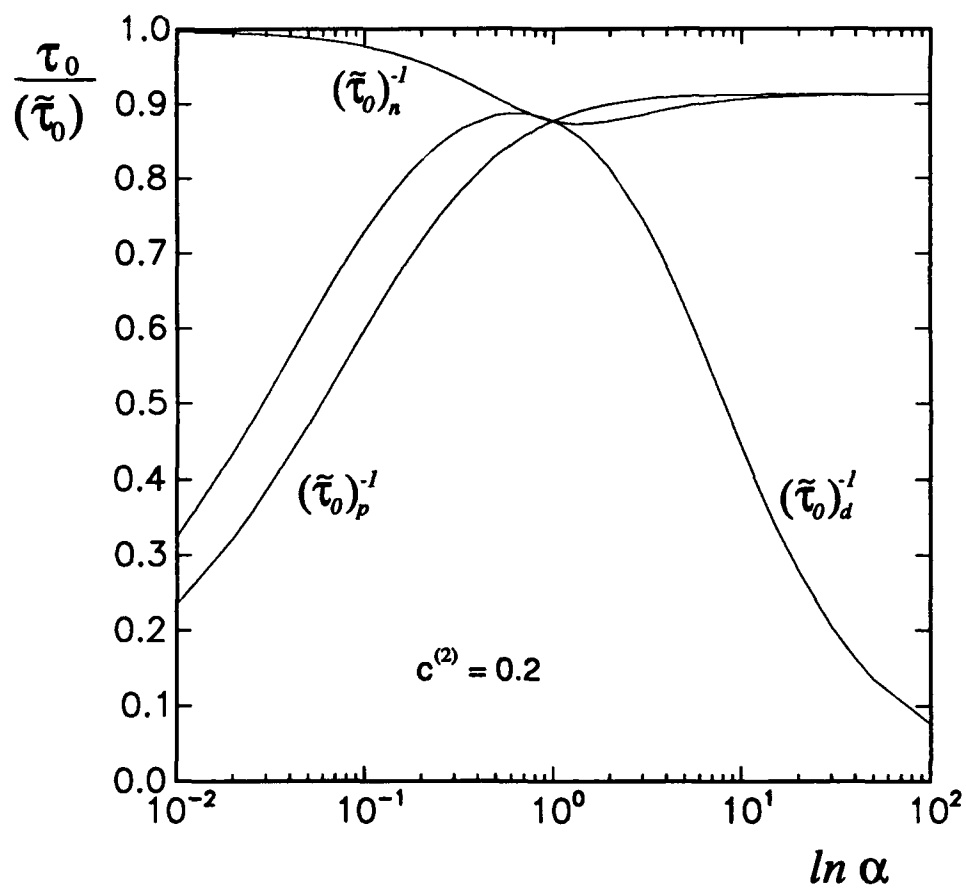


Fig. 5(a)

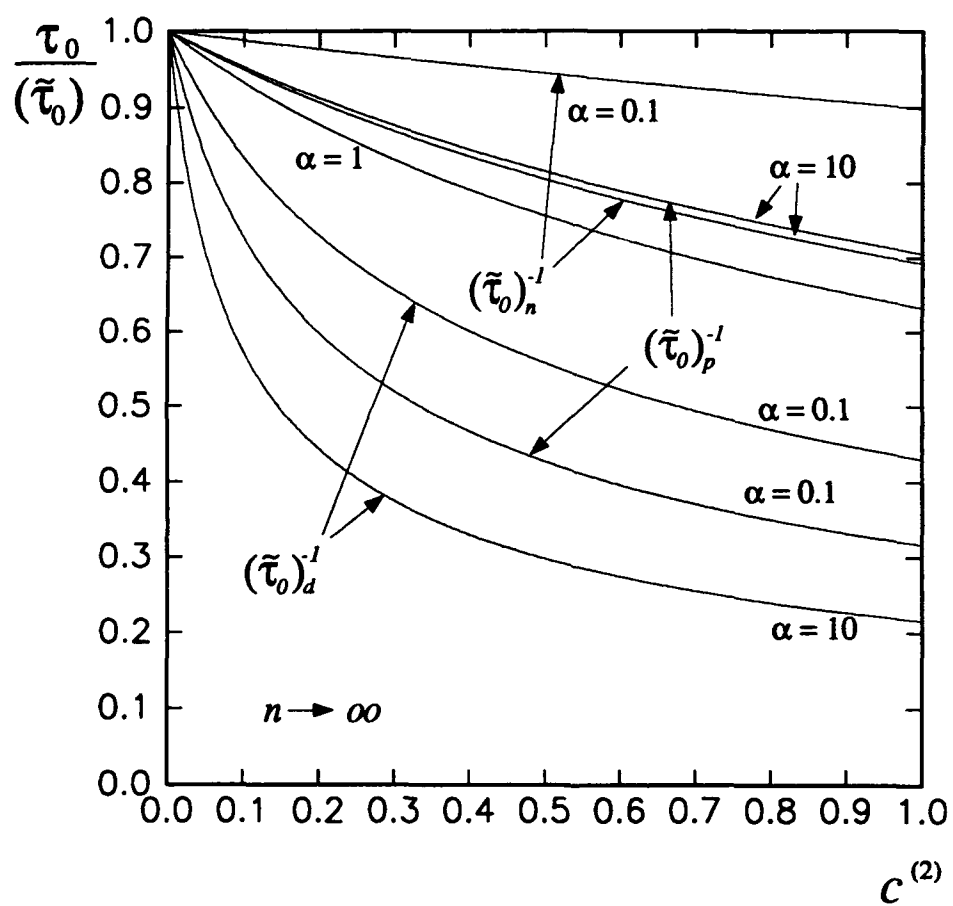


Fig. 5(b)

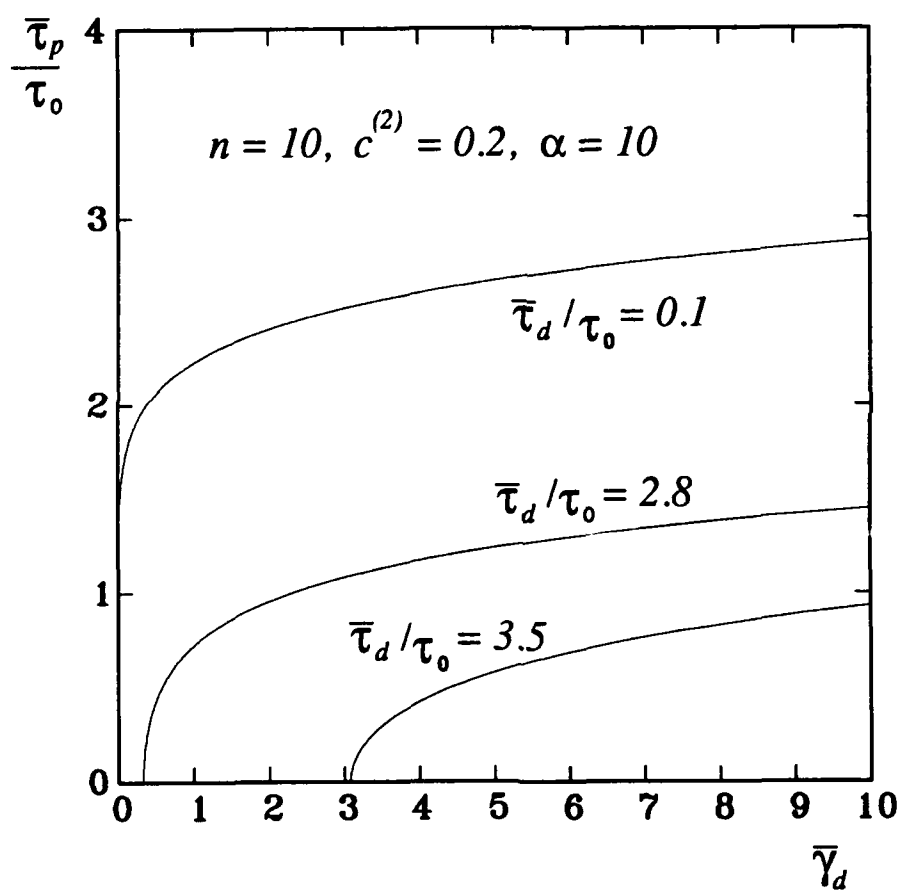


Fig. 6

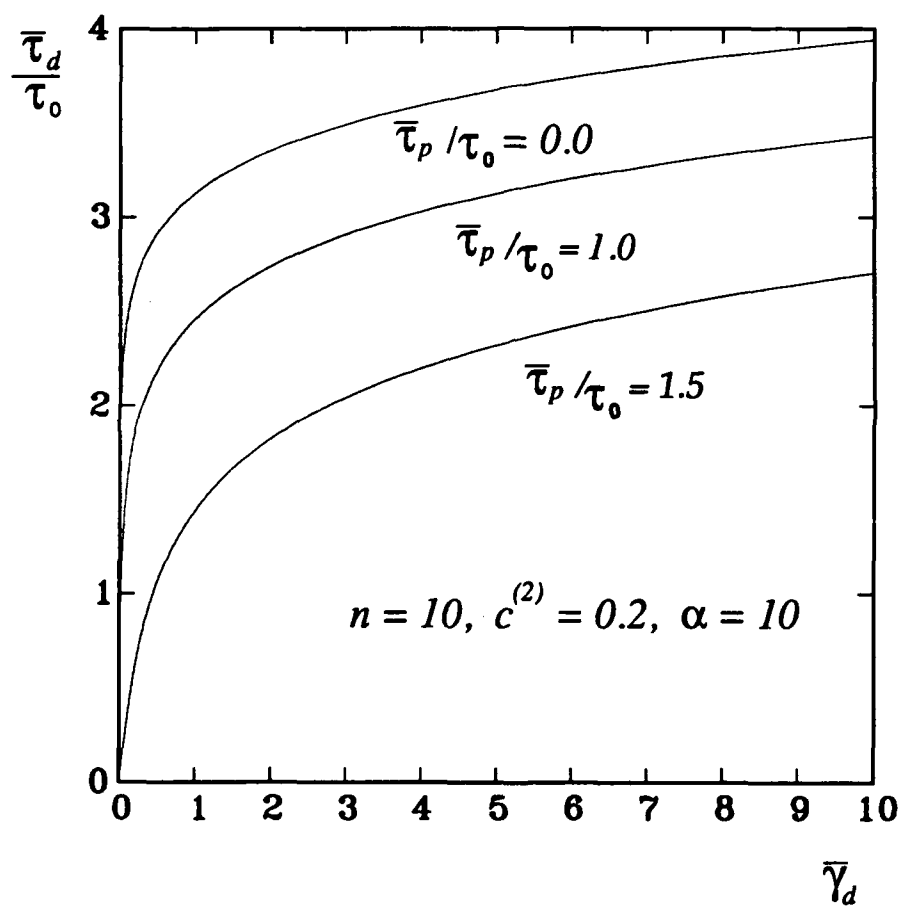


Fig. 7

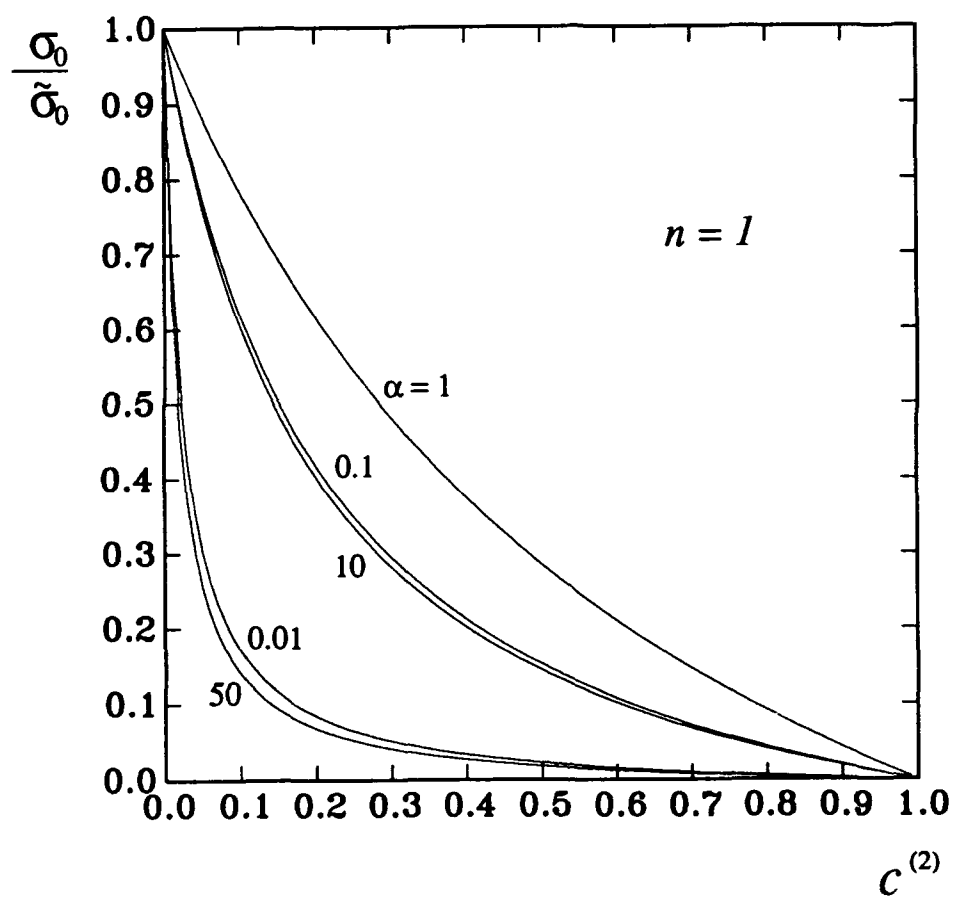


Fig. 8(a)

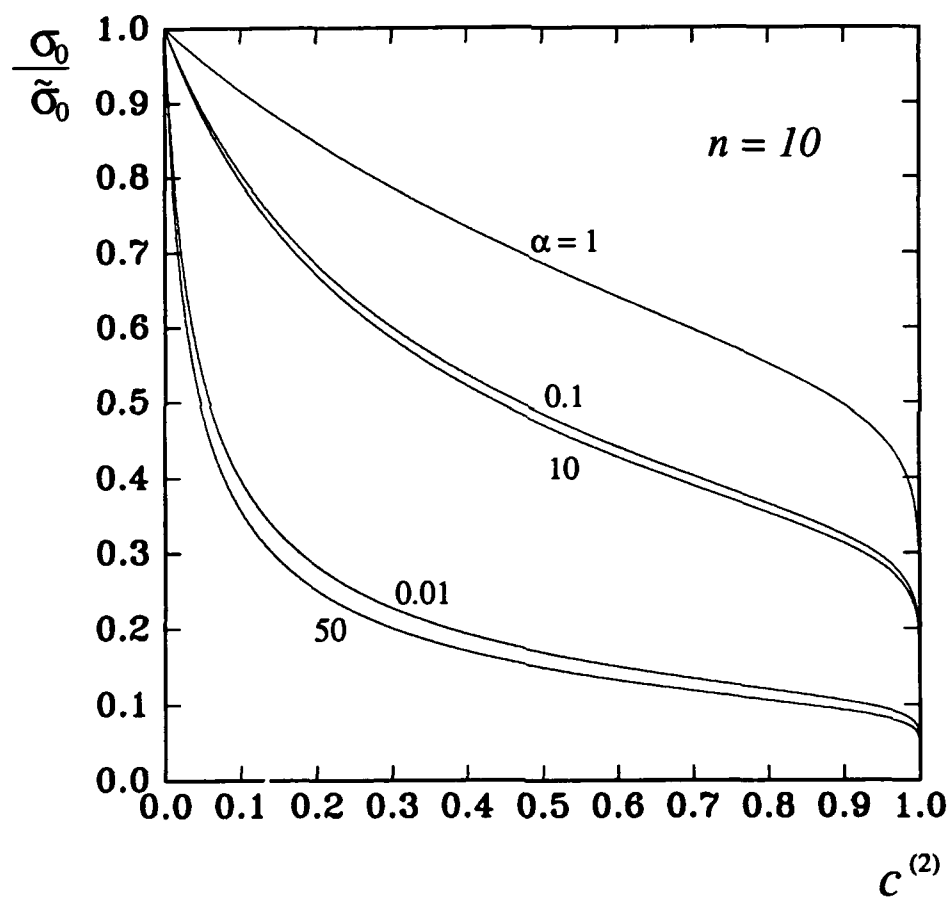


Fig. 8(b)

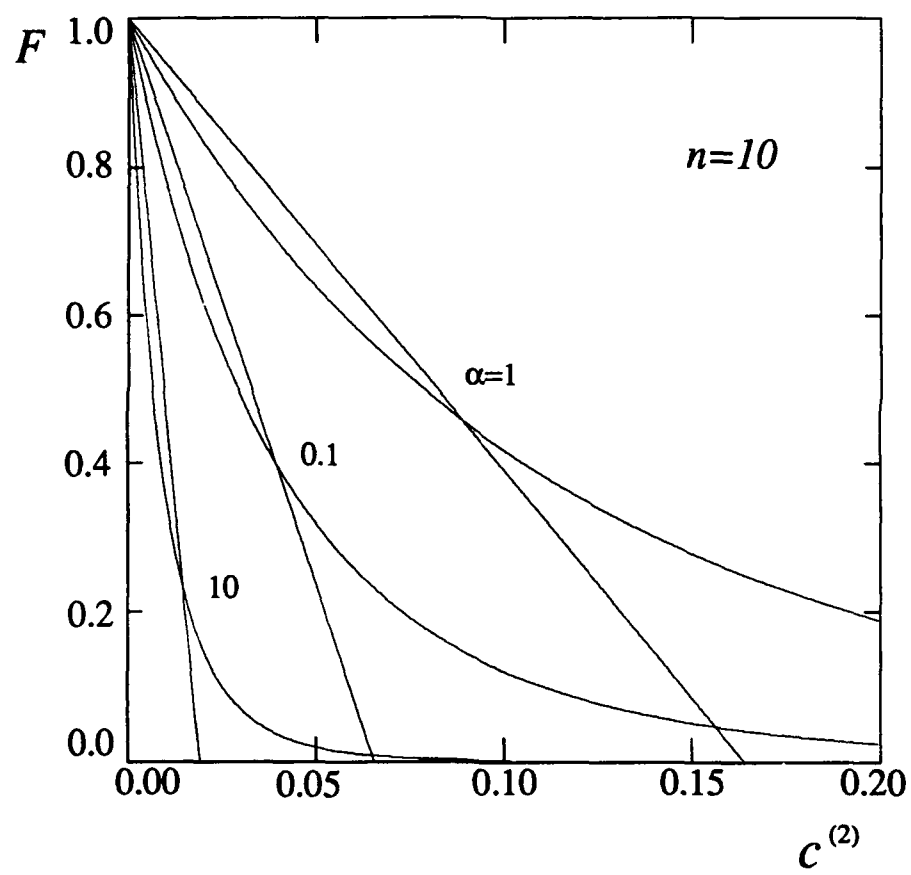


Fig. 9

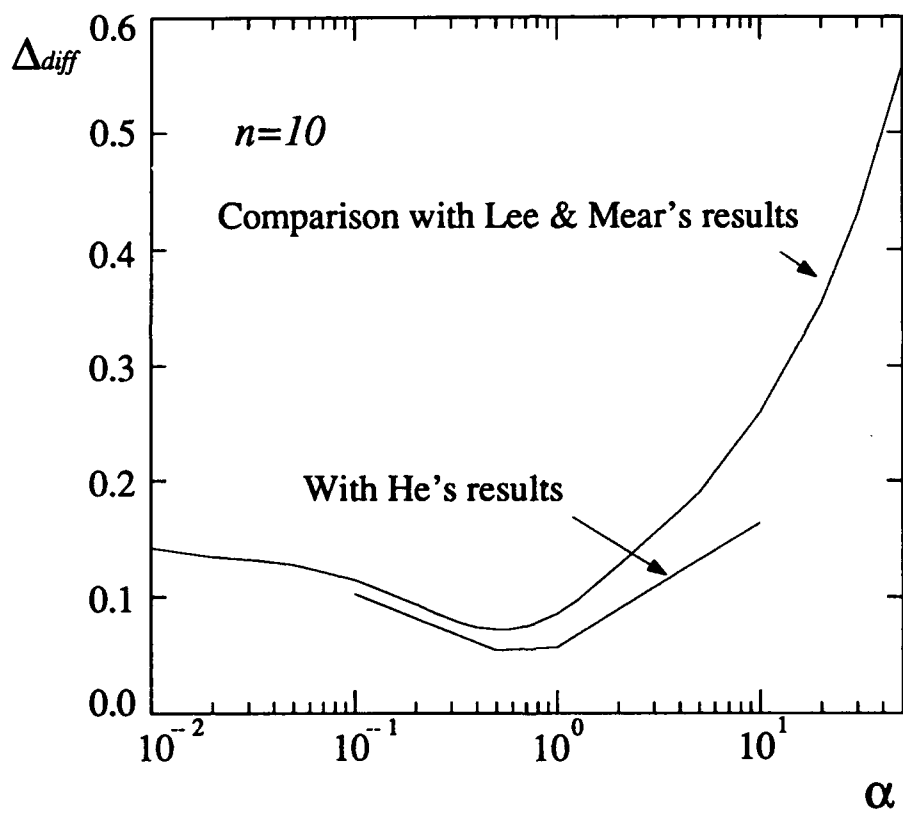


Fig. 10

Reference [25]

On the homogenized yield strength of two-phase composites†

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This paper is concerned with the determination of the effective yield strength of two-phase, rigid-perfectly plastic composite materials. The individual phases are assumed to satisfy, for simplicity, incompressible, isotropic yield criteria of the Mises type. The volume fractions of the constituent phases are prescribed, but their distribution within the composite is otherwise arbitrary. Using the homogenization framework of Suquet (1983) to define the homogenized, or effective, yield strength domain of rigid-perfectly plastic composites, a variational statement is introduced allowing the estimation of the associated effective dissipation functions of plastic composites in terms of the effective dissipation functions of corresponding classes of linearly viscous comparison composites. Thus the variational statement suggests a procedure for generating bounds and estimates for the effective yield strength of rigid-perfectly plastic composites from well-known bounds and estimates for the effective properties of the corresponding linear comparison composites. Sample results are given in the form of upper bounds and lower estimates of the Hashin-Shtrikman type for the effective yield strength of two-phase composites with overall isotropy. Additionally, estimates and bounds are also given for the effective strength domains of two-phase laminated and fibre-reinforced composites, with overall transverse isotropy.

1. Introduction

The problem of determining the effective yield strength of composite materials is one that has captured the interest of numerous investigators over the past 40 years. Among the earliest contributions, we may cite the work of Bishop & Hill (1951) to determine the effective yield strength of polycrystals comprised of plastic single crystals, and the work of Drucker (1959) to compute the limit load of a composite material made up of two rigid-perfectly plastic phases. A general overview of the subject, including the constitutive behaviour of plastic composites before 'extremal' yield, is given by Hill (1967) (see also Suquet 1987). Other contributions specifically addressing the computation of the effective yield strength of composite materials include the papers of Shu & Rosen (1967) and Majumdar & McLaughlin (1975) for the technological important case of fibre-reinforced composites, and the papers of Gurson (1977) and Bao *et al.* (1991) for porous and particulate composites, respectively.

While many of the above-mentioned studies are concerned with composites with completely determinate microstructures, the aim of this paper is different: we seek

† This paper was accepted as a rapid communication

to determine bounds and estimates for the effective yield strength of classes of composites, depending only on partial information about their microstructures, such as the volume fractions of the constituent phases and the overall symmetry of the composites. Thus, after an appropriate definition in §2 of the effective yield strength of rigid-perfectly plastic composites, we propose in §3 a method for bounding and estimating the effective yield strength of plastic composites in terms of the effective properties of appropriately chosen classes of linear comparison composites. The method is applied in §4 to the class of two-phase plastic composites with prescribed volume fractions and overall isotropy to obtain bounds and estimates for their effective yield strengths. These bounds are analogous to the bounds of Hashin & Shtrikman (1963) for the effective moduli of isotropic composites with linear constitutive behaviour of their phases. In §5 explicit expressions are derived for the effective yield strength domains of plastic laminated composites with prescribed volume fractions, thus providing a simple example of the application of the method to anisotropic composites. Finally, in §6 bounds and estimates are also determined for the effective yield strength domain of fibre-reinforced composites with prescribed volume fractions and overall transversely isotropic symmetry.

2. The effective yield strength of two-phase composites

In the development below, we follow the mathematical framework proposed by Suquet (1983) for the definition of the effective, or homogenized, yield strength of heterogeneous materials with periodic microstructure. The materials are assumed, for simplicity, to be made up of two isotropic phases with (non-negative) tensile yield strengths $k^{(1)}$ and $k^{(2)}$ in prescribed fractions $c^{(1)}$ and $c^{(2)}$, such that $c^{(1)} + c^{(2)} = 1$. Owing to the periodicity of their microstructures, the properties of the heterogeneous materials need only be specified over a unit cell of the microstructure Y , in terms of a position vector y defined over Y . Thus the strength domain of the heterogeneous material, which is assumed to be of the Mises type, is given by the set

$$P(y) = \{\sigma(y) \mid \sigma_e \leq k(y), \text{ with } k(y) = k^{(r)} \text{ if } y \in Y^{(r)}, r = 1, 2\}, \quad (1)$$

where $\sigma_e = \sqrt{\frac{1}{2} \sigma' \cdot \sigma'}$ is proportional to the magnitude of the deviatoric part of the stress tensor σ , and $Y^{(1)}$ and $Y^{(2)}$ are complementary subsets of Y ($Y^{(1)} + Y^{(2)} = Y$ and $Y^{(1)} \cap Y^{(2)} = \emptyset$) denoting the regions occupied by phases 1 and 2, respectively, within the unit cell. Below, we make use of the support function of P , denoted by π , and satisfying the relation

$$\pi(y, d) = \sup_{\tau \in P(y)} \{\tau \cdot d\}$$

(see Salençon 1977). The support function π has the physical interpretation of the plastic dissipation function corresponding to the strain rate (rate-of-deformation tensor) d . It then follows from (1) that, for the Mises plastic material,

$$\pi(y, d) = k(y) d_e + \delta_0(d_m), \quad (2)$$

where k is as given in (1), $d_e = \sqrt{\frac{1}{2} d' \cdot d'}$ is proportional to the magnitude of the deviatoric part of d , d_m is the hydrostatic component of d , and $\delta_0 = 0$ if $d_m = 0$, or $\delta_0 = \infty$ otherwise.

The effective yield strength domain of the above composite may be described (Suquet 1983) in terms of the average stress $\Sigma = \langle \sigma \rangle$ (where the angular brackets denote volume averages over Y) by the convex set

$$P^{\text{hom}} = \{\Sigma \mid \exists \sigma(y), \operatorname{div}_y \sigma = 0, \sigma(y) \in P(y), \sigma m \text{ is antiperiodic}, \langle \sigma \rangle = \Sigma\}, \quad (3)$$

where m denotes the normal to the boundary of Y . Dually, we may opt to determine P^{hom} from its support function, or effective dissipation function, given by

$$\pi^{\text{hom}}(D) = \inf_{d \in K} \int_Y \pi(y, d) dy, \quad (4)$$

where $D = \langle d \rangle$ is the average strain-rate tensor and K is the set of kinematically admissible strain rates, defined by

$$K = \{d \mid \exists v, d = \frac{1}{2}(\nabla v + (\nabla v)^T), v = Dy + v', \text{ and } v' \text{ is periodic}\}. \quad (5)$$

We remark that π^{hom} may be an anisotropic function of D . Further, in connection with the determination of P^{hom} from π^{hom} , it is useful to recall (see Salençon 1977) that $P = \operatorname{dom} \{\pi^*\}$, where π^* denotes the convex Legendre-Fenchel polar of π and $\operatorname{dom}(f)$ denotes the effective domain of the function f , i.e. the set $\{x \mid f(x) < \infty\}$ (Van Tiel 1984, §§6.1 and 5.11, respectively).

The usefulness of the above derivations arises from the fact that, at least for the case of no loads prescribed on the boundary, the limit strength of the heterogeneous material with yield domain $P(y)$ may be computed as if the composite was a homogeneous material with yield domain P^{hom} (Suquet 1983). On the other hand, if loads are prescribed on part of the boundary, then the limit load computed directly from P^{hom} yields a strict upper bound for the actual limit load of the composite (Bouchitte & Suquet 1987).

3. Estimates for the effective yield strength

In this section, we seek to develop a method for estimating the effective yield strength domain of the class of composites defined in the previous section. Rather than working with the set P^{hom} , we prefer to work with the corresponding dissipation function π^{hom} . The main result of this section depends on the following lemma.

Lemma. *The local dissipation function of the rigid-perfectly plastic composite, defined by relation (2) in the previous section, admits the representation*

$$\pi(y, d) = \inf_{\mu \geq 0} \{w(y, d) + v(y, \mu)\}, \quad (6)$$

where $w(y, d) = \frac{1}{2} \mu(y) d_e^2 + \delta_0(d_m)$ corresponds to the dissipation function of an incompressible, isotropic, linear comparison composite with viscosity coefficient μ , and where

$$v(y, \mu) = \sup_d \{\pi(y, d) - w(y, d)\}. \quad (7)$$

This result is obtained by defining the function $f: Y \times R^+ \rightarrow R^+$ (R^+ is the set of non-negative reals) by $f(y, u) = k(y) \vee u$, such that $\pi(y, d) = f(y, d_e^2) + \delta_0(d_m)$, and letting $v(y, \mu) = -f_*(y, \frac{1}{2}\mu)$, where f_* denotes the concave Legendre-Fenchel polar of f , namely (Van Tiel 1984, §7.14),

$$f_*(y, p) = \inf_{u \geq 0} \{up - f(y, u)\}. \quad (8)$$

Then, the result (6) follows from the fact that f is concave on R^+ , and therefore $f = f_{**}$ on R^+ . (Since the concave function f_{**} has the geometrical interpretation of the upper concave hull of f , this last result is simply a statement of the concavity of f .)

The next theorem is essentially the homogenized counterpart of relation (6), and thus gives an alternative expression for π^{hom} in terms of the effective dissipation

function of a linear heterogeneous comparison material. The idea is to be able to make use of well-established results for the effective behaviour of linear composites to generate corresponding results for the nonlinear rigid-perfectly plastic composites.

Theorem. *The effective dissipation function of the rigid-perfectly plastic composite admits the variational representation*

$$\pi^{\text{hom}}(\mathbf{D}) = \inf_{\mu^{(1)} \geq 0} \left\{ w^{\text{hom}}(\mathbf{D}) + \int_Y v(\mathbf{y}; \mu) dy \right\}, \quad (9)$$

where w^{hom} denotes the effective dissipation function of the incompressible, isotropic, linear comparison composite of the previous lemma, namely,

$$w^{\text{hom}}(\mathbf{D}) = \inf_{d \in K} \int_Y u(\mathbf{y}; d) dy. \quad (10)$$

This result is an adaptation of corresponding results by Ponte Castañeda (1992) for plastic solids with non-vanishing hardening. Thus the demonstration of relation (9) is formally the same as the demonstration of the corresponding result (relation (3.21) in the above reference) for hardening plastic solids, and it will not be repeated here. However, the essence of the argument of the proof is to substitute expression (6) for π into expression (4) for π^{hom} , and to interchange the order in which the resulting infima are evaluated. This leads to expression (9), together with (10).

We remark that the above variational statement for π^{hom} involves the evaluation of an infimum problem over the set of non-negative functions μ in some appropriate functional space, in addition to the determination of the effective behaviour of incompressible, isotropic linear composites with arbitrary periodic microstructures (as prescribed by the distribution of viscosity coefficients μ). This is, of course, at least as difficult as the direct approach based on the original definition (4) of π^{hom} ; however, the variational statement (9) lends itself to the following useful approximation.

Corollary. *The effective dissipation function of the two-phase, rigid-perfectly plastic composite may be estimated from the inequality*

$$\pi^{\text{hom}}(\mathbf{D}) \leq \inf_{\mu^{(1)}, \mu^{(2)} \geq 0} \left\{ w^{\text{hom}}(\mathbf{D}) + \sum_{r=1}^2 c^{(r)} v^{(r)}(\mu^{(r)}) \right\}, \quad (11)$$

where w^{hom} now corresponds to the effective dissipation function of an incompressible, isotropic, linear comparison composite with the same distribution of the two phases as the rigid-perfectly plastic composite. The two linear comparison phases have viscosity coefficients $\mu^{(1)}$ and $\mu^{(2)}$, and are prescribed in volume fractions $c^{(1)}$ and $c^{(2)}$, respectively. Further, the functions $v^{(r)}$ are determined from expression (7), specialized to each phase, and are hence independent of $\mathbf{y}$.

This result is obtained from relation (9) in the previous theorem by restricting the original set of (arbitrary) non-negative, comparison viscosity coefficients to the strictly smaller set of piecewise constant, non-negative viscosity coefficients, with a different constant in each of the two phases of the original nonlinear composite.

Application of result (11) may be considered for anisotropic composites, such as laminated composites and fibre-reinforced composites, as well as for isotropic composites. In the next section, we make use of the Hashin-Shtrikman bounds for two-phase, isotropic, linear composites with prescribed volume fractions to generate

corresponding results for the class of two-phase, isotropic perfectly plastic composites with prescribed volume fractions. In later sections, we consider laminated and fibre-reinforced composites.

4. Application to two-phase, isotropic composites

Under the assumption of overall isotropy, the problem of characterizing the effective yield strength of a rigid-perfectly plastic composite reduces to that of determining its effective uniaxial yield strength k^{hom} , such that $\pi^{\text{hom}}(\mathbf{D}) = k^{\text{hom}} D_e + \delta_0(D_m)$. In this section, we apply inequality (11) to the class of two-phase, isotropic plastic composites with prescribed volume fractions to obtain an upper bound for k^{hom} . Thus we require an upper bound for the effective dissipation function of the corresponding class of isotropic, incompressible linearly viscous composites. (We emphasize that the class of 'linearly isotropic' microstructures is larger than the class of 'nonlinearly isotropic' microstructures, and therefore a bound for the effective properties of linear composites with linearly isotropic microstructures is also a bound for the class of linear composites with nonlinearly isotropic microstructures.) Such a bound for the linear comparison composites may be expressed as an upper bound on the effective viscosity coefficient μ^{hom} , such that $w^{\text{hom}}(\mathbf{D}) = \frac{3}{2} \mu^{\text{hom}} D_e^2 + \delta_0(D_m)$. Because of the mathematical analogy between linearly elastic and linearly viscous behaviour, an upper bound for the effective viscosity coefficient may be obtained from the work of Hashin & Shtrikman (1963). This bound is known to be optimal (Francfort & Murat 1986) and may be expressed (Ponte Castañeda 1992) in the form of the one-dimensional optimization problem:

$$\mu^{\text{hom}} \leq \inf_{\omega} \left\{ c^{(1)} \mu^{(1)} (1 + c^{(2)} \omega)^2 + c^{(2)} \mu^{(2)} (1 - c^{(1)} \omega)^2 + \frac{3}{2} c^{(1)} c^{(2)} \mu^{(2)} \omega^2 \right\}, \quad (12)$$

where we have assumed that $\mu^{(1)} \leq \mu^{(2)}$. Note that, unlike the original expression for the Hashin-Shtrikman upper bound, this expression is linear in the viscosity coefficients, which will greatly simplify the derivation of the results that follow.

Therefore, assuming that $k^{(1)} \leq k^{(2)}$, we find that π^{hom} must satisfy the inequality

$$\begin{aligned} \pi^{\text{hom}}(\mathbf{D}) \leq & \inf_{\omega} \left\{ c^{(1)} \inf_{\mu^{(1)} \geq 0} \left[\frac{3}{2} \mu^{(1)} (1 + c^{(2)} \omega)^2 D_e^2 + c^{(1)} \mu^{(1)} \right] + \dots \right. \\ & \left. + c^{(2)} \inf_{\mu^{(2)} \geq 0} \left[\frac{3}{2} \mu^{(2)} ((1 - c^{(1)} \omega)^2 + \frac{3}{2} c^{(1)} \omega^2) D_e^2 + c^{(2)} \mu^{(2)} \right] + \delta_0(D_m) \right\}, \end{aligned} \quad (13)$$

where we have made use of (11), together with (12), and additionally we have interchanged the order of the infima and rearranged terms. By virtue of the local relation (6), specialized to each phase, we obtain the result that $\pi^{\text{hom}}(\mathbf{D}) \leq k_{\text{HS}}^{\text{hom}} D_e + \delta_0(D_m)$, where

$$k_{\text{HS}}^{\text{hom}} = \inf_{\omega} \left\{ c^{(1)} k^{(1)} [1 + c^{(2)} \omega] + c^{(2)} k^{(2)} [(1 - c^{(1)} \omega)^2 + \frac{3}{2} c^{(1)} \omega^2] \right\}. \quad (14)$$

This expression, which is analogous to the corresponding expression for the effective viscosity coefficient (12), and provides an upper bound for the effective yield strength of the class of two-phase, isotropic, rigid-perfectly plastic composites, may be simplified further to obtain the result

$$\frac{k_{\text{HS}}^{\text{hom}}}{k^{(2)}} = \frac{5c^{(1)}}{3 + 2c^{(1)}} \frac{k^{(1)}}{k^{(2)}} + \frac{3c^{(2)}}{3 + 2c^{(1)}} \sqrt{\left\{ 1 + \frac{2c^{(1)}}{3} \left[1 - \left(\frac{k^{(1)}}{k^{(2)}} \right)^2 \right] \right\}}. \quad (15)$$

This final expression for the Hashin-Shtrikman upper bound on k^{hom} is analogous to the corresponding bound for the effective viscosity coefficient of two-phase linear composites, and may additionally be thought of as an estimate for the effective yield strength of isotropic particulate composites, with the stronger phase playing the role of the matrix, and the weaker phase that of the inclusion (see Francfort & Murat 1986).

On the other hand, whereas it is well known that lower bounds also exist for the effective viscosity coefficients of linear viscous composites, we are unable here to determine non-trivial lower bounds for the effective yield strength of rigid-perfectly plastic composites; the reason being that lower bounds for the effective dissipation function of two-phase, linear comparison composites do not necessarily translate into corresponding lower bounds for the effective dissipation function of two-phase, rigid-perfectly plastic composites (this is a consequence of the inequality in (11)). However, an alternative point of view is to interpret the Hashin-Shtrikman lower bounds for the effective viscosity of two-phase, linear composites as estimates for the effective viscosity coefficient of particulate composites with the weaker phase playing the role of the matrix (see Francfort & Murat 1986). Then, application of relation (11) to the rigid-perfectly plastic composite leads to estimates for the effective yield strength of particulate composites with the weaker phase serving the role of the matrix. The procedure is formally identical to the procedure followed for the upper bound, starting with relation (12), except that $\mu^{(2)}$ is replaced by $\mu^{(1)}$ in the last term on the right-hand side. The final result for such a 'lower estimate' (not a bound) for k^{hom} may be expressed in the form

$$\frac{k_{\text{HS}+}^{\text{hom}}}{k^{(1)}} = \begin{cases} \frac{5c^{(2)} k^{(2)} + 3c^{(1)}}{3 + 2c^{(2)} k^{(1)} + 3 + 2c^{(2)}} \sqrt{\left\{1 + \frac{2c^{(2)}}{3} \left[1 - \left(\frac{k^{(2)}}{k^{(1)}}\right)^2\right]\right\}} & \text{if } \frac{2}{3} \sqrt{1 + \frac{2}{3} c^{(2)}} < k^{(1)}/k^{(2)} \leq 1, \\ \sqrt{1 + \frac{2}{3} c^{(2)}} & \text{if } k^{(1)}/k^{(2)} \leq \frac{2}{3} \sqrt{1 + \frac{2}{3} c^{(2)}}. \end{cases} \quad (16)$$

The above results for the Hashin-Shtrikman upper bound and lower estimate for k^{hom} , appropriately normalized by the yield strength of the stronger phase, are plotted in figure 1 as functions of the strength ratio of the two phases, for equal proportions of the two phases. In addition, we also show in this figure the upper bound of Bishop & Hill (1951) for k^{hom} , obtained by making use of a uniform strain-rate trial field in the minimum dissipation principle (4), and given by $k_+^{\text{hom}} = c^{(1)} k^{(1)} + c^{(2)} k^{(2)}$. Similarly, we also show in the figure a lower bound for k^{hom} , obtained trivially from definition (3), and given by $k_-^{\text{hom}} = k^{(1)}$. Thus in figure 1 we observe that the new Hashin-Shtrikman upper bound is tighter than the classical Bishop-Hill upper bound, although the difference between the two bounds is relatively small. On the other hand, the Hashin-Shtrikman lower estimate is significantly stronger than the trivial lower bound; in fact, it is very close to the upper bounds when the strength ratio is close to 1, but for strength ratios less than $\frac{2}{3} \sqrt{1 + \frac{2}{3} c^{(2)}}$, the Hashin-Shtrikman lower estimate becomes insensitive to the yield strength of the stronger (in this case the inclusion) phase, and quickly diverges from the upper bounds.

Interestingly, the above observations are very similar to those made by Bao *et al.* (1991) via a numerical implementation of the 'generalized self-consistent' model. We note, however, that their predictions for k^{hom} lie within our Hashin-Shtrikman estimates. This is best observed by comparing our figure 2, depicting the effect of

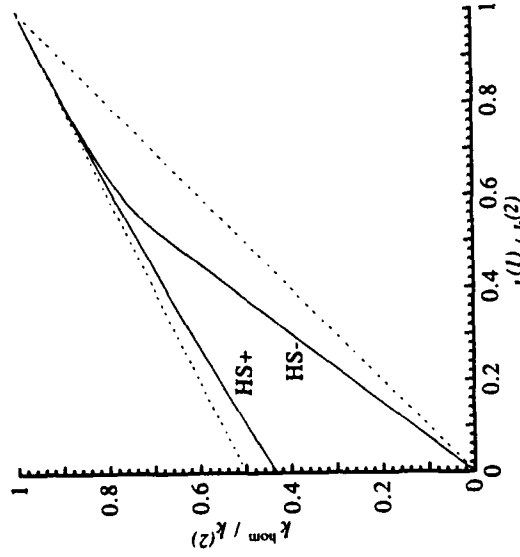


Figure 1. Bounds and estimates for k^{hom} for isotropic composites as functions of the strength ratio $k^{(1)}/k^{(2)}$, for a fixed volume fraction $c^{(2)} = 0.5$. The top and bottom continuous curves correspond to the Hashin-Shtrikman upper bound and lower estimate, respectively; the top and bottom dashed curves to the Bishop-Hill upper bound and trivial lower bound, respectively.

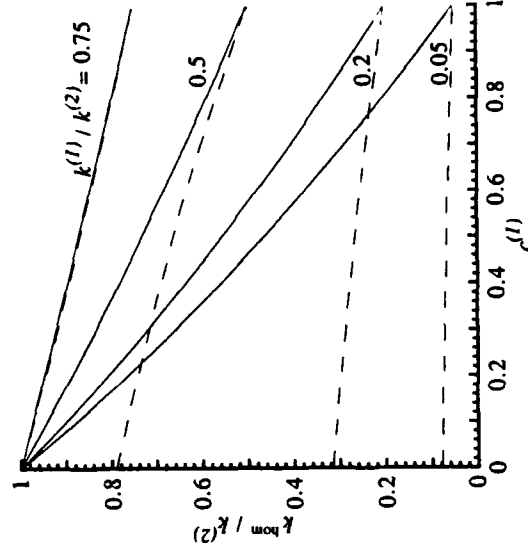


Figure 2. Hashin-Shtrikman upper bounds (continuous lines) and lower estimates (dashed lines) for k^{hom} for isotropic composites as functions of the volume fraction $c^{(1)}$, for four different values of the strength ratio $k^{(1)}/k^{(2)}$.

changes in the proportions of the two phases on the Hashin-Shtrikman bounds and estimates, with the appropriate range of figs 4 and 6 in Bao *et al.* (1991), showing results for composites with alternative choices of the stronger and weaker phase for

the inclusion and matrix, respectively. The difference between the two sets of predictions is accentuated for the lower estimates as the volume fraction of the weaker matrix phase becomes smaller (the lower self-consistent estimates approach 1 as $c^{(1)}$ approaches 0, whereas the lower Hashin-Shtrikman estimates approach numbers less than 1). This may be understood in terms of the observation that for the self-consistent model must of the 'matrix' phase associated with the lower estimate is made up of the 'homogenized' material (there is only a thin shell of the weaker material enclosed by a larger shell of the homogenized material), which is stronger than the weaker phase; whereas for the Hashin-Shtrikman configuration (which has a particulate microstructure with a well-defined matrix phase) corresponding to the lower estimate, the matrix phase is made up entirely of the weaker material.

5. Application to two-phase, laminated composites

We mentioned earlier that even if the phases are isotropic, the microstructure of the composite may be such that the composite is itself anisotropic. In this section, we consider the simple case of a laminated material made up of alternating layers of phases 1 and 2 with tensile yield strengths $k^{(1)}$ and $k^{(2)}$, as given by (1), in prescribed volume fractions $c^{(1)}$ and $c^{(2)}$, respectively. The orientation of the laminated composite is characterized by the unit normal to the layers $\mathbf{n}$. With these hypotheses, we observe that the composite will have transversely isotropic symmetry.

To make effective use of the theorem of §3, it is important to note that, under the assumption that the thickness of the layers is small compared with the overall size of the laminate, the strain-rate fields within the layers of the laminate are constant (a different constant in each phase). Then, it is easy to demonstrate that the comparison viscosity coefficients in (9) are also constant within each phase. It follows that the dissipation function of the laminate may be determined exactly from

$$\pi^{\text{hom}}(\mathbf{D}) = \inf_{\mu^{(1)}, \mu^{(2)} \geq 0} \left\{ w^{\text{hom}}(\mathbf{D}) + \sum_{r=1}^2 c^{(r)} \psi^{(r)}(\mu^{(r)}) \right\}, \quad (17)$$

where w^{hom} now corresponds to the effective dissipation function of a linearly viscous laminated composite with two phases with viscosity coefficients $\mu^{(1)}$ and $\mu^{(2)}$ distributed in precisely the same way as the phases in the corresponding perfectly plastic laminate. Thus, to determine π^{hom} we require an estimate for w^{hom} . Such a result may be extracted, for example, from the work of Walpole (1969), and may be expressed in the form

$$w^{\text{hom}}(\mathbf{D}) = \frac{2}{3} \mu_a (D_a^2 + D_p^2) + \frac{2}{3} \mu_n D_n^2 + \delta_0 (D_m), \quad (18)$$

where $\mu_a = c^{(1)} \mu^{(1)} + c^{(2)} \mu^{(2)}$ and

$$\mu_n = \inf_{\omega} \{ c^{(1)} \mu^{(1)} (1 + c^{(2)} \omega)^2 + c^{(2)} \mu^{(2)} (1 - c^{(1)} \omega)^2 \}$$

are the arithmetic and harmonic means of the viscosity coefficients, respectively. We note that the above form of μ_n is selected to facilitate the computations to follow. In relation (18),

$$D_a = \frac{1}{3} \text{tr}(\mathbf{D}) - \mathbf{n} \cdot (\mathbf{D} \mathbf{n}), \quad D_n = \frac{2}{3} \sqrt{((\mathbf{D} \mathbf{n}) \cdot (\mathbf{D} \mathbf{n}) - [\mathbf{n} \cdot (\mathbf{D} \mathbf{n})]^2)}, \quad \text{and} \quad D_p = \sqrt{(D_a^2 - D_n^2)}$$

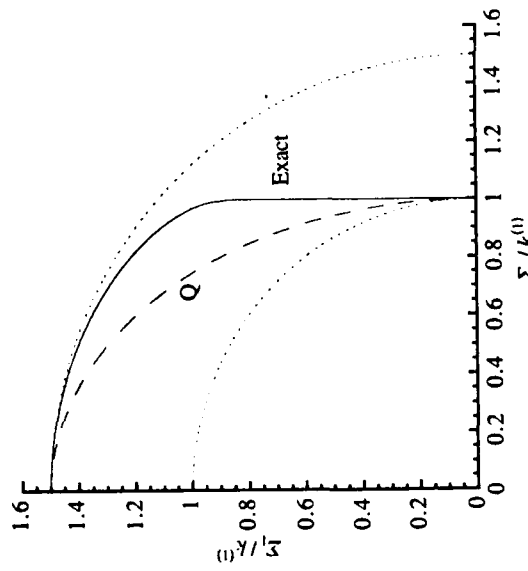


Figure 3. Estimates for the yield surface of a laminated composite with $k^{(2)}/k^{(1)} = 2$ and $c^{(2)} = 0.5$, plotted as functions of Σ_n and Σ_a . The continuous curve corresponds to the exact yield surface; the long-dash curve to the quadratic interpolation of Hill; and the top and bottom short-dash curves to the Bishop-Hill upper bound and the trivial lower bound, respectively.

(where D_p has been defined previously) are the incompressible transversely isotropic invariants of $\mathbf{D}$, corresponding physically to axisymmetric loading (aligned with $\mathbf{n}$), shear parallel to the layers of the composite, and shear transverse to the layers. We also define, for convenience, $D_1 = \sqrt{(D_a^2 + D_p^2)}$.

Then, following a procedure similar to that followed for the two-phase, isotropic plastic composite, we arrive at the expression

$$\pi^{\text{hom}}(\mathbf{D}) = \inf_{\omega} \{ c^{(1)} k^{(1)} \sqrt{[D_1^2 + (1 + c^{(2)} \omega)^2 D_n^2] + c^{(2)} k^{(2)} \sqrt{[D_1^2 + (1 - c^{(1)} \omega)^2 D_n^2]} + \delta_0 (D_m)} \}. \quad (19)$$

From this result, we may compute, by the procedure indicated in §2 ($P = \text{dom} \{\pi^*\}$), the effective yield strength domain of the laminate. The final result is

$$P^{\text{hom}} = \{ \Sigma_n \mid \Sigma_n \leq k^{(1)}, \Sigma_1 \leq c^{(1)} \sqrt{[(k^{(1)})^2 - \Sigma_n^2] + c^{(2)} \sqrt{[(k^{(2)})^2 - \Sigma_n^2]} \}, \quad (20)$$

where $\Sigma_n = \sqrt{[3] \{ (\Sigma \mathbf{n}) \cdot (\Sigma \mathbf{n}) - [\mathbf{n} \cdot (\Sigma \mathbf{n})]^2 \}}$ and $\Sigma_1 = \sqrt{(\Sigma_p^2 + \Sigma_a^2)}$ (with $\Sigma_p = \sqrt{(\Sigma_a^2 - \Sigma_n^2) - \Sigma_a^2}$ and $\Sigma_a = \frac{1}{3} \{ \text{tr}(\Sigma) - 3 \mathbf{n} \cdot (\Sigma \mathbf{n}) \}$). We note, in particular, that $\Sigma_n \leq k^{(1)}$ when $\Sigma_1 = 0$, and $\Sigma_1 \leq c^{(1)} k^{(1)} + c^{(2)} k^{(2)}$ when $\Sigma_n = 0$.

The yield surface associated with P^{hom} in (20) is plotted in figure 3 in terms of Σ_n and Σ_1 , for a strength ratio of two, and equal volume fractions of the two phases. The associated isotropic Bishop-Hill upper bound and trivial lower bound are also included in this figure for comparison with the exact estimate. They appear in the figure as quarter circles of radii $k_+^{\text{hom}} = c^{(1)} k^{(1)} + c^{(2)} k^{(2)}$, and $k_-^{\text{hom}} = k^{(1)}$, respectively. It may be observed that the Bishop-Hill upper bound is a good estimate for the effective yield surface of the laminated composite as long as $\Sigma_1/\Sigma_n > c^{(2)} \sqrt{[(k^{(2)}/k^{(1)})^2 - 1]}$ (approximately). However, for $\Sigma_1/\Sigma_n < c^{(2)} \sqrt{[(k^{(2)}/k^{(1)})^2 - 1]}$,

neither classical estimate is very good, except when Σ_f/Σ_n is close to zero when the trivial lower bound and the exact yield surface are in close agreement. We also show in figure 3 a quadratic interpolation between the Bishop-Hill upper bound and the trivial lower bound. We observe that this type of estimate, originally proposed by Hill (1948) for slightly anisotropic materials, is not accurate for the strongly anisotropic laminated composite shown in the figure. This is seen to be a consequence of the flat portion of the yield surface of the laminated composite ($\Sigma_n = k^{(1)}$). Further, the existence of the two distinct 'branches' in the yield surface defined by (20) is consistent with the general comments of Hashin (1980) for the analogous case of fibre-reinforced composites, and they correspond to two distinct 'failure' mechanisms. Thus the flat section of the yield surface, corresponding to shear parallel to the layers of the laminate (Σ_n), is controlled by the weaker phase (1), whereas the other 'mode' (a combination of the axisymmetric and transverse shear modes, Σ_d and Σ_p , respectively) depends on both the weaker and the stronger phase.

We note that homogenization results for the yield strength of rigid-perfectly plastic laminated composites have been obtained earlier by de Buhan (1983). However, to the knowledge of the authors, the explicit form (20) for the yield surface of the laminated composite is new. Also, a more comprehensive study of laminated materials including the effect of hardening and compressibility is given by deBotton & Ponte Castañeda (1992).

6. Application to two-phase, fibre-reinforced composites

In this section, we study the technologically important case of fibre-reinforced plastic composites made up of two phases with tensile yield strengths $k^{(1)}$ and $k^{(2)}$, as given by (1), in prescribed volume fractions $c^{(1)}$ and $c^{(2)}$, respectively. The cylindrical fibres, which may be made up of either phase, are assumed to have the same orientation, characterized by a unit normal n , aligned with the fibres. If we further assume that the distribution of the fibres in the transverse plane is random, then the fibre-reinforced composites exhibit transversely isotropic symmetry, in similarity with the laminated composites. However, the fibre-reinforced composites are unlike the laminated composites in that the above hypotheses on the microstructure do not suffice to completely specify the effective yield strength of the composites. In this sense, the fibre-reinforced composites are more like the isotropic composites discussed in §4. Thus we are concerned in this section with determining bounds and estimates for the effective yield strength domain of the class of two-phase, fibre-reinforced composites.

We follow a procedure that is an appropriate generalization of the procedure followed in §4 for the isotropic composites. Thus we begin with the application of inequality (11) to the class of fibre-reinforced plastic composites, and we require an upper bound for the effective dissipation function of the class of two-phase, incompressible, linearly viscous fibre composites with transversely isotropic symmetry. Such a bound follows from the work of Hill (1964) and Hashin (1965), and may be expressed in the form

$$w^{\text{hom}}(D) = \frac{3}{2} \mu_{\text{HS}+}^{\text{hom}} (D_n^2 + D_p^2) + \frac{3}{2} \mu_{\text{HS}} D_d^2 + \delta_0(D_m), \quad (21)$$

$$\text{where } \mu_{\text{HS}+}^{\text{hom}} = \inf \{ c^{(1)} \mu^{(1)} (1 + c^{(2)} \omega)^2 + c^{(2)} \mu^{(2)} (1 - c^{(1)} \omega)^2 + c^{(1)} c^{(2)} \mu^{(2)} \omega^2 \}$$

Proc. R. Soc. Lond. A (1992)

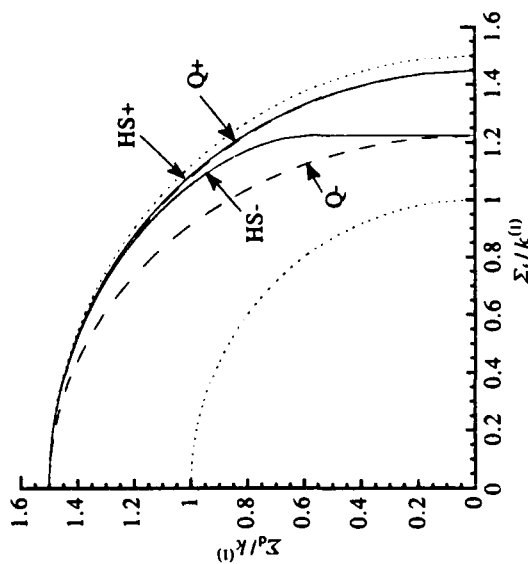


Figure 4. Bounds and estimates for the yield surface of fibre-reinforced composites with $k^{(2)}/k^{(1)} = 2$ and $c^{(1)} = c^{(2)} = 0.5$, plotted as functions of Σ_f and Σ_n . The continuous curves correspond to the Hashin-Shtrikman upper bound and lower estimate, respectively; the top and bottom long-dash curves to the corresponding quadratic interpolations of Hill; and the top and bottom short-dash curves to the Bishop-Hill upper bound and trivial lower bound, respectively.

(μ_{HS} is a given previously), and we have assumed once again that $\mu^{(1)} \leq \mu^{(2)}$. Therefore, assuming that $k^{(1)} \leq k^{(2)}$, we can show that $\pi^{\text{hom}}(D) \leq \pi_{\text{HS}+}^{\text{hom}}(D) + \delta_0(D_m)$, where $\pi_{\text{HS}+}^{\text{hom}}(D) = \inf \{ c^{(1)} k^{(1)} \sqrt{[D_d^2 + (1 + c^{(2)} \omega)^2 D_p^2] + c^{(2)} k^{(2)} \sqrt{[D_d^2 + (1 - c^{(1)} \omega)^2 + c^{(1)} \omega^2] D_p^2}} \}$, (22)

with $D_f = \sqrt{D_n^2 + D_p^2}$. This last result is similar in form to result (19) for the laminated composite; however, the determination of the corresponding effective yield domain $\pi_{\text{HS}+}^{\text{hom}}$ is more difficult (involving a quartic equation), and a simple result, analogous to (20) for the laminate, is not available. However, $\pi_{\text{HS}+}^{\text{hom}}$ may be computed numerically; the result depends on the axisymmetric shear stress invariant Σ_d , and on the following combination of the other two invariants, namely, $\Sigma_f = \sqrt{(\Sigma_n^2 + \Sigma_p^2)}$. It can also be shown that $\Sigma_d \leq c^{(1)} k^{(1)} + c^{(2)} k^{(2)}$ when $\Sigma_f = 0$, and $\Sigma_f \leq k_{\text{HS}+}^{\text{hom}}$, where $k_{\text{HS}+}^{\text{hom}}$ has an expression analogous, but not identical to (15), when $\Sigma_d = 0$.

In analogy with the results for the isotropic composites, it is not possible to use the inequality (11) to obtain a lower bound for π^{hom} , instead, we can only give a 'lower estimate' $\pi_{\text{HS}-}^{\text{hom}}(D) = \pi_{\text{HS}+}^{\text{hom}}(D) + \delta_0(D_m)$, where

$$\pi_{\text{HS}-}^{\text{hom}}(D) = \inf \{ c^{(1)} k^{(1)} \sqrt{[D_d^2 + (1 + c^{(2)} \omega)^2 + c^{(2)} \omega^2] D_p^2} + c^{(2)} k^{(2)} \sqrt{[D_d^2 + (1 - c^{(1)} \omega)^2] D_p^2} \}. \quad (23)$$

Once again, the associated effective strength domain $\pi_{\text{HS}-}^{\text{hom}}$ must be computed numerically. However, when $k^{(1)} \leq \frac{1}{2} \sqrt{(1 + c^{(2)} \omega)^2 + c^{(2)} \omega^2}$, it can be shown (see figure 4) that the yield surface associated with $\pi_{\text{HS}-}^{\text{hom}}$ has a flat section characterized by $\Sigma_f = \sqrt{(1 + c^{(2)} \omega)^2 + c^{(2)} \omega^2}$. On the other hand, when the above condition is not satisfied, the

Proc. R. Soc. Lond. A (1992)

yield surface lacks such a flat section, and $\Sigma_f = k_{HS}^{\text{hom}}$, where k_{HS}^{hom} has an expression analogous to (16)₁, when $\Sigma_d = 0$. Finally, we note that the yield surfaces for the Hashin-Shtrikman lower estimate and upper bound agree when $\Sigma_f = 0$. Thus $\Sigma_d = c^{(0)}k^{(0)} + c^{(2)}k^{(2)}$ when $\Sigma_f = 0$.

Figure 4 shows sample yield surfaces corresponding to P_{HS+}^{hom} and P_{HS-}^{hom} , plotted as functions of Σ_d and Σ_f for a strength ratio of two, and equal volume fractions of the two phases. The associated isotropic Bishop-Hill upper bound and trivial lower bound are also included in this figure for comparison with the new upper bound and lower estimate. It may be observed from this figure that the new Hashin-Shtrikman upper bound is more restrictive than the Bishop-Hill upper bound, but only slightly so. On the other hand, the Hashin-Shtrikman lower estimate is close to the upper bounds for a fairly large range of combined loading. However, when Σ_d is sufficiently small, the lower estimate diverges from the upper bounds: in fact, as pointed out earlier, the lower estimate yield surface may have a flat section in this range. We also note that the lower estimate is significantly stronger than the trivial lower bound. Additionally, it is worth mentioning that the quadratic estimate of Hill (1948) is a good approximation to the upper bound, but it is not good for the lower estimate.

Lipton (1992) has shown that in the linear case the upper and lower Hashin-Shtrikman bounds for the incompressible, fibre-reinforced composites are attained by (two-dimensional) particulate microstructures involving fibres (matrices) of the weaker (stronger), and stronger (weaker) materials, respectively. Thus, extending this interpretation to the rigid-perfectly plastic composites, we may think of the Hashin-Shtrikman upper bounds as estimates for composites with a distinct fibre phase made up of the weaker phase, and correspondingly of the lower estimates as estimates for composites with a distinct fibre phase made up of the stronger phase. In this context, it is interesting to note that the flat sections of the lower-estimate yield surfaces are consistent with the predictions of Hashin (1980) (see also Dvorak & Bahei-El-Din 1987), who argues, on physical grounds, the existence of two distinct failure modes for fibre-reinforced composites with a weaker matrix phase: a fibre mode in which the composite fails due to fibre failure in tension or compression along the fibres (Σ_d), and a matrix mode in which the matrix fails in shear transverse to, or along, the fibres (Σ_f). Thus, we can see that the flat portion of the lower-estimate yield surface in figure 4 corresponds to the matrix-dominated shear modes, and the smooth curved portion corresponds to the fibre-controlled axi-symmetric mode. Finally, we note that bounds for the yield strength of rigid-perfectly plastic, fibre-reinforced composites have also been obtained by Majumdar & McLaughlin (1975) and, more recently, by de Buhan & Taliercio (1991). However, these authors do not enforce transverse isotropy for the distribution of the fibres in the composites, and hence their results are different from the results of the present work.

This work was supported by the Air Force Office of Scientific Research (Grant no. 91-0161) and by the National Science Foundation under its Materials Research Laboratory program at the University of Pennsylvania (Grant no. DMR-91-20668).

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Submitted by M. E. Fisher; received 22 April 1992; accepted 13 May 1992

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A NEW VARIATIONAL PRINCIPLE AND ITS APPLICATION TO NONLINEAR HETEROGENEOUS SYSTEMS*

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Abstract. A new variational principle is proposed for estimating the effective properties of *nonlinear* heterogeneous systems. The variational principle expresses the effective energy-density function of a given nonlinear heterogeneous dielectric in terms of an infinite-dimensional optimization problem involving the effective energy-density functions of the class of *linear* heterogeneous comparison materials with variable dielectric coefficients. The variational principle can alternatively be used in an approximate fashion—by optimizing over finite-dimensional subspaces of the *original space of comparison dielectric coefficients*—to obtain bounds on the effective dielectric properties of nonlinear composite materials with isotropic constituent phases in given proportions. The application of the procedure is illustrated by computing an exact estimate for the effective energy-density function of a nonlinear laminated dielectric material (which has overall anisotropic effective behavior). It is also demonstrated that the Talbot-Willis nonlinear extension of the Hashin-Shtrikman variational principle can be generated from the new variational principle by estimating the effective energy of the linear comparison material (in the new variational principle) directly from the *Hashin-Shtrikman variational principle* for linear materials. However, the new variational principle is more general in the sense that it can be used in conjunction with other bounds and estimates for the linear comparison material to yield corresponding bounds and estimates for nonlinear composites.

Key words. nonlinear variational principles, effective properties, composite materials, nonlinear laminates

AMS(MOS) subject classification. 73

1. Introduction. This paper is concerned with the development of new variational principles with a view toward their application in the prediction of the *effective* properties of *nonlinear* heterogeneous systems. Although the results of the paper are applicable to a number of mathematically analogous disciplines, such as nonlinear conductivity, diffusion, and magnetostatics, we set our problem in the context of *nonlinear electrostatics*. Extensions of the results of this paper are also possible in the context of nonlinear infinitesimal elasticity and other analogous fields [20], [21]. Thus in this work we deal with the effective constitutive behavior of nonlinear heterogeneous dielectrics. The problem of the definition and determination of effective properties of heterogeneous systems has a long and distinguished history, which has rekindled the interest of mathematicians, physicists, and engineers in recent years. However, the great majority of the work in this interesting and important field has been in the context of linear constitutive behavior for the material systems. In the next two paragraphs, we briefly review some of the key developments in the field of linear heterogeneous systems that are relevant to the present study.

In the definition of effective properties, we follow the approach of Hill [8], where these properties are defined in terms of the *effective*, or *overall*, energy of the heterogeneous system subject to certain classes of *uniform* boundary conditions. For a general heterogeneous system, different boundary conditions lead to different effective properties for the system. However, it is assumed on physical grounds that, when the size of the typical heterogeneity is small compared to the size of the specimen under

* Received by the editors March 11, 1991; accepted for publication (in revised form) October 17, 1991. This research was supported in part by Air Force Office of Scientific Research grants 89-02388 and 91-0161, by National Science Foundation grant MSM-90-96324, and by the National Science Foundation/Materials Research Laboratory program of the University of Pennsylvania grant DMR-88-19885.

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conclusion, the heterogeneous material behaves like a homogeneous material with effective properties that are independent of the specific boundary conditions and forcing functions applied to the system. It is precisely the question of the existence (and uniqueness) of such effective properties in the limit of vanishing small microstructure that is the subject of study of *homogenization*. Thus this theory introduces an idealized material that corresponds to the limit of a sequence of heterogeneous materials with two distinct length scales: one microscopic L corresponding to the size of the typical heterogeneity, and one macroscopic L corresponding to the size of the specimen of interest and the scale of variation of the boundary conditions and forcing functions. The effective behavior of the homogenized material is then obtained by considering the limit of the behavior of the sequence of heterogeneous materials as the ratio of scales $\varepsilon = l/L$ tends to zero. We also refer to this idealized limit as a "composite," and to its effective properties as the *homogenized* properties. In this work, we are concerned with the effective properties of *general* heterogeneous systems, and we invoke results from homogenization theory whenever we find it interesting to specialize our general results to the special case of the homogenized system, or composite. Two standard references in the context of *periodic* homogenization (dealing exclusively with periodic microstructures) are given by Sanchez-Palencia [23] and Bensoussan, Lions, and Papanicolaou [2]. An interesting review article written in the context of linear elasticity dealing with recent developments, including nonperiodic microstructures, is given by Kohn [9].

Concerning the determination of effective properties, still in the context of linear constitutive behavior, several methods have been proposed in the literature. One approach is to identify specific microstructures for which the effective properties can be computed *exactly*. An alternative approach, perhaps more useful in practice (since often the exact microstructure is not known), is to assume only partial information (such as the volume fraction of the phases) about the microstructure of the heterogeneous material and to compute *bounds* for its effective properties, considering that the problem is not deterministic. Thus the problem becomes characterizing *range* of possible behaviors by specifying *optimal* (i.e., attainable) bounds on the effective properties of classes of materials systems (e.g., heterogeneous systems with given volume fractions of the constituent phases). A third, *approximate*, approach consists in the postulation of ad hoc models for systems that are based on some physical intuition about the microstructure of the system. Examples of the first approach are provided by the "homogenization" formulae for periodic composites [2], [23], the composite-spheres assemblage model of Hashin and Shtrikman for two-phase [7] and multiphase systems [17], and the sequentially layered laminates introduced by Bruggeman [5], which have been used extensively more recently by several investigators [13], [14], [19], [27]. Examples of the second approach are given by the Wiener bounds [30] and by the Hashin-Shtrikman bounds for isotropic [7], [4] and anisotropic systems [10], [11], [13], [14], [27], [31]. Finally, examples of the third approach are provided by the self-consistent and other effective-medium theories initiated by Bruggeman [5], and used more recently by numerous investigators [12], [18].

In contrast to the situation for linear heterogeneous systems, the number of works dealing with the effective properties of nonlinear heterogeneous systems is much smaller. Concerning the definition of effective properties, Willis [32] has generalized the approach of Hill [8] to nonlinear dielectrics, and Marcellini [15] has extended the results of homogenization theory to nonlinear media in the periodic context. In terms of the computation of effective properties for nonlinear heterogeneous materials, we refer to the work of Miksis [16], who obtained approximate results for the effective

properties of a periodic array and a random (in a self-consistent sense) distribution of nonlinear spherical inclusions in a linear matrix. Other approximate methods are provided by the works of Stroud and Hui [24] and Zeng et al. [35] for weakly nonlinear dielectrics. More significantly from our point of view, Willis [32] has proposed an extension of the Hashin-Shtrikman variational principle to nonlinear heterogeneous systems. A thorough derivation of this variational principle is given by Talbot and Willis [25]. This method has been applied to compute bounds and estimates for nonlinear heterogeneous dielectrics by Willis [32] and Talbot and Willis [26]. However, it is important to note that the procedure of Talbot and Willis normally yields only one-sided bounds if a linear comparison material is utilized in connection with a nonlinear composite (with nonquadratic growth for the energy function at infinity). Thus, Aranguren [1] has applied the Talbot-Willis variational principle to compute two-sided bounds on the effective properties of nonlinear dielectric composites by choosing nonlinear comparison materials and solving the nonlinear comparison problem approximately. Unfortunately, his results for the other bound are not very strong, and they do not compare favorably with the Wiener bounds that can be obtained directly from the principles of minimum energy and minimum complementary energy for nonlinear dielectrics.

In this paper, we present the development of a new variational principle that can be used to determine bounds and exact estimates for the effective properties of nonlinear heterogeneous systems. In the next section, we introduce the definition of effective properties through the principles of minimum energy and minimum complementary energy in connection with the two standard types of uniform boundary conditions. In the following section, we develop the new variational principles and prescribe hypotheses for which the new variational principles are precisely equivalent to the corresponding classical minimum energy principles. The new variational principles are designed to yield the effective properties of a nonlinear heterogeneous material in terms of the effective properties of a suitably optimized, heterogeneous, linear material. Given the large number of results for linear composites, this is clearly a worthwhile enterprise. In § 4 we provide a comparison of the new variational principle with the Talbot-Willis variational principles, and we show that, under appropriate hypothesis on the constitutive behavior of the phases, the Talbot-Willis principles can be obtained directly from the new variational principles by estimating the effective properties of the linear comparison materials in the new principles directly from the Hashin-Shtrikman variational principle for linear composites. Thus, in this sense, the new variational principle is more general because it allows for estimates of the effective energy of the linear heterogeneous comparison material other than the Hashin-Shtrikman estimate. This point is illustrated in § 5 by applying the new variational principle to the computation of the effective behavior of a nonlinear laminated material. Other more sophisticated applications, including bounds of the Hashin-Shtrikman and Beran types, are given elsewhere.

2. Effective properties. Consider a heterogeneous dielectric occupying a region in space of unit volume Ω . The constitutive behavior of the material can be characterized in terms of an electric energy-density function $w(\mathbf{x}, \mathbf{E})$, depending on the position vector $\mathbf{x}$ and the electric field $\mathbf{E}(\mathbf{x})$. Under the assumption of local isotropy, we can write

$$(2.1) \quad w(\mathbf{x}, \mathbf{E}) = \phi(\mathbf{x}, E),$$

where $\phi: \Omega \times R \rightarrow R$ is assumed to be integrable in $\mathbf{x}$ and convex and continuous in the magnitude of the electric field E . Here, R is the set of the extended real numbers.

Additionally, we assume that ϕ satisfies the physically based conditions

$$\begin{aligned} \phi(\mathbf{x}, \mathbf{E}) &\geq 0 \quad \forall \mathbf{x}, \mathbf{E}, \\ \phi(\mathbf{x}, 0) &= 0 \quad \forall \mathbf{x}. \end{aligned} \quad (2.2)$$

The constitutive relation between the electric displacement field $\mathbf{D}(\mathbf{x})$ and the electric field $\mathbf{E}(\mathbf{x})$ is then given by

$$\mathbf{D}(\mathbf{x}) = \partial_{\mathbf{E}} w(\mathbf{x}, \mathbf{E}), \quad (2.3)$$

where $\partial_{\mathbf{E}}$ denotes differentiation with respect to $\mathbf{E}$, assuming, of course, differentiability of w . If, on the other hand, w is not differentiable, we may still give $\partial_{\mathbf{E}}$ the interpretation of the subdifferential of convex analysis (Ekeland and Temam [6]).

The effective constitutive behavior of the heterogeneous dielectric can be defined by a relation analogous to (2.3) relating the spatial averages of the fields $\bar{\mathbf{D}}$ and $\bar{\mathbf{E}}$ through the effective energy of the nonlinear dielectric $\bar{W}(\bar{\mathbf{E}})$, given compactly in terms of the minimum energy principle by

$$\bar{W}(\bar{\mathbf{E}}) = \min_{\mathbf{E} \in \mathbf{K}} W(\mathbf{E}), \quad (2.4)$$

where $W(\mathbf{E}) = \int_{\Omega} w(\mathbf{x}, \mathbf{E}(\mathbf{x})) d\mathbf{x}$ is the pertinent energy functional, and

$$\mathbf{K} = \{\mathbf{E} \mid \mathbf{E} = -\nabla \varphi(\mathbf{x}) \text{ in } \Omega, \text{ and } \varphi = -\bar{\mathbf{E}} \cdot \mathbf{x} \text{ on } \partial\Omega\} \quad (2.5)$$

is the set of *admissible* electric fields in some appropriate functional space (see, for example, Toland and Willis [28]). Here φ is the electrostatic potential; the first condition in Ω implies the restricted version of Faraday's law

$$\nabla \times \mathbf{E} = 0, \quad (2.6)$$

and the second on the boundary $\partial\Omega$ ensures that

$$\bar{\mathbf{E}} = \int_{\Omega} \mathbf{E}(\mathbf{x}) d\mathbf{x}. \quad (2.7)$$

On the other hand, the Euler-Lagrange equation of the variational principle is Gauss's law for a vanishing distribution of free charge, namely,

$$\nabla \cdot \mathbf{D} = 0. \quad (2.8)$$

We note that, to guarantee the existence of a minimizer of (2.4), we must add some coercivity hypothesis on W (or ϕ). In fact, on physical grounds, we assume that W grows at least quadratically in $\mathbf{E}$ as $|\mathbf{E}| \rightarrow \infty$. Also, strict convexity of W guarantees uniqueness of the solution. Additionally, we note that, to ensure enough regularity of the minimizer to validate the differential operations implied by (2.6) and (2.8), additional assumptions would be required on W ; however, because heterogeneous materials often have discontinuous properties separated by sharp interfaces, we prefer to give these equations a weak interpretation equivalent to (2.4) with (2.5). Finally, we note that, under the given hypotheses, $\bar{W}(\bar{\mathbf{E}})$ is a convex function (see, for example, the Appendix of Ponte Castañeda and Willis [22]).

Thus, with the above definition of the effective energy $\bar{W}$, we can show [8], [26] that the average displacement field

$$\bar{\mathbf{D}} = \int_{\Omega} \mathbf{D}(\mathbf{x}) d\mathbf{x} \quad (2.9)$$

can be related to the average electric field $\bar{\mathbf{E}}$, as given by (2.7), via the effective relation

$$\bar{\mathbf{D}} = \partial_{\bar{\mathbf{E}}} \bar{W}(\bar{\mathbf{E}}). \quad (2.10)$$

A dual characterization of the effective constitutive behavior of the composite is possible [8], [33] through the dual energy function $\bar{U}$, which can be defined in terms of the principle of minimum complementary energy by

$$\bar{U}(\bar{\mathbf{D}}) = \min_{\mathbf{D} \in \mathbf{S}} U(\mathbf{D}), \quad (2.11)$$

where $U(\mathbf{D}) = \int_{\Omega} w^*(\mathbf{x}, \mathbf{D}(\mathbf{x})) d\mathbf{x}$ is the complementary energy functional, expressed in terms of the polar function (of convex analysis [6]) of the energy function w by

$$w^*(\mathbf{x}, \mathbf{D}) = \sup_{\mathbf{E}} \{\mathbf{E} \cdot \mathbf{D} - w(\mathbf{x}, \mathbf{E})\}, \quad (2.12)$$

and where

$$\mathbf{S} = \{\mathbf{D} \mid \nabla \cdot \mathbf{D} = 0 \text{ in } \Omega, \text{ and } \mathbf{D} \cdot \mathbf{n} = \bar{\mathbf{D}} \cdot \mathbf{n} \text{ on } \partial\Omega\} \quad (2.13)$$

is the set of admissible electric displacement fields. Note that the first condition must be given a weak interpretation if the displacement field happens to be discontinuous. Then, given the conditions implicit in (2.13), relation (2.11) is a weak statement of (2.6), and, if we make use of the definition (2.7) for the average electric field, we have the following relation analogous to (2.10):

$$\bar{\mathbf{E}} = \partial_{\bar{\mathbf{D}}} \bar{U}(\bar{\mathbf{D}}). \quad (2.14)$$

We should emphasize, however, that the duality relation between the local energy functions w and w^* (recall that w is convex and continuous, and by [6, Props. 1.3.1 and 1.4.1])

$$w(\mathbf{x}, \mathbf{E}) = \sup_{\mathbf{D}} \{\mathbf{E} \cdot \mathbf{D} - w^*(\mathbf{x}, \mathbf{D})\}, \quad (2.15)$$

does not generally carry over to the effective energy functions $\bar{W}$ and $\bar{U}$ (which are also known to be convex). In fact, Willis [33] has shown that

$$\bar{W}(\bar{\mathbf{E}}) \geq \bar{U}^*(\bar{\mathbf{E}}). \quad (2.16)$$

The reason for the inequality is related to the fact that definitions (2.4) for $\bar{W}$ and (2.11) for $\bar{U}$ correspond to distinct boundary conditions on the heterogeneous material (Dirichlet versus Neumann conditions), thus leading in general to different effective energies. As we mentioned in the Introduction, however, it follows from the results of homogenization theory that, for a composite (a heterogeneous material that is homogeneous in a large enough scale), strict equality could be satisfied in the above relation. For nonlinear periodic composites [15], for example, explicit expressions for the effective energy-density functions can be obtained in terms of relations analogous to (2.4) and (2.11), but applied to a unit cell of the periodic composite and subjected to periodic boundary conditions.

Finally, we record here for later reference that [6, Prop. 1.4.2]

$$w^*(\mathbf{x}, \mathbf{D}) = \phi^*(\mathbf{x}, \mathbf{D}), \quad (2.17)$$

where ϕ^* is the polar function of ϕ , and D is the magnitude of $\mathbf{D}$.

3. The new variational principles. The new variational principles, just as the complementary-energy and Hashin-Shtrikman variational principles, rely on the Legendre transformation. However, the transformation is carried out on a suitably modified set of variables to achieve comparison with a linear heterogeneous comparison material with effective energy that can be estimated using linear methods. Depending on whether the minimum energy or the complementary-energy formulation is used, we arrive at

two versions of essentially the same result. We begin by considering the minimum energy formulation, leaving consideration of the complementary energy formulation and the comparison between the two for later sections.

3.1. Minimum energy formulation. The starting point is a change of variables $u = h(E)$, with $h: R^+ \rightarrow R^+$ (R^+ is the set of nonnegative reals) given by $h(E) = E^2$. By composition of ϕ with h^{-1} , we obtain a function $f: \Omega \times R^+ \rightarrow R^+$ such that

$$(3.1) \quad f(x, u) = \phi(x, E) = w(x, E).$$

Note that f has the same dependence on x as ϕ and w . Other relevant properties of f are given in the Appendix and will be simply quoted in the body of this section as needed.

Defining the functional F induced by f as follows:

$$(3.2) \quad F(u) = \int_{\Omega} f(x, u(x)) dx,$$

we can rewrite the minimum energy principle (2.4) in the form

$$(3.3) \quad \tilde{W}(\bar{E}) = \min_{u \in K'} F(u),$$

where K' is the set of admissible fields $u(x)$ induced by K in (2.5).

The other key ingredient in the derivation of the variational principle is property (A.8) from the Appendix, stating that

$$(3.4) \quad f(x, u) \geq \sup_{p \geq 0} \{up - f^*(x, p)\},$$

where

$$(3.5) \quad f^*(x, p) = \sup_{u \geq 0} \{up - f(x, u)\}$$

is the convex polar function (or generalized Legendre transform) of f . Note that x is fixed in the above operations, and that the suprema are evaluated over the set of nonnegative p (this follows from the fact that f is nonnegative and such that $f(x, 0) = 0$; see the Appendix). Also, the right-hand side of inequality (3.4) is the bipolar of f , which has the geometric interpretation of the convex envelope of f , and hence the inequality. Propositions I.3.1 and I.4.1 of [6] ensure that equality is achieved in (3.4) if f is convex and continuous. For now, we do not enforce such an assumption on f , and thus our variational principle generally takes the form of an inequality; however, at the end of this development, we remind the reader of this condition yielding complete equivalence between the new variational principle and the corresponding classical principle.

Next, we note for immediate reference that if we define the polar of the functional F by

$$(3.6) \quad F^*(p) = \sup_{u(x) \geq 0} \left\{ \int_{\Omega} u(x)p(x) dx - F(u) \right\},$$

with an analogous expression for the bipolar functional F^{**} , then, by [6, Props. IX.2.1 and IX.2.2], we have that

$$(3.7) \quad F^*(p) = \int_{\Omega} f^*(x, p(x)) dx$$

and

$$(3.8) \quad F^{**}(u) = \int_{\Omega} f^{**}(x, u(x)) dx,$$

respectively. Once again, the restriction over the set of nonnegative functions $u(x)$ follows from the assumption that f (and thus also F) is nonnegative and vanishes at zero (see the Appendix). It follows from (3.4) and (3.8), respectively, that

$$\begin{aligned} F(u) &\equiv \int_{\Omega} f^{**}(x, u(x)) dx \\ &= F^{**}(u) \\ &= \sup_{p(x) \geq 0} \left\{ \int_{\Omega} u(x)p(x) dx - F^*(p) \right\} \\ &= \sup_{p(x) \geq 0} \left\{ \int_{\Omega} u(x)p(x) dx - \int_{\Omega} f^*(x, p(x)) dx \right\}, \end{aligned} \quad (3.9)$$

where the last two identities follow from the definition of the bipolar and from (3.7). Note that equality holds if f is convex.

Combining this result with the variational statement (3.3), we arrive at the result

$$(3.10) \quad \tilde{W}(\bar{E}) \equiv \tilde{W}_-(\bar{E}),$$

where

$$(3.11) \quad \tilde{W}_-(\bar{E}) = \min_{u \in K'} \sup_{p \geq 0} L(u, p),$$

and L is the saddle functional

$$(3.12) \quad L(u, p) = \int_{\Omega} u(x)p(x) dx - \int_{\Omega} f^*(x, p(x)) dx.$$

Note that equality holds in (3.10) if f is convex.

The functional (3.12) is affine (and hence convex) in u and concave (since f^* is convex) in p . Furthermore, $\lim_{p_0 \rightarrow \infty} L(u, p_0) = \infty$ for $p_0 > 0$ fixed (since $u \in K' \Rightarrow u \geq 0$), and hence by [6, Prop. VI.2.3], we have that

$$(3.13) \quad \min_{u \in K'} \sup_{p \geq 0} L(u, p) = \sup_{p \geq 0} \inf_{u \in K'} L(u, p),$$

and therefore

$$(3.14) \quad \tilde{W}_-(\bar{E}) = \sup_{p(x) \geq 0} \left\{ \inf_{u \in K'} \int_{\Omega} p(x)u(x) dx - \int_{\Omega} f^*(x, p(x)) dx \right\}.$$

This last expression can be rewritten in the form

$$(3.15) \quad \tilde{W}_-(\bar{E}) = \sup_{p(x) \geq 0} \{ \tilde{W}_0(\bar{E}) - V(\epsilon_0) \},$$

where p has been identified with $\frac{1}{2}\epsilon_0$, and u with E^2 , so that $\tilde{W}_0$ corresponds to the effective energy function of a linear, heterogeneous comparison material with local energy function $w_0(x, E) = \frac{1}{2}\epsilon_0(x)E^2$. Thus

$$(3.16) \quad \tilde{W}_0(\bar{E}) = \min_{E \in K'} \int_{\Omega} w_0(x, E) dx,$$

and

$$(3.17) \quad V(\epsilon_0) = \int_{\Omega} v(x, \epsilon_0(x)) dx$$

is the functional generated by the function $v(\mathbf{x}, \varepsilon_0) = f^*(\mathbf{x}, \frac{1}{2}\varepsilon_0)$. We note that the comparison dielectric coefficient $\varepsilon_0(\mathbf{x})$ in (3.15) is an arbitrary nonnegative function in some appropriate functional space. Also, we have the following physical interpretation of the variational principle given by (3.10) together with (3.15): The effective energy function of a *linear* heterogeneous dielectric can be estimated by the coefficient $\varepsilon_0(\mathbf{x})$, whose precise variation is determined by the new variational principle. Thus statements (3.10) and (3.15) for the effective energy function constitute a global version of the corresponding statement (3.4) for the local energy function of the heterogeneous material. Note that this last relation can also be written in the form

$$(3.18) \quad w(\mathbf{x}, \mathbf{E}) \geq \sup_{\varepsilon_0 \geq 0} \{w_0(\mathbf{x}, \mathbf{E}) - v(\mathbf{x}, \varepsilon_0)\},$$

where

$$(3.19) \quad v(\mathbf{x}, \varepsilon_0) = \sup_{\mathbf{E}} \{w_0(\mathbf{x}, \mathbf{E}) - w(\mathbf{x}, \mathbf{E})\}$$

is, in turn, equivalent to (3.5).

We complete this section by making the following two observations. First, we remind the reader that if the local energy function for the nonlinear composite w is assumed to be such that f is convex, then equality applies in (3.4), and hence in (3.18). More importantly for the purposes of this investigation, however, equality would also then hold in (3.10), and we have complete equivalence between the classical minimum energy principle (2.4) and the new variational principle (3.15) under such a hypothesis. The second observation relates to the use of the new variational principle in applications. In the form (3.15), the new variational principle is, at best, as intractable as the classical minimum principle. This is because (3.15) requires the solution of a linear problem with arbitrary inhomogeneity, namely, the solution of (3.16) for arbitrary $\varepsilon_0(\mathbf{x})$. This would be very difficult, in general, but it lends itself to easy approximation by restricting the class of functions to which $\varepsilon_0(\mathbf{x})$ belongs to a smaller class, such as, for example, the class of piecewise constant functions. We note that the result of this approximation would still yield a lower bound in (3.10), even if f is convex. We pursue this point in a later section.

3.2. Complementary energy formulation. Here we proceed similarly to the way we proceeded in the minimum energy formulation by introducing an analogous change of variables $v = h(D)$, where the function h has been specified in the previous section. This change of variables induces (by composition) a function $g: \Omega \times R^+ \rightarrow R^+$ such that

$$(3.20) \quad g(\mathbf{x}, v) = \phi^*(\mathbf{x}, D) = w^*(\mathbf{x}, D).$$

We note, as in the previous section, that the relevant properties of g are given in the Appendix.

We can then express the effective complementary energy of the nonlinear heterogeneous dielectric via

$$(3.21) \quad \tilde{U}(\bar{D}) = \min_{v \in S} G(v),$$

where G is the functional given by

$$(3.22) \quad G(v) = \int_{\Omega} g(\mathbf{x}, v(\mathbf{x})) \, dx,$$

and S is the set of admissible functions $v(\mathbf{x})$ induced by the set S in (2.13).

Next, we introduce, following (A.9) in the Appendix, the concave polar of g (see [29, § 7.14])

$$(3.23) \quad g_*(\mathbf{x}, q) = \inf_{v \geq 0} \{vq - g(\mathbf{x}, v)\}.$$

Whereas the former prescription (3.5) is useful when the function f is convex, the latter prescription (3.23) is more useful if we anticipate that the function g may be concave. Indeed, we have good reason to anticipate different behaviors for g than for f . It is related to the growth conditions for the energy-density functions as the intensity of the fields becomes large. Thus, for a *nonlinear* dielectric, we may wish to impose (on physical grounds) the condition that $w(\mathbf{x}, \mathbf{E}) \sim E^{1+n}$ ($n > 1$) as $E \rightarrow \infty$; i.e., w is stronger than quadratic. Then f is stronger than affine at infinity, which is consistent with convexity of f . On the other hand, the above assumption for w implies that $w^*(\mathbf{x}, D) \sim D^{1-1/n}$ as $D \rightarrow \infty$, and therefore g is weaker than affine at infinity, which is consistent with concavity of g .

According to (A.10) in the Appendix, we have that

$$(3.24) \quad g(\mathbf{x}, v) \leq \inf_{q \geq 0} \{vq - g_*(\mathbf{x}, q)\},$$

with equality if g is concave and continuous. Noting that the concave polar of G is given by

$$(3.25) \quad G_*(q) = \int_{\Omega} g_*(\mathbf{x}, q(\mathbf{x})) \, dx,$$

with an analogous expression for the bipolar G_{**} , result (3.24) yields

$$(3.26) \quad G(v) \leq \inf_{q(\mathbf{x}) \geq 0} \left\{ \int_{\Omega} q(\mathbf{x})v(\mathbf{x}) \, dx - \int_{\Omega} g_*(\mathbf{x}, q(\mathbf{x})) \, dx \right\}.$$

This last result, in turn, implies that

$$(3.27) \quad \tilde{U}(\bar{D}) \leq \tilde{U}_*(\bar{D}),$$

where

$$(3.28) \quad \tilde{U}_*(\bar{D}) = \inf_{q(\mathbf{x}) \geq 0} \left\{ \int_{\Omega} q(\mathbf{x})v(\mathbf{x}) \, dx - \int_{\Omega} g_*(\mathbf{x}, q(\mathbf{x})) \, dx \right\}.$$

In this derivation, we use the fact that infima can be interchanged. Then, by identifying q with $(2\varepsilon_0)^{-1}$ and introducing the notation $w_0^*(\mathbf{x}, D) = (1/2\varepsilon_0(\mathbf{x}))D^2$, we can express the previous result in the form

$$(3.29) \quad \tilde{U}_*(\bar{D}) = \inf_{\varepsilon_0(\mathbf{x}) \geq 0} \{\tilde{U}_0(\bar{D}) - Y(\varepsilon_0)\},$$

where

$$(3.30) \quad \tilde{U}_0(\bar{D}) = \min_{D \in S} \int_{\Omega} w_0^*(\mathbf{x}, D) \, dx$$

and

$$(3.31) \quad Y(\varepsilon_0) = \int_{\Omega} g_*(\mathbf{x}, \frac{1}{2\varepsilon_0(\mathbf{x})}) \, dx.$$

Thus expression (3.29) gives an alternative formulation of the new variational principle, yielding an estimate for the effective complementary-energy function of the nonlinear heterogeneous material in terms of the effective complementary-energy functions of the class of linear heterogeneous comparison materials (3.30) and the functional (3.31).

We note further that if g is concave, equality holds in (3.27). In this case, the classical complementary-energy principle and the new variational principle are equivalent. This suggests that $\tilde{U}_*$ and $\tilde{W}_*$ should be related by duality in exactly the same way as $\tilde{U}$ and $\tilde{W}$. This is explored in the next section.

3.3. On the connection between the two formulations. We start by recalling result (2.16), which can be expressed in the form

$$(3.32) \quad \tilde{W}(\tilde{\mathbf{E}}) \cong \sup_{\tilde{\mathbf{D}}} \{\tilde{\mathbf{D}} \cdot \tilde{\mathbf{E}} - \tilde{U}(\tilde{\mathbf{D}})\}.$$

We note that this result applies not only to the nonlinear heterogeneous material with effective energy functions $\tilde{U}$ and $\tilde{W}$, but also, in particular, to the linear comparison material with effective energy density functions $\tilde{U}_0$ and $\tilde{W}_0$. Next, we make use of (3.27), which, together with (3.32), leads to

$$(3.33) \quad \tilde{W}(\tilde{\mathbf{E}}) \cong \tilde{U}^*(\tilde{\mathbf{E}}) \cong \tilde{U}^*(\tilde{\mathbf{E}}),$$

where

$$(3.34) \quad \begin{aligned} \tilde{U}^*(\tilde{\mathbf{E}}) &= \sup_{\tilde{\mathbf{D}}} \{\tilde{\mathbf{D}} \cdot \tilde{\mathbf{E}} - \inf_{\epsilon_0(\mathbf{x}) \geq 0} \{\tilde{U}_0(\tilde{\mathbf{D}}) - Y(\epsilon_0)\}\} \\ &= \sup_{\tilde{\mathbf{D}}} \sup_{\epsilon_0(\mathbf{x}) \geq 0} \{\tilde{\mathbf{D}} \cdot \tilde{\mathbf{E}} - \tilde{U}_0(\tilde{\mathbf{D}}) + Y(\epsilon_0)\} \\ &= \sup_{\epsilon_0(\mathbf{x}) \geq 0} \{\sup_{\tilde{\mathbf{D}}} \{\tilde{\mathbf{D}} \cdot \tilde{\mathbf{E}} - \tilde{U}_0(\tilde{\mathbf{D}})\} + Y(\epsilon_0)\}. \end{aligned}$$

Here, we have used the facts that suprema can be interchanged and also that $\sup\{-f\} = -\inf\{f\}$. Finally, using Result A.1 in the Appendix, stating that $g_*(\mathbf{x}, q) = -f^*(\mathbf{x}, p)$ with $p = 1/(4q)$, we arrive at $Y(\epsilon_0) = -V(\epsilon_0)$, and hence we have that

$$(3.35) \quad \tilde{U}^*(\tilde{\mathbf{E}}) = \sup_{\epsilon_0(\mathbf{x}) \geq 0} \{\tilde{U}_0^*(\tilde{\mathbf{E}}) - V(\epsilon_0)\}.$$

Evidently, this lower bound for $\tilde{W}$, obtained by dualizing the upper bound for $\tilde{U}$ from the complementary energy formulation of § 3.2, is identical in form to the lower bound $\tilde{W}_*$, given by (3.15), obtained directly from the minimum energy formulation. The only difference here is that the effective energy function of the linear comparison material is determined from the complementary energy formulation (3.30), instead of the minimum energy formulation (3.16). Hence, comparing the two lower bounds for $\tilde{W}$, and making use of (3.32), as applied to the linear heterogeneous material, we have the important result that

$$(3.36) \quad \tilde{W}(\tilde{\mathbf{E}}) \cong \tilde{W}_*(\tilde{\mathbf{E}}) \cong \tilde{U}^*(\tilde{\mathbf{E}}).$$

We remark that if the effective properties of the heterogeneous material do not depend on the type of boundary condition applied (Dirichlet versus Neumann), as is the case for a composite, then equality is obtained in (3.32) (in particular, also for the linear comparison material), and thus we also have equality between $\tilde{W}$ and $\tilde{U}^*$. Furthermore, we have already noted that, under appropriate assumptions on the local energy function w , namely, convexity of f and concavity of g , we also obtain equality between $\tilde{W}$ and $\tilde{W}_*$, and $\tilde{U}$ and $\tilde{U}_*$, respectively. Combining these last two remarks for a composite, we have exact equivalence between the classical minimum principles (2.4) and (2.11), and the new variational principles (3.15) and (3.29). Result A.2 in the Appendix shows that concavity of g implies convexity of f (and hence convexity of w). Thus we have (see Remark A.2) that concavity of g provides a sufficient hypothesis for the complete equivalence of the classical and new variational principles. Remark

A.1 in the Appendix points out the relationship between the convexity (concavity) hypothesis on $f(g)$ and the growth conditions on $w(w^*)$. We note in this connection that, while the analysis carried out in this investigation assumed that the growth of w was stronger than quadratic as the intensity of the fields becomes large, the whole analysis could be repeated for the converse hypothesis on w (subquadratic growth), essentially by interchanging the roles of w and w^* .

4. The Talbot-Willis variational principles. As an application of the new variational principles, in this section we provide an alternative derivation of the Talbot-Willis [25] extension of the Hashin-Shtrikman variational principles for nonlinear heterogeneous systems. The strategy is to obtain an expression for the effective complementary energy function $\tilde{U}_*$ of the linear heterogeneous comparison material via the standard Hashin-Shtrikman variational principle. Then we use this result in the new variational principle to obtain an estimate for the effective energy of the nonlinear composite. Finally, we observe that the result of such a procedure agrees with the statement of the nonlinear Talbot-Willis variational principle.

The Hashin-Shtrikman variational principle in its complementary-energy form, as applied to the linear comparison material, can be written in the form (see [31])

$$(4.1) \quad \tilde{U}_0(\tilde{\mathbf{D}}) = \inf_{\mathbf{p}} \inf_{\mathbf{D} \in S} \left\{ \int_{\Omega} [w_0^*(\mathbf{D}) + \mathbf{D} \cdot \mathbf{p}] dx - \int_{\Omega} (w_0^* - w_0^*)_*(\mathbf{x}, \mathbf{p}) dx \right\},$$

where

$$(4.2) \quad (w_0^* - w_0^*)_*(\mathbf{x}, \mathbf{p}) = \inf_{\mathbf{D}} \{\mathbf{D} \cdot \mathbf{p} - [w_0^*(\mathbf{x}, \mathbf{D}) - w_0^*(\mathbf{D})]\},$$

where

$$(4.3) \quad w_0^*(\mathbf{D}) = \frac{1}{2\epsilon_0} D^2$$

corresponds to a linear homogeneous material with constant dielectric coefficient $\epsilon_0 \geq 0$. We note that the result provided by (4.1) can be valid only if $\epsilon_0 \leq \min_{\mathbf{x}} \epsilon_0(\mathbf{x})$. Otherwise, the concave polar function defined by (4.2) would become unbounded (at some $\mathbf{x}$), and an alternative definition of the polar function would be required, leading to a different version of the Hashin-Shtrikman variational principle (the sup/inf form instead of the inf/inf form). The choice of the above version of the Hashin-Shtrikman variational principle was made based on the anticipated growth condition of the nonlinear heterogeneous system, and on computational convenience. We note that the result can also be obtained by specializing to a linear heterogeneous system the appropriate form of the Talbot-Willis variational principles (namely, form (3.17) in [25]).

Continuing the demonstration, we combine the above estimate for $\tilde{U}_0$ with the estimate for the effective energy of the nonlinear material $\tilde{U}$, as given by the complementary-energy form of the new variational principles (3.29), where $\tilde{U}_0$ is chosen to correspond to the effective energy function of the linear heterogeneous comparison material (4.1).

Thus we arrive at

$$(4.4) \quad \begin{aligned} \tilde{U}_*(\tilde{\mathbf{D}}) &= \inf_{\epsilon_0 \geq 0} \inf_{\mathbf{p}} \inf_{\mathbf{D} \in S} \left\{ \int_{\Omega} [w_0^*(\mathbf{D}) + \mathbf{D} \cdot \mathbf{p}] dx - \int_{\Omega} (w_0^* - w_0^*)_*(\mathbf{x}, \mathbf{p}) dx + V(\epsilon_0) \right\} \\ &= \inf_{\mathbf{p}} \inf_{\mathbf{D} \in S} \left\{ \int_{\Omega} [w_0^*(\mathbf{D}) + \mathbf{D} \cdot \mathbf{p}] dx - H_1(\mathbf{p}) \right\}. \end{aligned}$$

where

$$H_\epsilon(\mathbf{p}) = \sup_{\epsilon_0 \geq 0} \left\{ \int_{\Omega} (w_\epsilon^* - w_\epsilon^0)_*(\mathbf{x}, \mathbf{p}) \, d\mathbf{x} - V(\epsilon_0) \right\}, \quad (4.5)$$

and where we have used the fact that infima can be interchanged. We note that (4.5) can be expressed in the form

$$H_\epsilon(\mathbf{p}) = \left\{ \int_{\Omega} h_\epsilon(\mathbf{x}, \mathbf{p}) \, d\mathbf{x} \right\}, \quad (4.6)$$

where

$$\begin{aligned} h_\epsilon(\mathbf{x}, \mathbf{p}) &= \sup_{\epsilon_0 \geq 0} \left\{ (w_\epsilon^* - w_\epsilon^0)_*(\mathbf{x}, \mathbf{p}) + g_*(\mathbf{x}, \frac{1}{2\epsilon_0}) \right\} \\ &= \sup_{\epsilon_0 \geq 0} \left\{ \inf_{\mathbf{D}} \{ \mathbf{D} \cdot \mathbf{p} - [w_\epsilon^0(\mathbf{X}, \mathbf{D}) - w_\epsilon^*(\mathbf{D})] \} + g_*(\mathbf{x}, \frac{1}{2\epsilon_0}) \right\} \\ &= \sup_{q \geq 0} \left\{ \inf_{\mathbf{D}} \{ \mathbf{D} \cdot \mathbf{p} + w_\epsilon^*(\mathbf{D}) - qD^2 + g_*(\mathbf{x}, q) \} \right\} \\ &= \inf_{\mathbf{D}} \left\{ \mathbf{D} \cdot \mathbf{p} + w_\epsilon^*(\mathbf{D}) - \inf_{q \geq 0} \{ qD^2 - g_*(\mathbf{x}, q) \} \right\} \\ &\equiv (w^* - w_\epsilon^0)_*(\mathbf{x}, \mathbf{p}). \end{aligned} \quad (4.7)$$

In deriving this last result, we use the identification of q with $(2\epsilon_0)^{-1}$ and the fact that the supremum and the infimum in the third equality can be exchanged. This is accomplished via the appropriate version of the saddle point theorem, using the fact that the argument function is convex in $\mathbf{D}$ (provided that ϵ_0 can be chosen such that $\epsilon_0(\mathbf{x}) \geq \epsilon_1$ at every $\mathbf{x}$) and concave in q . Additionally, in the last step, we use the fact that

$$(4.8) \quad w^*(\mathbf{x}, \mathbf{D}) = g(\mathbf{x}, D^2) \equiv g_{**}(\mathbf{x}, D^2).$$

Hence we observe that we have equality in (4.7) if g is concave. Thus summarizing the above results, we have that, in general,

$$(4.9) \quad \tilde{U}_{TW}(\bar{\mathbf{D}}) \equiv \tilde{U}_*(\bar{\mathbf{D}}),$$

where

$$(4.10) \quad \tilde{U}_{TW}(\bar{\mathbf{D}}) = \inf_{\mathbf{p}} \inf_{\mathbf{D} \in S} \left\{ \int_{\Omega} [w_\epsilon^*(\mathbf{D}) + \mathbf{D} \cdot \mathbf{p}] \, d\mathbf{x} - \int_{\Omega} (w^* - w_\epsilon^0)_*(\mathbf{x}, \mathbf{p}) \, d\mathbf{x} \right\}$$

is a statement of the Talbot-Willis variational principle for the nonlinear heterogeneous material. It is important to note that, under the hypothesis that g is concave, we have from our previous results that equality is attained between $\tilde{U}$ and $\tilde{U}_*$. Because of (4.8), however, we have that equality is also attained between $\tilde{U}_*$ and $\tilde{U}_{TW}$ under this hypothesis, and so we have complete equivalence between the classical, Talbot-Willis, and new variational principles. This is, of course, provided that $w^* - w_\epsilon^0$ is concave,

for otherwise it is easy to show that $\tilde{U} \equiv \tilde{U}_{TW}$, in which case the new variational principle is tighter than the Talbot-Willis variational principle ($\tilde{U}_* \equiv \tilde{U}_{TW}$). However, we can choose ϵ_0 as small as needed (to minimize the lack of concavity of $w^* - w_\epsilon^0$), as long as it is nonnegative.

On the other hand, inequality (4.9) suggests the possibility that the Talbot-Willis variational principle may actually be stronger than the new variational principle under hypotheses that are more general than convexity of g (but still with concavity of $w^* - w_\epsilon^0$). This possibility could be interpreted in terms of the following heuristic argument. The new variational principle locally approximates the nonlinear material with energy function w^* by a heterogeneous linear material with a quadratic energy function w_ϵ^0 , whereas the Talbot-Willis variational principle approximates the nonlinear energy function by a homogeneous quadratic energy function w_ϵ^* plus an inhomogeneous linear term, depending on the polarization ($\mathbf{p} \cdot \mathbf{D}$). Thus the Talbot-Willis principle may be better at approximating materials that, although satisfying the appropriate growth conditions in w^* , may have a dominating affine term for small intensities of the fields. However, this potential advantage of the Talbot-Willis variational procedure must be weighted against the previously mentioned disadvantage when $w^* - w_\epsilon^0$ is not concave. Also, it appears that the hypotheses of concavity of g and of convexity of $w^* - w_\epsilon^0$ are unrelated.

We note further that both variational principles can be applied to the characterization of the effective properties of composite materials with homogeneous phases. The method of Talbot and Willis leads to an upper bound for the effective energy function of the composite $\tilde{U}$ by evaluating the infimum in (4.10) over the restricted class of piecewise homogeneous polarizations (with different constant values in each phase), and choosing an appropriate linear (homogeneous) comparison material. The new method also leads to an upper bound for $\tilde{U}$ by evaluating the infimum in (3.29) over the restricted class of linear comparison composites with dielectric properties that are constant over each phase. Examples of the application of the first method are given by the works of Willis [32] and Ponte Castañeda and Willis [22] in the contexts of elastostatics and nonlinear infinitesimal elasticity, whereas applications of the second method have been given by Ponte Castañeda [20], [21] in the context of nonlinear infinitesimal elasticity. It is interesting to note that, while the results based on the new method for a porous material [20] were initially stronger than the corresponding results of [22] based on the older Talbot-Willis method for a choice of material behavior such that the difference between the energy function of the nonlinear material and that of the linear comparison material ($w^* - w_\epsilon^0$) was not concave, Willis [34] has shown recently that the improved results of [20] can also be obtained directly from the Talbot-Willis method by means of a better ("optimal") choice of the comparison material. However, the choice of such an optimal material is complicated by the lack of concavity of $w^* - w_\epsilon^0$, and this was the reason behind the initial deficiency in the Talbot-Willis bounds of [22]. This complication appears to be intrinsic to the Talbot-Willis method. The question remains as to the *weakest* (i.e., weaker than concavity of g) conditions that can be stated, ensuring complete equivalence between the *exact* versions of the two variational principles. However, this question is outside of the scope of this work and is not pursued here.

We note for completeness that the variational principles of Talbot and Willis are more general than the new variational principles, as developed so far, in the sense that the derivation of the new variational principles assumed isotropic behavior for the phases, whereas the Talbot-Willis variational principles also hold for anisotropic phases.

We conclude this section by remarking that, under appropriate hypothesis on w , there are exact dual versions of (4.9) and (4.10), as provided by

$$(4.11) \quad \tilde{W}_{TW}(\tilde{\mathbf{E}}) \equiv \tilde{W}_-(\tilde{\mathbf{E}}),$$

and

$$(4.12) \quad \tilde{W}_{TW}(\tilde{\mathbf{E}}) = \sup_{\mathbf{p}} \inf_{\mathbf{E} \in K} \left\{ \int_{\Omega} [w_{\mathbf{E}}(\mathbf{E}) + \mathbf{E} \cdot \mathbf{p}] \, dx - \int_{\Omega} (w - w_{\mathbf{E}})^*(\mathbf{x}, \mathbf{p}) \, dx \right\},$$

with $(w - w_{\mathbf{E}})^*(\mathbf{x}, \mathbf{p}) = \sup_{\mathbf{E}} \{ \mathbf{E} \cdot \mathbf{p} - [w_{\mathbf{E}}(\mathbf{x}, \mathbf{E}) - w_{\mathbf{E}}(\mathbf{E})] \}$. We note, finally, that if the roles of the assumed growth conditions in w and w^* are interchanged, then the other two versions of the variational principles of Talbot and Willis [25] can also be related to the corresponding versions of the new variational principles (3.15) and (3.29).

5. Application to nonlinear laminated composites. As a means of illustrating the use of the newly developed variational principles, we consider in this section the application to laminated materials. Such laminated materials consist of N homogeneous phases, which we assume to be isotropic, occupying nonintersecting layered regions $\Omega^{(r)}$ ($r = 1, 2, \dots, N$), with union Ω , that are oriented perpendicular to the lamination direction $\mathbf{n}$. Thus we express the local energy-density function for the laminated material in terms of the characteristic functions $\chi^{(r)}(\mathbf{x} \cdot \mathbf{n})$, and the homogeneous energy-density functions $w^{(r)}(\mathbf{E})$ of each phase via the relation

$$(5.1) \quad w(\mathbf{x}, \mathbf{E}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{x} \cdot \mathbf{n}) w^{(r)}(\mathbf{E}).$$

We also note for later reference that the volume fractions $c^{(r)}$ of the constituent phases are determined by the characteristic functions $\chi^{(r)}$ via the relations

$$(5.2) \quad c^{(r)} = \int_{\Omega} \chi^{(r)}(\mathbf{x} \cdot \mathbf{n}) \, dx.$$

The goal is to determine the effective energy function of the heterogeneous material from which the effective constitutive relation for the laminate can be determined via (2.10). If the thickness of the typical layer is small compared to the size of the laminate (i.e., the laminate is a composite), it is well known that, away from a boundary-layer region close to the boundary of the composite, the fields within the layers are constant. Hence the original problem reduces to an algebraic one involving the determination of the constant fields in each of the layers satisfying certain jump conditions across the layers. These conditions are three in number, and can be expressed in terms of the continuity of the normal component of the displacement field and continuity of the (two) tangential components of the electric field (they are weak statements of (2.8) and (2.6), respectively). Although in principle this task is easily carried out, in practice it may be difficult to obtain explicit results because the jump conditions take the form of sets of complicated nonlinear algebraic equations. However, if the composite is made up of linear phases (with quadratic energy functions in each phase), the jump conditions are also linear, and they can be solved in closed form.

Thus, for a linear laminate with dielectric coefficient given by

$$(5.3) \quad \epsilon_{\mathbf{E}}(\mathbf{x}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{x} \cdot \mathbf{n}) \epsilon_{\mathbf{E}}^{(r)},$$

where the $\epsilon_{\mathbf{E}}^{(r)}$ are the dielectric constants in each phase, the effective energy is given by

$$(5.4) \quad \tilde{W}_0(\tilde{\mathbf{E}}) = \frac{1}{2} \tilde{\mathbf{E}} \cdot (\tilde{\epsilon}_0 \tilde{\mathbf{E}}),$$

where the dielectric constant of the laminate is given by an anisotropic tensor $\tilde{\epsilon}_0$, with principal values $\epsilon_0^h = (\epsilon_0^{-1})^{-1}$ (the harmonic mean) and $\epsilon_0^a = \bar{\epsilon}_0$ (the arithmetic mean) along the normal and tangential (to the layers) directions, respectively. An alternative way to write this result for the special case of a laminate with two anisotropic phases due to Tartar [27] and others is

$$(5.5) \quad c^{(1)}(\tilde{W}_0 - w_0^{(2)})^* = (w_0^{(1)} - w_0^{(2)})^* + c^{(2)} N_0^{(2)},$$

where $N_0^{(2)}$ denotes a quadratic form with degenerate dielectric constant $(\mathbf{n} \otimes \mathbf{n}) / (\mathbf{n} \cdot \epsilon_0^{(2)} \mathbf{n})$, depending on the dielectric constant of one of the phases, say $\epsilon_0^{(2)}$, and the lamination orientation $\mathbf{n}$. This formula, which can be derived easily from the Hashin-Shtrikman variational principle, has the advantage that it lends itself to iteration and has been used to prove the optimality of the Hashin-Shtrikman bounds for two-phase composites with linear phases [13], [14], [27]. By comparison, result (5.4) is limited to laminates with isotropic phases, which is our main concern here.

We then can use the above exact result (5.4) for a linear laminate to estimate the effective energy function of a nonlinear laminate with the same microstructure (same $\chi^{(r)}$). This is done by means of the new variational principle (3.15), which yields

$$(5.6) \quad \tilde{W}(\tilde{\mathbf{E}}) = \sup_{\epsilon_{\mathbf{E}}^{(r)} \geq 0} \left\{ \tilde{W}_0(\tilde{\mathbf{E}}) - \sum_{r=1}^N \epsilon_{\mathbf{E}}^{(r)} w^{(r)}(\epsilon_{\mathbf{E}}^{(r)}) \right\},$$

where

$$(5.7) \quad v^{(r)}(\epsilon_{\mathbf{E}}^{(r)}) = \sup_{\mathbf{E}} \{ \frac{1}{2} \epsilon_{\mathbf{E}}^{(r)} \mathbf{E}^2 - w^{(r)}(\mathbf{E}) \}.$$

In the above expression for the effective energy of the nonlinear laminate, we use the fact that the fields are constant in each layer, and hence it suffices to optimize over piecewise constant comparison dielectric constants $\epsilon_{\mathbf{E}}^{(r)}$. Additionally, we assume convexity of f to ensure equality in (5.6). The above prescription involves two optimizations for each phase, but it can be simplified by the following procedure, which we illustrate for a two-phase laminate.

First, we note that the effective energy of the linear comparison laminate can be expressed in the form

$$(5.8) \quad \tilde{W}_0(\tilde{\mathbf{E}}) = \frac{1}{2} \epsilon_0^a [\mathbf{E}^2 - (\mathbf{E} \cdot \mathbf{n})^2] + \frac{1}{2} \epsilon_0^h (\mathbf{E} \cdot \mathbf{n})^2.$$

Next, we take note of the following identity:

$$(5.9) \quad \epsilon_0^h = \inf_{\omega} \{ c^{(1)} \epsilon_0^{(1)} (1 - c^{(2)} \omega)^2 + c^{(2)} \epsilon_0^{(2)} (1 + c^{(1)} \omega)^2 \},$$

which, together with (5.6) and (5.8), leads to the result

$$\tilde{W}(\tilde{\mathbf{E}}) = \inf_{\omega} \sup_{\epsilon_{\mathbf{E}}^{(r)} \geq 0} \{ [c^{(1)} \epsilon_0^{(1)} (s^{(1)}(\omega))^2 + c^{(2)} \epsilon_0^{(2)} (s^{(2)}(\omega))^2] - c^{(1)} v^{(1)}(\epsilon_{\mathbf{E}}^{(1)}) - c^{(2)} v^{(2)}(\epsilon_{\mathbf{E}}^{(2)}) \},$$

where

$$(5.10) \quad \begin{aligned} s^{(1)}(\omega) &= \sqrt{E^2 - (\mathbf{E} \cdot \mathbf{n})^2} + (1 - c^{(2)} \omega)^2 (\mathbf{E} \cdot \mathbf{n})^2, \\ s^{(2)}(\omega) &= \sqrt{E^2 - (\mathbf{E} \cdot \mathbf{n})^2} + (1 + c^{(1)} \omega)^2 (\mathbf{E} \cdot \mathbf{n})^2, \end{aligned}$$

and where we have interchanged the suprema and the infimum because the hypotheses of the appropriate version of the saddle point theorem are satisfied. Hence, by recourse to relations analogous to (3.18) for each phase (with equality), the above expression simplifies to the final result

$$(5.11) \quad \tilde{W}(\tilde{\mathbf{E}}) = \inf_{\omega} \{ c^{(1)} \phi^{(1)}(s^{(1)}(\omega)) + c^{(2)} \phi^{(2)}(s^{(2)}(\omega)) \},$$

where the reader is referred to relations (2.1) and (5.1) for a reminder of the definition of the functions $\phi^{(1)}$. This remarkably simple expression reduces the original problem involving three algebraic equations (jump conditions) to one nonlinear equation expressed by the optimization operation implicit in (5.11). The result can be generalized to multiple phases by introducing auxiliary optimization problems of the type of (5.9)—one for each additional interface. We note that deBotton and Ponte Castañeda [3] have obtained results analogous to (5.11) for nonlinear elastic systems with isotropic phases. In this situation, two optimizations are needed for each interface because two independent constants are required to characterize isotropic elastic phases. By starting with the dual version of the new variational principle (3.29), we can also determine an expression for the effective complementary energy of the composite that is completely analogous to (5.11).

Finally, we attempt to establish comparisons between our approach and an alternative approach based on the Talbot-Willis variational principle. The derivation of result (5.5) for the linear laminate starting from the Hashin-Shtrikman variational principle can be extended to nonlinear composites by starting with the Talbot-Willis variational principle. Equivalently, the problem can be restated in terms of polarizations, which greatly simplifies the statement of the jump conditions. Following either method, we obtain an expression for the effective energy of the composite of the form

$$(5.12) \quad [(\bar{W} - w_c)^* + N_c]^* = [(w - w_c)^* + N_c]^*,$$

where w_c is the energy function of the linear homogeneous comparison material in the Talbot-Willis variational principle, and N_c is the quadratic form associated with the degenerate dielectric constant tensor $(n \otimes n)/(n \cdot \epsilon \cdot n)$. For the case of two-phase laminates, with one linear phase (say 2), this result simplifies to a result exactly analogous to (5.5) for the linear laminate

$$(5.13) \quad c^{(1)}(\bar{W} - w^{(2)})^* = (w^{(1)} - w^{(2)})^* + c^{(2)}N^{(2)},$$

where the linear phase has dielectric constant $\epsilon^{(2)}$. Thus we observe that the number of optimizations required to determine the effective energy of the nonlinear composite from expressions (5.12) and (5.13) is significantly larger than that required by the procedure based on the new variational principle. Furthermore, the relations based on the Talbot-Willis variational principle suffer from the difficulty associated with energy functions w such that $w - w_c$ is not convex. On the other hand, the results based on the Talbot-Willis variational principle hold for anisotropic phases, whereas the results based on the new variational principle (as developed so far) are limited to isotropic phases.

6. Concluding remarks. In this paper, dual variational principles have been proposed for nonlinear heterogeneous systems with locally isotropic phases. These variational principles are equivalent to the classical minimum principles under appropriate hypotheses. However, the new variational principles have the advantage that they allow the estimation of the effective energy-density functions of nonlinear heterogeneous systems in terms of optimization problems involving the effective energy functions of families of linear heterogeneous comparison systems with arbitrary inhomogeneity. Thus the new variational principles lend themselves to useful approximations in either one of two ways. If the microstructure is specified, the variational principles can be used to estimate, either exactly or in the form of bounds, the effective energy of nonlinear systems in terms of exact estimates for the effective energy of linear comparison systems. Results were given in this work for the special case of a nonlinear laminated composite, where the estimates can be shown to be exact. Results for other, more complicated

structures for which the estimates are also exact will be given elsewhere. If, on the other hand, the microstructure is not completely specified, optimal bounds characterizing the effective behavior of linear comparison systems can be used to generate bounds for the effective behavior of nonlinear systems with identical microstructures. For instance, for a composite linear dielectric with two isotropic phases in specified volume fractions, it is known that the Hashin-Shtrikman bounds characterize the range of all possible behaviors. Thus these bounds can be used in the context of the new variational principle to generate bounds for a nonlinear composite with the same microstructure. This approach was followed in [20], [21] for the case of nonlinearly elastic systems with overall isotropy. For the specific case of the Hashin-Shtrikman characterization, there exists an alternative approach provided by the Talbot-Willis [25] variational principles, and we have shown that these variational principles can, in fact, be derived from the new variational principles. More generally, however, other bounds and estimates for the effective behavior of linear comparison systems can be used to generate, via the new variational principles, corresponding bounds and estimates for nonlinear systems with identical microstructures.

Given the versatility of the proposed variational principles vis-à-vis the wealth of results available for the effective behavior of linear systems, and the ease of use of the approximate methods that follow from the new variational principles, it is anticipated that these variational principles will play an important role in the future characterization of the effective behavior of nonlinear systems.

Appendix. In this section, we derive some results to be used in the main body of the paper. We begin by noting that, under the hypotheses on ϕ given at the beginning of § 2, it is easily shown that ϕ^* is also nonnegative and that $\phi^*(x, 0) = 0$. Then it can be seen that, for $D \geq 0$ and $E \geq 0$, respectively, we have that

$$(A.1) \quad \phi^*(x, D) = \sup_{E \geq 0} \{ED - \phi(x, E)\}$$

and

$$(A.2) \quad \phi(x, E) = \sup_{D \geq 0} \{ED - \phi^*(x, D)\}.$$

Results (A.1) and (A.2) are counterparts of (2.12) for w^* and (2.15) for w provided by identifications (2.1) and (2.17). The reductions in the domains of optimization (from the set of all reals to the set of nonnegative reals) in (A.1) and (A.2) for $D \geq 0$ and $E \geq 0$, respectively, are geometrically evident.

Next, we introduce the function $f: \Omega \times R^+ \rightarrow R^+$ induced by the change of variables $u = E^2$ such that

$$(A.3) \quad f(x, u) = \phi(x, E).$$

Note that f is continuous, but not necessarily convex. Also, f is nonnegative such that $f(x, 0) = 0$.

Similarly, we let the function $g: \Omega \times R^+ \rightarrow R^+$ be given by the change of variables $v = D^2$ such that

$$(A.4) \quad g(x, v) = \phi^*(x, D).$$

Note that g is also continuous, nonnegative, such that $g(x, 0) = 0$, and not necessarily concave.

It follows from (A.1) that

$$(A.5) \quad g(x, v) = \sup_{u \geq 0} \{\sqrt{uv} - f(x, u)\},$$

and from (A.2) that

$$(A.6) \quad f(x, u) = \sup_{v \geq 0} \{\sqrt{uv} - g(x, v)\}.$$

Using arguments similar to the arguments that led to (A.1) and (A.2), we can show that the polar function of f can be expressed in the form

$$(A.7) \quad f^*(x, p) = \sup_{u \geq 0} \{up - f(x, u)\},$$

and therefore we have that

$$(A.8) \quad f(x, u) \geq \sup_{p \geq 0} \{up - f^*(x, p)\},$$

with equality if f should be convex.

Similarly, anticipating that g may be concave, we define the concave polar (see [29, § 7.14]) of g via

$$(A.9) \quad g_*(x, q) = \inf_{v \geq 0} \{vq - g(x, v)\},$$

and hence

$$(A.10) \quad g(x, v) \leq \inf_{q \geq 0} \{vq - g_*(x, q)\},$$

with equality if g is concave.

With these definitions and preliminaries, we have the following two results.

RESULT A.1. Let f and g be defined as above. Then

$$(A.11) \quad g_*(x, q) = -f^*(x, p),$$

for all $p, q \geq 0$ such that $p = 1/(4q)$.

Proof. We use (A.5) and (A.9) to find that

$$\begin{aligned} g_*(x, q) &= \inf_{v \geq 0} \left\{ vq - \sup_{u \geq 0} \{ \sqrt{uv} - f(x, u) \} \right\} \\ &= \inf_{v \geq 0} \inf_{u \geq 0} \{ vq - \sqrt{uv} + f(x, u) \} \\ &= \inf_{u \geq 0} \left\{ \inf_{v \geq 0} \{ vq - \sqrt{uv} \} + f(x, u) \right\}, \end{aligned}$$

where we use the fact that infima can be interchanged. However,

$$\inf_{v \geq 0} \{ vq - \sqrt{uv} \} = -\frac{u}{4q},$$

and therefore

$$\begin{aligned} g_*(x, q) &= \inf_{u \geq 0} \left\{ -\frac{u}{4q} + f(x, u) \right\} \\ &= -\sup_{u \geq 0} \{ pu - f(x, u) \} \\ &= -f^*(x, p), \end{aligned}$$

where we have identified p with $1/(4q)$.

Remark A.1. The above result implies that f^* is finite if and only if g_* is finite. Clearly, the satisfaction of these conditions requires certain growth conditions on f and g as $u, v \rightarrow \infty$. For example, a sufficient condition ensuring that f^* is finite for finite p is that f should be bounded for finite u and grow stronger than linearly as $u \rightarrow \infty$. This implies, on the other hand, that g should grow weaker than linearly as $v \rightarrow \infty$. Correspondingly, these results imply that $w(w^*)$ should grow stronger (weaker) than quadratically as $E \rightarrow \infty$, consistently with our prior coercivity assumptions on w .

RESULT A.2. If, in addition to the above hypotheses on f and g , we further assume that g is concave, then f is convex.

Proof. By means of results (A.6) and (A.10) with concavity of g , we have that

$$\begin{aligned} f(x, u) &= \sup_{v \geq 0} \left\{ \sqrt{uv} - \inf_{q \geq 0} \{ vq - g_*(x, q) \} \right\} \\ &= \sup_{v \geq 0} \sup_{q \geq 0} \{ \sqrt{uv} - vq + g_*(x, q) \} \\ &= \sup_{q \geq 0} \left\{ \sup_{v \geq 0} \{ \sqrt{uv} - vq \} + g_*(x, q) \right\} \\ &= \sup_{q \geq 0} \left\{ \frac{u}{4q} - f^*\left(x, \frac{1}{4q}\right) \right\} \\ &= f^{**}(x, u), \end{aligned}$$

where we use the fact that suprema can be interchanged, as well as Result A.1. Hence f is convex.

Remark A.2. If we assume that f is convex, and use arguments analogous to the ones used in the proof of Result A.2, we only arrive at the result that $g \leq g_{**}$, which is not enough to ensure concavity of g . Hence it appears that the assumption of concavity of g is stronger than that of convexity of f . Also, note that since concavity of g implies convexity of f , and this, in turn, implies convexity of w , we have that concavity of g implies convexity of w and w^* . It would be of general interest to determine more precise conditions relating f and g .

Acknowledgments. The author acknowledges insightful discussions with Professor J. R. Willis. The author also thanks Professors R. V. Kohn and G. W. Milton for valuable discussions. The observation that the effective energy of nonlinear laminates can be expressed in the forms (5.12) and (5.13) is due to them (unpublished work). The observation that the effective energy of the two-phase nonlinear laminate can be reduced to a one-dimensional optimization problem comes from the author's collaborative work with G. deBotton in the context of nonlinear infinitesimal elasticity [3]. Finally, the author thanks G. deBotton for his valuable comments on an earlier version of this manuscript.

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Reference [24]

Bounds and estimates for the properties of nonlinear heterogeneous systems

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Contents

1. Introduction	532
2. Effective properties	535
(a) Definition	535
(b) Classical minimum energy principles	536
(c) New variational principles	538
3. Bounds	541
(a) Weiner lower bound	543
(b) Hashin-Shtrikman lower bound	545
(c) Beran lower bound	547
(d) Upper bounds and estimates	548
4. Exact estimates	549
(a) Simple laminates	553
(b) Iterated laminates	554
5. Application to two-phase composites	558
(a) The nonlinear material with perfectly conducting inclusions	559
(b) The nonlinear material with perfectly insulating inclusions	560
(c) Special results for pure power materials	561
6. Concluding remarks	564
Appendix	565
References	566

A powerful and versatile variational principle, allowing the estimation of the effective properties of nonlinear heterogeneous systems, has been introduced recently by Ponte Castañeda (1992). The central idea is to express the effective energy-density function of a given nonlinear composite in terms of an optimization problem involving the effective energy-density functions of linear comparison composites with similar microstructure. This permits the computation of bounds and estimates for the effective properties of given classes of nonlinear heterogeneous systems directly from well-known bounds and estimates for the effective properties of corresponding classes of linear comparison composites. In this paper, we review the variational principle and apply it to determine bounds and estimates for the effective properties of certain classes of nonlinear composite dielectrics with homogeneous, isotropic phases. Thus, nonlinear bounds of the Hashin-Shtrikman and Beran types are obtained for composites with overall isotropy and prescribed volume fractions (of the phases). While nonlinear (second-order) bounds of the Hashin-Shtrikman type have been obtained previously, in different form, by other methods, the nonlinear

(higher-order) Beran bounds are the first of their type. Finally, exact estimates are also obtained for nonlinear composites with 'sequentially layered' microstructures. These special composites, which have proved to be extremely useful in assessing the optimality of bounds for linear systems, are also useful, although to a lesser extent, in assessing the sharpness of the nonlinear bounds.

1. Introduction

This paper deals mainly with the development of methods for estimating and bounding the effective properties of nonlinear heterogeneous systems. Although the presentation of the analyses and results will be phrased in the context of nonlinear electrostatics, the emphasis is not on determining the nonlinear dielectric constant of any specific material system, but rather to develop methods that would be of general application in connection with several of the physical and mechanical properties of material systems. Thus, we could equally well have chosen to set our problem in the context of nonlinear conductivity, magnetostatics or diffusion. More rudimentary versions of the methods in this paper have also been applied by Ponte Castañeda (1991*a, b*) to more complex systems, such as nonlinearly viscous materials.

The study of the effective behaviour of heterogeneous systems is a classical problem that has attracted the attention of numerous investigators in many different fields. However, most of the efforts thus far have concentrated on the effective behaviour of linear systems. In particular, the problem of estimating the effective dielectric constant (or, analogously, the effective conductivity) of a linear dielectric (or conductor) has been a central one. On the other hand, the study of nonlinear heterogeneous systems had not received much attention until very recently, except in some highly specialized areas, such as nonlinear optics. Some examples of nonlinear material behaviour in the context of electrical-transport phenomena are provided by dielectric breakdown, burning out of fuses and laser phenomena. Many other examples could be given in the realm of electrical, and other physical and mechanical properties of matter. In the next few paragraphs, I briefly review some of the key relevant developments in the theory of linear heterogeneous systems, as well as some of the more recent developments in the emerging nonlinear theories.

As is by now well known, the effective properties of most heterogeneous systems are not characterized by a simple average of the properties of the constituent phases weighted by their respective volume fractions, and in general involve dependence on microstructural parameters other than the volume fractions. Thus a proper definition of effective properties is required to have a well-founded theory. One possibility that applies to a large number of systems, introduced by Hill (1963) and Hashin (1964) in the context of elasticity, is to define these properties in terms of the effective, or overall, energy of the heterogeneous system resulting from special classes of uniform boundary conditions. According to this definition, different boundary conditions will in general lead to different effective properties for the system. However, on physical grounds, it is assumed that, as the size of the typical heterogeneity becomes small compared to the size of the specimen under consideration, the heterogeneous material behaves like a homogeneous material with effective properties that are independent of the specific boundary conditions applied to the system.

This intuitive notion of effective properties has been made mathematically rigorous by the field of homogenization, which deals with the study of the existence

and uniqueness of such effective properties in the limit of vanishingly small microstructure. Thus homogenization theory is concerned with the description of an idealized material that corresponds to the limit of a sequence of heterogeneous materials with two distinct length scales: one microscopic, l , corresponding to the size of the typical heterogeneity, and one macroscopic, L , corresponding to the size of the specimen of interest and the scale of variation of the boundary conditions and forcing functions applied to the system. The effective behaviour of the homogenized materials is then obtained by considering the limit of the behaviour of the sequence of heterogeneous materials as the ratio of scales, $\epsilon = l/L$, tends to zero. I also refer to this idealized limit material as a 'composite'. Two standard references in the context of periodic homogenization are provided by the monographs of Sanchez-Palencia (1980) and Bensoussan *et al.* (1978).

Many different methods have been proposed in the literature to predict the effective properties of linear heterogeneous systems, and they can generally be classified into three different categories. The first class of methods consists in identifying specific microstructures for which the effective properties can be computed exactly (e.g. periodic microstructures). An alternative approach, which is often more useful in practice (because the exact microstructure of most composite materials is usually not known precisely), is to define classes of composites for which the microstructure is only partially specified in terms of some known microstructural parameters. The goal of this approach is then to determine the range of possible behaviours for a given material class. Thus this approach seeks to characterize the effective behaviour of classes of material systems by specifying optimal bounds (i.e. bounds that are attainable by special members of the class) on the effective properties of the system. A third approach consists in the postulation of approximate models attempting to capture the essential features of the microstructure of a given system, or class of systems.

Examples of the first approach are provided by the periodic computations of McPhedran & McKenzie (1978), the composite spheres assemblage models of Hashin & Shtrikman (1962) and the sequentially layered laminates, introduced by Bruggeman (1935), and used more recently by Schulgasser (1976) and Milton (1986), among others. Examples of the second approach are given by the bounds of Wiener (1912), Hashin & Shtrikman (1962) and Beran (1965). The Hashin-Shtrikman bounds have been extended and rederived in a number of fundamentally different ways by several authors. Walpole (1966, 1969), Willis (1977), and Kohn & Milton (1988) proposed extensions of the Hashin-Shtrikman variational principles to include, in particular, anisotropic effective behaviour. Bergman (1978) made use of analytic function theory to obtain an alternative derivation of the Hashin-Shtrikman bounds. Tartar (1985) proposed yet a different method, developed jointly with Murat (1978) (see also Murat & Tartar 1985), for bounding the effective properties of anisotropic composites making use of the notions of compensated compactness. Lurie & Cherkov (1984, 1986) independently proposed closely related methods, which also were designed to deal with anisotropic composites. A detailed comparison between the different methods is given by Kohn & Milton (1986) and Milton (1990). The more stringent bounds of Beran have also been given a simpler form by Milton (1981) for the case of two phase isotropic composites in terms of one geometric parameter (additional to the volume fractions) containing third order statistical information. (The Hashin-Shtrikman bounds contain only up to second order information.) Finally, examples of the third approach are provided by the self

consistent and other effective-medium theories, also initiated by Bruggeman (1935), and used more recently by numerous investigators (see Landauer 1978; Milton 1985). For nonlinear heterogeneous systems, the number of investigations dealing with effective behaviour is comparatively small. Thus, in the context of the definition of effective properties, Willis (1986) has extended the approach of Hill (1963) to nonlinear dielectrics, and Marcellini (1978) has extended the results of homogenization theory to nonlinear media in the periodic context. On the other hand, the actual computation of effective properties in the nonlinear context has involved for the most part approximate methods that are generally problem-specific. For example, Miksis (1983) obtained results for the effective properties of periodic arrays, and random (in a self-consistent sense) distributions of nonlinear spherical inclusions in a linear matrix. Other approximate methods have been proposed recently by Stroud & Hui (1988) and Zeng *et al.* (1988) for weakly nonlinear materials. In terms of bounding methods, Willis (1986) and Talbot & Willis (1985) have proposed extensions of the Hashin-Shtrikman variational principles to nonlinear heterogeneous systems. Additionally, Talbot & Willis (1987) have used this method to compute bounds for the effective properties of nonlinear heterogeneous dielectrics, and compared them with the results of some self-consistent embedding procedures.

In this work, I apply variational principles (Ponte Castañeda 1992) to establish bounds and exact estimates for the effective properties of nonlinear heterogeneous systems with isotropic constituents. I begin, in the next section, by introducing the definition of effective properties in terms of the classical principles of minimum energy and minimum complementary energy. Additionally, I summarize the variational principles of Ponte Castañeda (1992) (see Theorems 2.1 and 2.2). These variational principles are designed to yield the effective properties of nonlinear heterogeneous systems in terms of the effective properties of suitably optimized linear heterogeneous systems. This procedure enables the translation of the large number of results available for linear composites directly into corresponding estimates for nonlinear composites. In §3, I use the variational principles (see Corollaries 3.1 and 3.2) to determine rigorous bounds for the effective properties of arbitrarily nonlinear heterogeneous dielectrics with prescribed volume fractions. Thus I show that the so-called Wiener bounds for arbitrarily anisotropic composites, which can be obtained directly from the classical minimum principles, can also be recovered from the new variational principles. More interestingly, I also develop sharper bounds for the class of isotropic nonlinear heterogeneous dielectrics by making use of the corresponding Hashin-Shtrikman bounds for linear dielectrics, and obtain even sharper bounds still for the class of two-phase, isotropic nonlinear systems by applying the Beran bounds for linear systems to the new variational principles. In §4, I introduce special microgeometries for which the effective nonlinear properties can be computed exactly. The new variational principles also play a central role in the derivation of these exact results (see Corollaries 4.1 and 4.2). The special microgeometries correspond to sequentially laminated materials, which have been found to yield the extremal properties of many classes of linear systems, both isotropic and anisotropic, and hence have been used in the linear theories to study the sharpness and optimality of bounds. Thus in §4, I also show that the nonlinear Wiener bounds are sharp. More importantly, I find in §§4 and 5 that, at least for two-phase systems, sequentially laminated nonlinear materials can be constructed closely approximating the nonlinear isotropic bounds of the Hashin-Shtrikman type obtained in §3. This suggests that, although the nonlinear bounds of

§3 may not be optimal in general, they are probably not too far from the optimal bounds (which, presumably, would be extremely difficult to determine precisely).

2. Effective properties

(a) Definition

I consider a heterogeneous dielectric occupying a region in space of unit volume Ω . The nonlinear constitutive behaviour of the material may be characterized in terms of an electric energy density function, $w(x, E)$, depending on the position vector x and the electric field $E(x)$, such that the electric displacement field $D(x)$ is expressed by

$$D(x) = \partial_E w(x, E), \quad (2.1)$$

where ∂_E denotes differentiation with respect to E , assuming differentiability of w . However, in the context of the present work, it will suffice to assume convexity and continuity of w on E . In this event, ∂_E can still be given the interpretation of the subdifferential of convex analysis (Ekeland & Temam 1974; §1.5). As stated in §1, I further assume local isotropy, so that I can write

$$w(x, E) = \phi(x, E), \quad (2.2)$$

where $\phi: \Omega \times R^3 \rightarrow R$ is continuous, convex and coercive (in a sense to be specified later) in the magnitude of the electric field E . Here, R is the set of the extended real numbers. Additionally, I assume that ϕ satisfies the conditions

$$\phi(x, E) \geq 0 \quad \forall x, E, \quad \phi(x, 0) = 0 \quad \forall x. \quad (2.3)$$

It was shown by Hill (1963) (see also Hashin 1964; Willis 1986) that the effective constitutive behaviour of the heterogeneous dielectric may be defined by a relation analogous to (2.1):

$$\bar{D} = \partial_E \bar{w}(E), \quad (2.4)$$

relating the spatial averages of the fields, $\bar{D}$ and $\bar{E}$, through the effective energy-function of the nonlinear dielectric, $\bar{w}(E)$. I emphasize that the effective behaviour of the heterogeneous dielectric, as characterized by $\bar{w}(E)$, may in general be anisotropic, even though the phases themselves are assumed to be isotropic.

In principle, $\bar{w}(E)$ may be determined by solving the electrostatic problem on Ω , given by the restricted version of Faraday's law

$$\nabla \times E = 0, \quad (2.5)$$

and Gauss's law for a vanishing distribution of free charge

$$\nabla \cdot D = 0, \quad (2.6)$$

subject to a uniform boundary condition

$$\varphi = -\bar{E} \cdot x \text{ on } \partial\Omega, \quad (2.7)$$

where φ is the electrostatic potential and is such that $E = -\nabla\varphi(x)$ in Ω (which is equivalent to Faraday's law). Note that boundary condition (2.7) ensures that the average of the electric field is in fact $\bar{E}$, in the sense that

$$\bar{E} = \int_{\Omega} E(x) dx. \quad (2.8)$$

On the other hand, the average displacement field is defined by the similar relation

$$\bar{D} = \int_{\Omega} D(x) dx. \quad (2.9)$$

Thus the effective energy of the heterogeneous dielectric $\bar{W}(\bar{E})$ is computed by evaluating the pertinent energy functional for the heterogeneous dielectric

$$W(E) = \int_{\Omega} w(x, E(x)) dx,$$

at the actual electric field solving the electrostatic problem, as defined by (2.5) to (2.7), for a given microstructure. The effective constitutive behaviour of the heterogeneous material, in the form of a functional relation between $\bar{E}$ and $\bar{D}$, then follows from (2.4). However, due to the complexity of the microstructure of actual materials, it is often impractical to solve the electrostatic problem. For this reason, alternative variational formulations of the problem are helpful. There are two standard (dual) variational formulations of the electrostatic problem: the minimum energy and minimum complementary-energy principles. These will be used in the next subsection to provide alternative descriptions of the effective energy functions of the nonlinear heterogeneous dielectrics.

(b) Classical minimum energy principles

The first standard variational principle is the minimum energy principle. This variational principle, expressed in terms of the energy functional $\bar{W}$, may be used to obtain the following expression for the effective energy of the heterogeneous dielectric, namely,

$$\bar{W}(\bar{E}) = \min_{E \in K} W(E), \quad (2.10)$$

where

$$K = \{E | E = -\nabla q(x) \text{ in } \Omega, \text{ and } q = -\bar{E} \cdot x \text{ on } \partial\Omega\} \quad (2.11)$$

is the set of admissible electric fields.

Note that to guarantee the existence of a minimizer of (2.10) certain conditions on the growth of w (or ϕ) as $E \rightarrow \infty$ are needed. Thus I assume that w is coercive in the sense that $w \rightarrow \infty$ as $E \rightarrow \infty$. Additionally, I remark that strict convexity of w guarantees uniqueness of the solution, and that convexity of w ensures the convexity of $\bar{W}$ (see, for example, the Appendix of Ponte Castañeda & Willis 1988). Finally, note that if the fields are smooth enough, statement (2.10) is equivalent to the electrostatics problem, as given by (2.5) to (2.7). More generally, however, (2.10) provides a weak statement of the electrostatics problem allowing the possibility of discontinuous fields, such as those that would arise in materials with discontinuous properties separated by sharp interfaces.

The second characterization of the effective constitutive behaviour of the heterogeneous dielectric is obtainable from the effective complementary-energy function for the dielectric, $\bar{U}$. This energy function may be defined in terms of the principle of minimum complementary energy via

$$\bar{U}(\bar{D}) = \min_{D \in S} U(D), \quad (2.12)$$

where

$$U(D) = \int_{\Omega} w^*(x, D(x)) dx$$

Properties of nonlinear heterogeneous systems

is the complementary energy functional, expressed in terms of the Legendre-Fenchel dual (convex polar) of the energy function w ,

$$w^*(x, D) = \sup_E \{E \cdot D - w(x, E)\}, \quad (2.13)$$

and where

$$S = \{D | \nabla \cdot D = 0 \text{ in } \Omega, \text{ and } D \cdot n = \bar{D} \cdot n \text{ on } \partial\Omega\}, \quad (2.14)$$

is the set of admissible electric displacement fields. Note that the first condition in this set must be given a weak interpretation if the displacement field happens to be discontinuous. Then, given the conditions implicit in (2.14), relation (2.12) is a weak statement of (2.5). Further, if (2.8) is reinterpreted as a definition for the average electric field, we have the following relation analogous to (2.4), namely,

$$\bar{E} = \partial_D \bar{U}(\bar{D}), \quad (2.15)$$

where it now follows from (2.14) that $\bar{D}$ must satisfy the relation (2.9)

I emphasize, however, that the duality relation between the local energy functions

w and w^* (recall that w is convex and continuous, and hence by Propositions 1.3.1 and 1.4.1 of Ekeland & Temam (1974))

$$w(x, E) = \sup_D \{E \cdot D - w^*(x, D)\}, \quad (2.16)$$

does not necessarily carry over to the effective energy functions $\bar{W}$ and $\bar{U}$ (which is also known to be convex). In fact, Willis (1989) has shown that, in general,

$$\bar{W}(\bar{E}) \geq \bar{U}^*(\bar{E}). \quad (2.17)$$

The reason for the inequality is related to the fact that definitions (2.10) for $\bar{W}$ and (2.12) for $\bar{U}$ correspond to different boundary conditions on the heterogeneous material (Dirichlet versus Neumann conditions), thus leading to generally distinct effective energies.

However, as I mentioned in §1, it follows from the results of homogenization theory that strict equality holds in the above relation for a composite (in the sense of a heterogeneous material that is homogeneous in a large enough scale). In particular, Marcellini (1978) has defined the effective properties of a nonlinear composite with periodic microstructure in terms of relations analogous to (2.4) with (2.10) (or, dually, (2.15) with (2.12)). In this context, the effective properties of the composite are defined by the solution of the electrostatics problem over a unit cell of the composite, with the uniform boundary conditions in (2.11) (or (2.14)) being replaced by periodic conditions over the unit cell. One advantage of this formulation is that equality then holds exactly in (2.17). Although either definition could be used in the present work, I prefer the earlier definition of effective properties (i.e. (2.4) with (2.10), or dually (2.15) with (2.12)), because it has fewer technical requirements (since the limit as the size of the typical heterogeneity vanishes is never evaluated explicitly).

Finally, I record here for later reference that (Ekeland & Temam 1974; §14.2), under the hypotheses given by (2.2) and (2.3),

$$w^*(x, D) = \phi^*(x, D), \quad (2.18)$$

where ϕ^* is the convex polar function (Legendre transform) of ϕ , and D is the magnitude of D .

(c) *New variational principles*

In this subsection, I discuss briefly the variational principles recently proposed by Ponte Castañeda (1992), which will be used in the next sections to determine bounds and estimates for the effective energy functions of nonlinear composites. These variational principles have been shown to be equivalent to the standard variational principles discussed in the previous subsection, under appropriate hypothesis on the energy-density function. Also, the variational principles, just like the classical complementary-energy and Hashin-Shtrikman variational principles, are based on the Legendre transformation; however, the transformation is carried out on a suitably modified set of variables to achieve comparison with a linear heterogeneous comparison material with effective energy that can be estimated by using the linear theory. Depending on whether I start from the minimum energy or complementary-energy formulation, I obtain two versions of essentially the same result. I begin by considering the minimum energy formulation.

The new variational principle centres around a change of variables $r = h(E)$, with $h: R^+ \rightarrow R^+$ (R^+ is the set of non-negative reals) given by $h(E) = E^2$. By composition of ϕ with h^{-1} , we obtain a function $f: \Omega \times R^+ \rightarrow R^+$, such that

$$f(x, r) = \phi(x, E) = w(x, E). \quad (2.19)$$

We note that f has the same dependence on x as ϕ and w , and that it is continuous and coercive (but not necessarily convex) in r . Also, from (2.3), f is a non-negative function satisfying the condition that $f(x, 0) = 0 \forall x$. Then, if I define the Legendre transform (convex polar) of f by

$$f^*(x, p) = \sup_{r \geq 0} \{rp - f(x, r)\}, \quad (2.20)$$

it follows that

$$f(x, r) \geq \sup_{p \geq 0} \{rp - f^*(x, p)\}. \quad (2.21)$$

Note that x is fixed in the above operations, and that the suprema are evaluated over the sets of non-negative r and p , respectively (this follows from the fact that f is non-negative and such that $f(x, 0) = 0$; see the Appendix of Ponte Castañeda (1992)). Also, the right-hand side of inequality (2.21) is the bipolar of f , which has the geometric interpretation of the convex envelope of f , and hence the inequality. Propositions 1.3.1 and 1.4.1 of Ekeland & Temam (1974) ensure that equality is achieved in (2.21) if f is convex and continuous (in r). Therefore, assuming that the energy function w in (2.20) is such that f is convex (note that f convex implies that w is convex), I obtain from (2.21) the following representation for the local energy-density function of the nonlinear heterogeneous material, namely,

$$w(x, E) = \sup_{\epsilon_0 \geq 0} \{w_0(x, E) - r(x, \epsilon_0)\}, \quad (2.22)$$

where (from (2.20))

$$r(x, \epsilon_0) = \sup_E \{w_0(x, E) - w(x, E)\}. \quad (2.23)$$

In the above relations, p has been identified with $\frac{1}{2}\epsilon_0$ and r with E^2 , in such a fashion that $w_0(x, E) = \frac{1}{2}v_0(x)E^2$ and $r(x, \epsilon_0) = f^*(x, \frac{1}{2}\epsilon_0)$. Thus w_0 corresponds to the local energy-density function of a linear, heterogeneous comparison material with arbitrary (not necessarily constant) non-negative dielectric coefficient $\epsilon_0(x)$.

Phil Trans R Soc Lond A (1992)

The minimum energy formulation of the variational principle follows by making use of the representation (2.22) in the classical minimum energy principle (2.10), and interchanging the order of the infimum in (2.20) and the supremum in (2.22), which is allowed by an appropriate version of the saddle-point theorem. The details of the proof are given in Ponte Castañeda (1992, §3.1), but the result is as follows.

Theorem 2.1. *Let the local energy density function w of a given nonlinear heterogeneous material with isotropic phases satisfy condition (2.19) with f a non-negative, continuous, coercive and convex function of $r = E^2$ further satisfying $f(x, 0) = 0 \forall x$. Then, the effective energy function of the nonlinear heterogeneous material $\bar{w}$ is determined by the variational principle*

$$\bar{w}(E) = \sup_{\epsilon_0(x) \geq 0} \{\bar{w}_0(E) - V(\epsilon_0)\}, \quad (2.24)$$

where

$$V(\epsilon_0) = \int_{\Omega} v(x, \epsilon_0(x)) dx, \quad (2.25)$$

is the functional generated by the function $v(x, \epsilon_0)$, and where $\bar{w}_0$ denotes the effective energy function of a linear heterogeneous comparison material with local energy function w_0 , such that

$$\bar{w}_0(E) = \min_{E \in K} \int_{\Omega} w_0(x, E) dx \quad (2.26)$$

I emphasize that the dielectric coefficient $\epsilon_0(x)$ of the comparison material in (2.24) is an arbitrary non-negative function in some appropriate functional space, so that (2.24) is indeed a variational statement. Later, however, I make use of piecewise constant approximations of $\epsilon_0(x)$, leading to finite dimensional optimization problems for bounds and estimates on $\bar{w}$.

The dual, or complementary-energy, formulation of the new variational principle follows in a similar fashion from the change of variables $s = h(D)$, where h is the same as before. This change of variables induces (by composition) a function $g: \Omega \times R^+ \rightarrow R^+$ such that

$$g(x, s) = \phi^*(x, D) = w^*(x, D). \quad (2.27)$$

Again, we note that g has the same dependence on x as ϕ^* and w^* , and that it is continuous and coercive in s . Similarly, it follows from (2.3) that ϕ^* , and therefore g , are non-negative functions satisfying the condition that $g(x, 0) = \phi^*(x, 0) = 0 \forall x$. Then, if we define the concave polar of g by (see Van Tiel 1974, §7.14)

$$g_*(x, q) = \inf_{\epsilon \geq 0} \{sq - g(x, s)\}, \quad (2.28)$$

it follows that

$$g(x, s) \leq \inf_{q \geq 0} \{sq - g_*(x, q)\}, \quad (2.29)$$

with equality if g is concave. Then, assuming that the complementary energy function w^* of the nonlinear heterogeneous material is such that g in (2.27) is concave, it follows from (2.29) that

$$w^*(x, D) = \inf_{\epsilon_0 \geq 0} \{w_0^*(x, D) + v(x, \epsilon_0)\}, \quad (2.30)$$

where q has been identified with $(2\epsilon_0)^{-1}$ and s with D^2 , such that $w_0^*(x, D) = (1/2\epsilon_0(x))D^2$ is the complementary-energy function of the linear, heterogeneous

Phil Trans R Soc Lond A (1992)

comparison material with arbitrary non-negative dielectric coefficient $\epsilon_0(\mathbf{x})$. Here, $\epsilon(\mathbf{x}, \epsilon_0) = -g_*(\mathbf{x}, 1/2\epsilon_0)$, which can alternatively be expressed in the form

$$\epsilon(\mathbf{x}, \epsilon_0) = \sup_{\mathbf{D}} \{w^*(\mathbf{x}, \mathbf{D}) - w_0^*(\mathbf{x}, \mathbf{D})\}. \quad (2.31)$$

We note that this latter result is consistent with definition (2.23) of v , as can be seen by making use of (2.13) in the right hand side of (2.31), and interchanging the resulting suprema.

The complementary energy form of the variational principle then follows by letting w^* in (2.12) take the form given by (2.30), and interchanging the order of the infima. The final result may be stated (see Ponte Castañeda 1992, §3.2) in the following form.

Theorem 2.2. *Let the (convex) local complementary-energy function w^* of a given nonlinear heterogeneous material with isotropic phases satisfy condition (2.27) with g a non-negative, continuous, coercive and concave function of $s = D^2$, further satisfying $g(\mathbf{x}, 0) = 0 \forall \mathbf{x}$. Then, the effective complementary energy function of the nonlinear heterogeneous material $\tilde{V}$ is given by*

$$\tilde{V}(\mathbf{D}) = \inf_{\epsilon_0 > 0} \{ \tilde{V}_\epsilon(\mathbf{D}, \epsilon; V(\epsilon_0)) \}, \quad (2.32)$$

$$\text{where} \quad \tilde{V}_\epsilon(\mathbf{D}) = \min_{\mathbf{D} \leq \mathbf{D}_0} \int_{\mathbf{D}_0} w_0^*(\mathbf{x}, \mathbf{D}) d\mathbf{x} \quad (2.33)$$

stands for the effective complementary energy function of the linear comparison composite, and the functional V is as described by (2.25).

In the above developments of the energy and complementary-energy formulations of the variational principle, the respective hypotheses of convexity of f and concavity of g were introduced independently of each other. It is natural to ask whether there is any relation between these two hypotheses. Further, it is important to emphasize that, without these hypotheses, equivalence between the classical minimum energy and new variational principles would not hold. Both of these issues are addressed in Ponte Castañeda (1992, §3.3), where it is proved that concavity of g implies convexity of f , and thus that concavity of g suffices to ensure the lack of a duality gap between the two versions of the variational principle (2.24) and (2.32), when applied to the same composite. Thus, in the developments to follow, I assume, unless stated otherwise, that w is such that g in (2.27) is concave.

I conclude this section by considering the implications of the hypothesis of concavity of g on the growth conditions on w . Recall that so far we have only assumed explicitly that w is convex and coercive ($w \rightarrow \infty$ as $E \rightarrow \infty$). Since concavity of g implies convexity of f , it implies in turn that $w \geq \alpha E^2$ (for some $\alpha > 0$) as $E \rightarrow \infty$. Thus a sensible growth condition may be that $w(\mathbf{x}, \mathbf{E}) \sim E^{1/\alpha}$ ($\alpha \geq 1$) as $E \rightarrow \infty$ (i.e. w is stronger than, or as strong as, quadratic). Then, f is stronger than, or as strong as, affine at infinity, which is consistent with convexity of f . On the other hand, the above assumption for w implies that $w^*(\mathbf{x}, \mathbf{D}) \sim D^{1/(1-\alpha)}$ as $D \rightarrow \infty$, and therefore g is weaker than, or as weak as, affine at infinity, which is consistent with concavity of g . For a more detailed discussion of this point the reader is referred to Ponte Castañeda (1992, §3.3), but in the developments to follow the above growth conditions will be assumed implicitly. Other growth conditions are possible, but the bounds/estimates that follow may need reinterpretation, if these are different. For

instance, we could let n in the above conditions be such that $0 < n \leq 1$. Then, the suprema and infima in the above relations would have to be replaced by infima and suprema, respectively. Further, the role of the bounds and estimates in the following discussion on bounds will also have to be appropriately adjusted.

3. Bounds

This section is concerned with the determination of bounds and estimates for the effective energy functions of classes of nonlinear heterogeneous dielectrics that are defined by the specification of appropriate statistical information on their microstructure, such as, for example, the volume fractions of their constituent phases. The idea is to make use of corresponding bounds and estimates for linear heterogeneous comparison materials in the context of the variational principles discussed in the previous section. Although the prescriptions determined by this approach can be applied to more general types of bounds and estimates, in this section, we limit our attention to bounds of the Wiener, Hashin-Shtrikman and Beran types.

In particular, we are interested in heterogeneous materials with n homogeneous phases, characterized by the isotropic energy functions $\phi^{(n)}(r = 1, \dots, n)$, such that the local energy function of the heterogeneous material w , as defined by (2.2), takes the form

$$w(\mathbf{x}, \mathbf{E}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) \phi^{(r)}(\mathbf{E}), \quad (3.1)$$

where $\chi^{(r)}(\mathbf{x})$ is the characteristic function of phase r (this function vanishes unless $\mathbf{x}$ is in phase r , in which case it equals unity). We further assume that the volume fractions $c^{(r)}$ of the constituent phases are fixed. These are given by the relations

$$c^{(r)} = \int_D \chi^{(r)}(\mathbf{x}) d\mathbf{x}, \quad \text{and are such that } \sum_{r=1}^n c^{(r)} = 1$$

Additionally, we may, or may not, prescribe further information on the microstructure, such as overall isotropy. From hereon, I refer to the material characterized by (3.1) as a nonlinear composite (even though the relation may also correspond to a family of composites), and hence I assume that the scale of variation of the characteristic functions is small enough.

Because of the approximations that need to be made in the application of the variational principle (2.24) and its dual (2.32), most of the results of this section will be in the form of lower bounds for $\tilde{V}$. However, corresponding upper estimates for rather estimates for the upper bounds, are also proposed. Thus §3a deals with the Wiener lower bound and is concerned with generally anisotropic nonlinear composites with isotropic phases in prescribed volume fractions. On the other hand, §§3b and c, dealing with Hashin-Shtrikman and Beran lower bounds, respectively, are concerned with nonlinear composites with isotropic phases in prescribed volume fractions, which are additionally distributed in such a way that the composite depicts overall isotropy. In addition, §3d addresses the difficulty in obtaining rigorous upper bounds for $\tilde{V}$. More specifically, it is shown that, although the well-known Wiener upper bounds are easily obtainable from the new variational principles, upper bounds of the Hashin-Shtrikman and Beran varieties are much harder to determine. Instead 'upper estimates' of these types are proposed and computed.

We remark that the above choices for classes of bounds are given on account of

and where the function $v^{(r)}$ is obtained by specializing relation (2.31) (or (2.23)) to the r th phase.

Proof. The derivation of this result is analogous to the derivation of the previous result. In this case, we make use of expression (2.32) (Theorem 2.2) and of the fact that the infimum over the set of piecewise fields is larger than the infimum over the set of arbitrarily variable fields.

(a) Wiener lower bound

In this subsection, I make use of the lower bound of Wiener (1912) for linear, anisotropic composites with prescribed volume fractions to generate a corresponding bound for nonlinear, anisotropic composites. I refer to the resulting bound as the nonlinear Wiener bound. I emphasize that, although the nonlinear Wiener bounds may be obtained in exactly the same way as the corresponding linear bounds (directly from the classical variational principles), here I make use of the alternative variational principles of §2c for two reasons. First, the basic ideas in the derivation of the Wiener lower bound via the new variational principles are essentially the same as those to be used later in the derivation of the Hashin Shtrikman and Beran bounds, and thus it is useful to illustrate the general method in the simplest possible case. Second, the resulting form of the Wiener lower bound for the case under consideration (isotropic phases) is new, and simpler than (although equivalent to) the alternative form resulting from direct application of the classical variational principle.

The Wiener lower bound may be specified as a bound on the effective energy functions of linear composites with dielectric constants $\epsilon_0^{(1)}, \epsilon_0^{(2)}, \dots, \epsilon_0^{(n)}$ in prescribed volume fractions $c^{(1)}, c^{(2)}, \dots, c^{(n)}$ via the relation

$$\bar{W}_0(\mathbf{E}) \geq \frac{1}{2} \left(\sum_{r=1}^n \frac{c^{(r)}}{\epsilon_0^{(r)}} \right)^{-1} E^2, \quad (3.4)$$

where $\bar{W}_0(\mathbf{E}) = \frac{1}{2} \bar{\epsilon}_0(\mathbf{E}) \cdot \mathbf{E}$ is the effective energy function of linear composites with effective dielectric tensors $\bar{\epsilon}_0$. Note that beyond the volume fractions of the constituent phases, nothing else is specified about the microstructure of the composite, and that, in particular, the composite may be anisotropic.

The nonlinear Wiener lower bound for the effective energy functions $\bar{W}$ of the class of nonlinear composites with prescribed volume fractions is obtained by applying (3.2) of Corollary 3.1 to the set of nonlinear composites with prescribed volume fractions, and combining this result with the lower bound (3.4) for the class of linear comparison composites with prescribed volume fractions. Therefore,

$$\bar{W}(\mathbf{E}) \geq \sup_{\epsilon_0^{(r)} > 0} \left\{ \frac{1}{2} \left(\sum_{r=1}^n \frac{c^{(r)}}{\epsilon_0^{(r)}} \right)^{-1} E^2 - \sum_{r=1}^n c^{(r)} v^{(r)}(\epsilon_0^{(r)}) \right\}, \quad (3.5)$$

$$\text{where, explicitly,} \quad v^{(r)}(\epsilon_0^{(r)}) = \sup_{\epsilon_0^{(r)} > 0} \{ \frac{1}{2} \epsilon_0^{(r)} s^2 - \phi^{(r)}(s) \}, \quad (3.6)$$

(Clearly, the optimizations implicit in (3.5) and (3.6) are $2n$ in number, but this number may be significantly reduced by means of the following identity, first utilized by deBotton & Ponte Castañeda (1992) and proved in the Appendix, namely,

$$\left(\sum_{r=1}^n \frac{c^{(r)}}{\epsilon_0^{(r)}} \right)^{-1} = \inf_{\omega_0^{(r)} > 0} \left\{ \sum_{r=1}^n c^{(r)} \epsilon_0^{(r)} (1 - \omega_0^{(r)})^2 \right\}. \quad (3.7)$$

their simplicity, and to illustrate the general method. (Other possibilities would include application to the linear bounds and self-consistent estimates of Willis (1977) for anisotropic composites with prescribed two-point correlation functions, and to the bounds of Murat & Tartar (1985) and Lurie & Cherkashin (1984, 1986) for generally anisotropic composites with prescribed volume fractions only (i-closures). This latter class of bounds on the effective conductivity tensor are sharper than the Wiener bounds, but would require, for implementation purposes, alternative expression in the form of energy bounds, analogous to those determined by Kohn & Lipton (1988) and Allaire & Kohn (1991) for linearly elastic systems. Before proceeding with the determination of the bounds and estimates, I state the following useful corollaries to Theorems 2.1 and 2.2.

Corollary 3.1. Let (3.1) characterize the local energy-density function of an n -phase nonlinear composite, additionally satisfying the hypotheses of Theorem 2.1. Then, the effective energy function $\bar{W}$ of the composite satisfies the inequality

$$\bar{W}(\mathbf{E}) \geq \sup_{\epsilon_0^{(r)} > 0} \left\{ \bar{W}_0(\mathbf{E}) - \sum_{r=1}^n c^{(r)} v^{(r)}(\epsilon_0^{(r)}) \right\}, \quad (3.2)$$

where $\bar{W}_0$ denotes the effective energy function of a linear comparison composite with n phases of dielectric constant $\epsilon_0^{(r)}$ in volume fraction $c^{(r)}$, such that the dielectric coefficient of the comparison composite is given by

$$\epsilon_0(\mathbf{x}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) \epsilon_0^{(r)}.$$

and where the function $v^{(r)}$ is obtained by specializing relation (2.23) to the r th phase.

Note that the supremum in (3.2) is evaluated over the set of constants $\epsilon_0^{(r)} (r = 1, \dots, n)$.

Proof. This result follows from expression (2.24) in Theorem 2.1 by restricting the set of (arbitrarily variable) comparison dielectric coefficients $\epsilon_0(\mathbf{x})$ to the set of piecewise constant fields, with a different dielectric constant over each nonlinear phase of the composite (namely, $\epsilon_0^{(r)}$ over phase r). The inequality is a consequence of the fact that the supremum over the set of piecewise constant fields is smaller than the supremum over the original (larger) set of fields. Note that the linear comparison composite has the same microstructure as the nonlinear composite, and that, in particular, it is constituted by n phases of dielectric constants $\epsilon_0^{(r)}$ in volume fractions $c^{(r)}$.

Corollary 3.2. Let the appropriate dual version of (3.1) characterize the local complementary-energy function v^* of an n -phase nonlinear composite, additionally satisfying the hypotheses of Theorem 2.2. Then, the effective complementary-energy function $\bar{V}$ satisfies

$$\bar{V}(\mathbf{D}) \leq \inf_{\epsilon_0^{(r)} > 0} \left\{ \bar{V}_0(\mathbf{D}) + \sum_{r=1}^n c^{(r)} v^*(\epsilon_0^{(r)}) \right\}, \quad (3.3)$$

where $\bar{V}_0$ denotes the effective complementary-energy function of a linear comparison composite with n phases of dielectric constant $\epsilon_0^{(r)}$ in volume fraction $c^{(r)}$, such that the dielectric coefficient of the comparison composite is given by

$$\epsilon_0(\mathbf{x}) = \sum_{r=1}^n \chi^{(r)}(\mathbf{x}) \epsilon_0^{(r)}.$$

where the infimum is over the set of variables $\omega^{(r)}$ ($r = 1, \dots, n$), which are subject to a zero-average constraint

$$\bar{\omega} = \sum_{r=1}^n c^{(r)} \omega^{(r)} = 0.$$

This identity, when applied to the nonlinear lower bound for $\tilde{W}$ in (3.5), yields the result

$$\tilde{W}(\tilde{E}) \geq \sup_{\omega^{(r)} \geq 0} \left\{ \inf_{\omega^{(r)} \geq 0} \left\{ \sum_{r=1}^n c^{(r)} \left[\frac{1}{2} \epsilon^{(r)} (1 - \omega^{(r)})^2 E^2 - \nu^{(r)} (\epsilon_0^{(r)}) \right] \right\} \right\}, \quad (3.8)$$

which in turn leads to

$$\tilde{W}(\tilde{E}) \geq \inf_{\omega^{(r)} \geq 0} \left\{ \sum_{r=1}^n c^{(r)} \sup_{\omega^{(r)} \geq 0} \left\{ \frac{1}{2} \epsilon^{(r)} (1 - \omega^{(r)})^2 E^2 - \nu^{(r)} (\epsilon_0^{(r)}) \right\} \right\}. \quad (3.9)$$

In this last step, I have made use of the fact that the argument of the nested supremum and infimum is concave in the $\epsilon_0^{(r)}$ (since the functions $\nu^{(r)}$ are convex in $\epsilon_0^{(r)}$) and convex in the $\omega^{(r)}$, and of the saddle-point theorem to justify the interchange of the supremum and infimum operations.

Then, finally, application of relation (2.22), specialized to each phase in the form

$$\phi^{(r)}(s) = \sup_{\epsilon_0^{(r)} \geq 0} \left\{ \frac{1}{2} \epsilon_0^{(r)} s^2 - \nu^{(r)} (\epsilon_0^{(r)}) \right\}, \quad (3.10)$$

leads to the following result for the lower bound

$$\tilde{W}(\tilde{E}) \geq \inf_{\omega^{(r)} \geq 0} \left\{ \sum_{r=1}^n c^{(r)} \phi^{(r)}(1 - \omega^{(r)}) \tilde{E} \right\}. \quad (3.11)$$

The form of this bound is much simpler than that given by (3.5) and (3.6), and involves only a n -dimensional optimization, with one linear constraint. This constraint can easily be embedded in the optimization by suitable relabelling of the optimization variables. For instance, for the case of a two-phase composite, the above bound reduces to

$$\tilde{W}(\tilde{E}) \geq \inf_{\omega} \left\{ c^{(1)} \phi^{(1)}(1 - c^{(2)} \omega / \tilde{E}) + c^{(2)} \phi^{(2)}(1 + c^{(1)} \omega / \tilde{E}) \right\}, \quad (3.12)$$

where the optimization variable ω is unconstrained.

Next, for completeness, note that an alternative expression for this lower bound may be obtained dually from expression (3.3) of Corollary 3.2. In this event, I obtain the following upper bound for the effective complementary-energy function $\tilde{V}$ of the nonlinear composite, namely,

$$\tilde{V}(\tilde{D}) \leq \inf_{\omega^{(r)} \geq 0} \left\{ \frac{1}{2} \left(\sum_{r=1}^n \frac{c^{(r)}}{\epsilon_0^{(r)}} \right) \tilde{D}^2 + \sum_{r=1}^n c^{(r)} \nu^{(r)} (\epsilon_0^{(r)}) \right\}, \quad (3.13)$$

and thus

$$\tilde{V}(\tilde{D}) \leq \sum_{r=1}^n c^{(r)} (\phi^{(r)})^*(\tilde{D}),$$

where I have used the result that

$$(\phi^{(r)})^*(\tilde{D}) = \inf_{\epsilon_0^{(r)} \geq 0} \left\{ \frac{1}{2\epsilon_0^{(r)}} \tilde{D}^2 + \nu^{(r)} (\epsilon_0^{(r)}) \right\}, \quad (3.15)$$

which follows from (2.18), together with the fact that the Legendre transform of the energy function $\phi^{(r)}$ (or complementary-energy function) is given by result (2.30), specialized to phase r . We remark that, not surprisingly, the alternative form for the bound (3.14) is precisely the result that would be obtained by applying the classical minimum complementary energy principle directly to the nonlinear composite. Then, by dualizing this last relation, and recalling relation (2.17), I obtain

$$\tilde{W}(\tilde{E}) \geq \left(\sum_{r=1}^n c^{(r)} (\phi^{(r)})^* \right)^*(\tilde{E}). \quad (3.16)$$

For a given composite, we know from the lack of a duality gap (see discussion at the end of §2c) between the two versions of the new variational principle that expressions (3.11) and (3.16) for the lower bound for $\tilde{W}$ must be equivalent. However, the latter form involves $n+1$ Legendre transforms, which in general may be more difficult to evaluate than the earlier equivalent prescription. For this reason, I prefer the earlier prescription, and from hereon I denote the bound implied by (3.11) (or by (3.16)) as $\tilde{W}_{\text{W}}$, and refer to it as the Wiener lower bound.

(b) Hashin-Shtrikman lower bound

In this subsection, I apply the structure developed in the previous subsection to obtain lower bounds of the Hashin-Shtrikman type for the class of nonlinear isotropic composites with prescribed volume fractions.

We begin by noting that the effective dielectric tensor of a linear isotropic heterogeneous material is isotropic (i.e. $\tilde{\epsilon}_0 = \tilde{\epsilon}_0 I$, where I is the identity tensor in $\mathbb{R}^d$, with $d = 2$ or 3 representing the dimension of the space). Then, the Hashin-Shtrikman (H-S) lower bound $\tilde{\epsilon}$ for the effective dielectric constant $\tilde{\epsilon}_0$, satisfying $\tilde{\epsilon}_0 \geq \tilde{\epsilon}$, is given by the expression (see, for instance, Kohn & Milton 1988)

$$\tilde{\epsilon} = \left(\sum_{r=1}^n \frac{c^{(r)}}{\epsilon_0^{(r)}} + (d-1)\epsilon \right)^{-1} - (d-1)\epsilon, \quad (3.17)$$

where $\epsilon = \inf_{\omega} \{ \epsilon_0^{(r)} \}$. This result may be rewritten by means of the identity (3.7), shifted by $(d-1)\epsilon$, in the form

$$\tilde{\epsilon} = \inf_{\omega^{(r)} \geq 0} \left\{ \sum_{r=1}^n c^{(r)} (1 - \omega^{(r)})^2 + (d-1)\epsilon (\omega^{(r)})^2 \right\}. \quad (3.18)$$

Next, we observe that the effective energy functions $\tilde{W}$ of the class of nonlinear composites with overall isotropy may be estimated once again from relation (3.2) of Corollary 3.1, where $\tilde{W}_0$ now represents the effective energy functions of the class of linear comparison composites with phases of dielectric constants $\epsilon_0^{(r)}$ in prescribed volume fractions $c^{(r)}$. Further, the distribution of the phases in the linear comparison composite is such that the nonlinear compositing must be isotropic; therefore, the distribution of the phases in the corresponding linear comparison composites must also be isotropic. I emphasize that, while not all microstructures that are isotropic for linear composites are also isotropic in the nonlinear context (for example, whereas a linear periodic composite with cubic symmetry is isotropic, the corresponding nonlinear periodic composite is not necessarily isotropic), nonlinear isotropic microstructures must also be isotropic in the linear context (since linear is a special case of nonlinear). Thus it follows that a lower bound for the effective energy functions of the class of linear, isotropic comparison composites is also a lower bound

for the subclass of linear comparison composites with 'nonlinearly isotropic' microstructure.

Therefore, replacing $\bar{W}_0$ in (3.2) by the lower bound specified by (3.18) induces a lower bound for the class of nonlinear isotropic composites. The result is

$$\begin{aligned}\bar{W}(\mathbf{E}) &\geq \sup_{\substack{\omega^{(n)} > 0 \\ \epsilon^{(n)} > 0}} \left\{ \inf_{\substack{\omega^{(n)} > 0 \\ \epsilon^{(n)} > 0}} \left\{ \sum_{n=1}^{\infty} c^{(n)} \left[\frac{1}{2} (\epsilon^{(n)} (1 - \omega^{(n)})^2 + (d-1) \epsilon_-(\omega^{(n)})^2) \mathbf{E}^2 - v^{(n)}(\epsilon_0^{(n)}) \right] \right\} \right\} \\ &= \inf_{\substack{\omega^{(n)} > 0 \\ \epsilon^{(n)} > 0}} \left\{ \sup_{\substack{\omega^{(n)} > 0 \\ \epsilon^{(n)} > 0}} \left\{ \sum_{n=1}^{\infty} c^{(n)} \left[\frac{1}{2} (\epsilon^{(n)} (1 - \omega^{(n)})^2 + (d-1) \epsilon_-(\omega^{(n)})^2) \mathbf{E}^2 - v^{(n)}(\epsilon_0^{(n)}) \right] \right\} \right\},\end{aligned}\quad (3.19)$$

where, once again, I have made use of the saddle point theorem to interchange the supremum and infimum operations. Then, making use of (3.10) and remarking that the minimum $\epsilon_0^{(n)}$ (i.e. ϵ) depends on the intensity of the applied electric field $\mathbf{E}$, and may occur in any given phase, I conclude that

$$\begin{aligned}\bar{W}(\mathbf{E}) &\geq \min_{\substack{\omega^{(n)} > 0 \\ \epsilon^{(n)} > 0}} \left\{ \inf_{\substack{\omega^{(n)} > 0 \\ \epsilon^{(n)} > 0}} \left\{ \sum_{n=1}^{\infty} c^{(n)} \phi^{(n)} [(1 - \omega^{(n)}) \mathbf{E}] \right. \right. \\ &\quad \left. \left. + c^{(n)} \phi^{(n)} \left[\sqrt{(1 - \omega^{(n)})^2 + (d-1) \frac{1}{\epsilon_0^{(n)}} \sum_{i=1}^n c^{(i)} (\omega^{(i)})^2} \mathbf{E}} \right] \right\} \right\}.\end{aligned}\quad (3.20)$$

I refer to the bound implied by this relation as the H-S lower bound for nonlinear isotropic composites with isotropic phases in prescribed volume fractions, and denote it by $\bar{W}_{\text{HS}}$. We remark that a dual version of this result, with very similar form, may also be obtained from the complementary-energy version of the variational principle (Corollary 3.2). Note further that for the special case of two-phase composites, the nonlinear H-S lower bound reduces to

$$\bar{W}_{\text{HS}}(\mathbf{E}) = \min_{\substack{\omega > 0 \\ \epsilon > 0}} \left\{ \inf_{\substack{\omega > 0 \\ \epsilon > 0}} \left\{ \sum_{n=1}^{\infty} c^{(n)} \phi^{(n)} [(1 - \epsilon^{(n)} \omega) \mathbf{E}] + c^{(2)} \phi^{(2)} \left[\sqrt{(1 + \epsilon^{(1)} \omega)^2 + (d-1) \epsilon^{(1)} \omega^2} \mathbf{E} \right] \right\} \right\},\quad (3.21)$$

where the absolute minimum of the two infimum problems normally depends on $\mathbf{E}$.

Note that lower bounds of the Hashin-Shtrikman type for nonlinear dielectrics have been obtained earlier by Willis (1986), using the general method of Talbot & Willis (1985). This method, which is a generalization of the Hashin-Shtrikman (1962) variational principles for nonlinear problems, makes use of a linear homogeneous comparison material (unlike the heterogeneous comparison material of the new method) to obtain results that, at least in some cases, agree with the predictions for the bounds obtained via the new method proposed in this work. According to the work of Willis (1991), in a different physical context (nonlinear infinitesimal elasticity), the key to recovering the bounds predicted by the new method from the Talbot-Willis method centres around an optimal choice of the homogeneous comparison material (in the Talbot-Willis method). Thus Willis finds that the improvement in the bounds noted in Ponte Castañeda (1991a) (using essentially the new method) with reference to the bounds determined by Ponte Castañeda & Willis (1988) (using the Talbot-Willis method), for some special types of nonlinear composite materials, was a consequence of a non-optimal choice for the comparison

material in the earlier work. It is anticipated that analogous results will hold for nonlinear dielectrics. However, I emphasize that the form of the bounds given in this work is different (and simpler) from the form of the bounds given by Willis (1986) (see also Talbot & Willis 1987). In particular, I find that, for the special case of two-phase nonlinear composites, the bounds given in this work involve only one optimization, whereas the bounds given by Willis (1986) involve, in general, two optimizations (including the optimization of the homogeneous comparison material). Further comparison between the two methods is given by Willis (1991) and by Ponte Castañeda (1992), but one distinct advantage of the new method utilizing the linear heterogeneous comparison material is that it can be used in conjunction with linear bounds and estimates, other than Hashin-Shtrikman bounds, to yield corresponding nonlinear bounds and estimates (assuming, of course, that such linear bounds and estimates are available). As an illustration of this feature of the new method, in the next subsection, I derive lower bounds of the Beran type for two-phase, nonlinear isotropic composites. Later, in §5c we also give an application to self-consistent estimates.

(c) Beran lower bound

In the context of linear, isotropic dielectric composites, bounds that are more restrictive than the Hashin-Shtrikman bounds, and contain additional microstructural information in the form of three-point correlation functions have been proposed by Beran (1965). For the case of two-phase systems, Milton (1981) has proposed a simple form of the Beran bounds that depends on the volume fraction of the phases, and on only one additional geometric parameter (that can be obtained from the three-point correlation functions). Milton's form for the lower bound on the effective dielectric constant $\hat{\epsilon}_0$ is given by

$$\hat{\epsilon} = \left(\sum_{i=1}^2 \frac{c^{(i)}}{\epsilon_0^{(i)}} + (d-1) \hat{\epsilon}_- \right)^{-1} - (d-1) \hat{\epsilon}_-, \quad (3.22)$$

which is identical in form to (3.17), except that $\hat{\epsilon}$ must be replaced by

$$\hat{\epsilon} = \left(\sum_{i=1}^2 \frac{\xi^{(i)}}{\epsilon_0^{(i)}} \right)^{-1}.$$

Thus this expression depends on the dielectric constants $\epsilon_0^{(1)}$ and $\epsilon_0^{(2)}$, the volume fractions $c^{(1)}$ and $c^{(2)}$, and the third-order geometric parameters $\xi^{(1)}$ and $\xi^{(2)} = 1 - \xi^{(1)}$, both lying in the interval [0, 1].

Substitution of this result into the lower bound approximation (3.2) of Corollary 3.1, and following a procedure very similar to that used for the Hashin-Shtrikman bound (making use of identity (3.7) twice; once for expression (3.22) and again for the expression of $\hat{\epsilon}$), we arrive at the following lower bound for the nonlinear energy function, namely,

$$\begin{aligned}\bar{W}_H(\mathbf{E}) &= \inf_{\omega, \gamma} \left\{ c^{(1)} \phi^{(1)} \left[\sqrt{(1 - c^{(2)} \omega)^2 + (d-1) c^{(2)} \xi^{(1)} \omega^2} (1 - \xi^{(2)} \gamma)^2 \mathbf{E} \right] + \dots \right. \\ &\quad \left. + c^{(2)} \phi^{(2)} \left[\sqrt{(1 + c^{(1)} \omega)^2 + (d-1) c^{(1)} \xi^{(2)} \omega^2} (1 + \xi^{(1)} \gamma)^2 \mathbf{E} \right] \right\}.\end{aligned}\quad (3.23)$$

We remark that the corresponding nonlinear H-S lower bound (3.21) follows immediately from this result by choosing either $\xi^{(1)} = 0$ or $\xi^{(2)} = 1$, whichever yields the lowest value (note that the infimum problem over γ becomes trivial in either case). This is completely analogous to the corresponding result for linear two-phase

composites. Finally, note that a dual version of this result, with slightly simpler form (involving only one optimization), may also be obtained by application of Corollary 3.2.

(d) Upper bounds and estimates

The determination of upper bounds for the effective energy functions $\tilde{W}$ of classes of nonlinear composites is intrinsically harder than the determination of the corresponding lower bounds. This is because approximations of the type (3.2) clearly do not work in this case; instead, we must resort to the exact versions of the variational principles. As we see below, it is possible to make use of the exact version of the variational principles to obtain the Wiener upper bound, but I was not able to make use of the exact version of the variational principles to obtain upper bounds of the Hashin-Shtrikman and Beran types. It is interesting to note that the method of Talbot & Willis (1985) exhibits the same limitation in determining nonlinear H-S upper bounds, if attention is restricted to linear comparison materials. Because of this difficulty, I provide instead 'upper estimates', or rather lower estimates for the upper bound, of the Hashin-Shtrikman and Beran types. The rationale behind this is that, in practice, the bounds are often used as estimates for the behaviour of the 'weakest' and 'strongest' composite possible with the given microstructural information. In this less rigorous interpretation, it is probably as useful to have a lower estimate for the 'weakest' material, as to have a lower estimate for the 'strongest' material, provided that the estimates are not too weak. In §§4 and 5, we pursue these ideas further.

The derivation of the Wiener upper bound is made possible by the corresponding upper bound for linear composites with arbitrarily variable dielectric coefficient $\epsilon_0(\mathbf{x})$. This bound may be expressed in the form

$$\tilde{W}_0(\mathcal{E}) \leq \frac{1}{2} \left(\int_U \epsilon_0(\mathbf{x}) d\mathbf{x} \right) \mathcal{E}^2, \quad (3.24)$$

where $\tilde{W}_0$ is the effective energy function of the linear composite. Then, application of (3.24) to (2.24) of Theorem 2.1 leads to

$$\begin{aligned} \tilde{W}(\mathcal{E}) &\leq \sup_{\epsilon_0(\mathbf{x}) \geq 0} \left\{ \frac{1}{2} \left(\int_U \epsilon_0(\mathbf{x}) d\mathbf{x} \right) \mathcal{E}^2 - \int_U r(\mathbf{x}, \epsilon_0(\mathbf{x})) d\mathbf{x} \right\} \\ &= \int_U \sup_{\epsilon_0 \geq 0} \left\{ \frac{1}{2} \epsilon_0 \mathcal{E}^2 - r(\mathbf{x}, \epsilon_0) \right\} d\mathbf{x} \\ &= \int_U \phi(\mathbf{x}, \mathcal{E}) d\mathbf{x}, \end{aligned} \quad (3.25)$$

which, via (3.1), leads in turn to the nonlinear Wiener upper bound ($\tilde{W}_{W+}$)

$$\tilde{W}(\mathcal{E}) \leq \sum_{r=1}^n r^{(r)} \phi^{(r)}(\mathcal{E}). \quad (3.26)$$

This result may, of course, be obtained directly from the classical minimum energy principle; inclusion of the present alternative derivation in this paper is made only to emphasize that the exact version of the variational principles of §2c also lead (at least in principle, and sometimes in practice) to upper bounds. Finally, we remark that a dual alternative expression analogous to expression (3.11) for the Wiener lower bound is also possible, if use is made of Theorem 2.2 instead of Theorem 2.1.

Phil Trans R Soc Lond A (1992)

A similar approach may be attempted for the nonlinear H-S upper bound. However, I was unable to obtain an explicit result analogous to (3.26). The reason is essentially that the evaluation of the pertinent term involving $\epsilon_s = \max_{\mathbf{x}} \{\epsilon_0(\mathbf{x})\}$ in the appropriate expression of the H-S upper bound for the linear isotropic comparison composite with continuously varying microstructure is extremely difficult. I do not pursue this point further, and proceed with the determination of estimates for the H-S upper bound, or 'upper estimates'. As discussed earlier, this is accomplished by application of approximation (3.2) of Corollary 3.1 to the H-S upper bounds for the linear comparison composite. The upper bound for the effective energy function of the linear comparison composite may be given in terms of the upper bound for its effective dielectric constant, namely,

$$\tilde{\epsilon}_s = \left(\sum_{r=1}^n \frac{r^{(r)}}{\epsilon_0^{(r)}} + (d-1)\epsilon_s \right)^{-1} - (d-1)\epsilon_s, \quad (3.27)$$

where $\epsilon_s = \sup_{\mathbf{x}} \{\epsilon_0(\mathbf{x})\}$.

It is evident that the procedure of §3b, utilizing the lower bound (3.17) for the linear comparison composite to obtain a lower bound for the nonlinear composite, may be repeated making use of (3.27) instead of (3.17) to obtain an 'upper estimate' for the nonlinear composite. The result is the same as expressions (3.20) and (3.21) for the n -phase and two-phase nonlinear composites, respectively, with the difference that the outermost minimum operations must now be replaced by maximum operations. I denote this result by $\tilde{W}_{HSC}$, and refer to it as the H-S 'upper estimate' for $\tilde{W}$. In §4, we see that $\tilde{W}_{HSC}$ is not, in general, an upper bound for $\tilde{W}$. This is accomplished by computing the exact effective energy function of a specific nonlinear composite, and noting that it exceeds $\tilde{W}_{HSC}$.

Finally, note that the same comments that we have just made about the H-S upper bounds also apply to the Beran upper bounds. Therefore, I provide here only an 'upper estimate' of the Beran type. Such an estimate is made possible by Milton's expression for the upper bound of the effective dielectric constant of the linear comparison composite, $\tilde{\epsilon}_l$, which is given by expression (3.22), but with $\hat{\epsilon}$ replaced by

$$\hat{\epsilon}_s = \sum_{r=1}^n r^{(r)} \epsilon_0^{(r)}.$$

Then, the corresponding form for the 'upper estimate' (not in general an upper bound, for the same reason as for the Hashin-Shtrikman upper estimate) of the class of nonlinear isotropic composites with prescribed volume fractions and third order statistical information (in the form of $\zeta^{(r)}$) is given by

$$\begin{aligned} \tilde{W}_{H+}(\mathcal{E}) &= \inf_{\omega} \{ c^{(0)} \phi^{(0)}(\chi [(1 - c^{(2)}\omega)^2 + (d-1)c^{(2)}\zeta^{(2)}\omega^2] \mathcal{E}) + \\ &\quad \dots + c^{(2)} \phi^{(2)}(\chi [(1 + c^{(0)}\omega)^2 + (d-1)c^{(0)}\zeta^{(0)}\omega^2] \mathcal{E}) \}. \end{aligned} \quad (3.28)$$

This bound also reduces to the corresponding Hashin-Shtrikman 'upper estimate', by letting either $\zeta^{(0)} = 0$, or $\zeta^{(2)} = 1$, whichever yields the highest value.

4. Exact estimates

In the previous section, I made use of the variational principles of §2c (Theorems 2.1 and 2.2) to obtain bounds on the effective behaviour of nonlinear composites. I first studied the case of generally anisotropic composites and discovered that the

Phil Trans R Soc Lond A (1992)

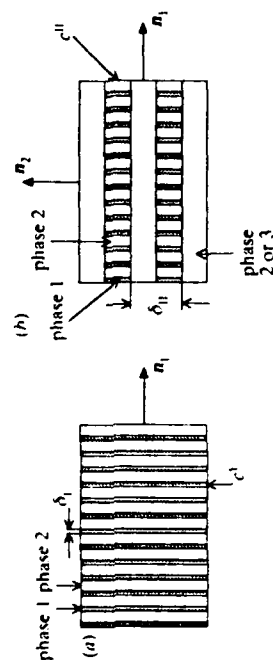


Figure 1. (a) A rank-I laminate and (b) a rank-II laminate.

well-known Wiener bounds for nonlinear composites could be recovered from these variational principles. I then considered the special, but important, case of isotropic composites, and was able to determine new lower bounds, but only 'upper estimates', in general, for the effective energy functions of these nonlinear composites. This was accomplished by making use of the Hashin-Shtrikman and Beran bounds for linear, isotropic comparison materials, with the same distribution of phases as the nonlinear composites, in the variational principles. The question then arises as to the quality of the lower bounds, and also, as to whether the 'upper estimates' could in fact be rigorous bounds. In this section, I attempt to answer these and other related questions by analysing the effective properties of sequentially laminated materials. This class of materials, although of little practical value, has proved to be of great theoretical value in the study of linear composites. For instance, in the particular case of two-phase linear dielectric composites, it has been shown by Tartar (1985) and Lurie & Cherkov (1984, 1986) that sequentially laminated materials can be constructed attaining the corresponding linear Hashin-Shtrikman bounds. Here, I attempt a similar study for nonlinear sequentially laminated materials. I find that the variational principles of §2c are specially suited to the study of this class of nonlinear materials.

A sequentially laminated material (or laminate, for short) is an iterative construction obtained by layering laminated materials (which in turn have been obtained from lower-order lamination procedures) with other laminated materials, or directly with the homogeneous phases that make up the composite, in such a way as to produce hierarchical microstructures of increasing complexity. The 'rank' of the laminate refers to the number of layering operations required to reach the final iterated microstructure. Thus, as shown in figure 1a, a rank-I (I use roman numerals to denote rank) laminate is obtained by mixing layers of two homogeneous phases (1 and 2) to obtain a simple laminate with layering direction n_1 . A rank-II laminate (see figure 1b) is obtained by layering the rank-I laminate with a third phase (3), or alternatively with one of the original phases (say 2), in a different layering direction n_2 . In general, n_1 and n_2 can take on any orientation, but in figure 1b they are depicted as being orthogonal. Higher rank laminates are obtained by iterating this procedure (see Milton 1986). (One important observation in connection with this procedure is that the length scale of the embedded laminates is assumed to be small compared with the length scale of the embedding laminates (for example, in figure 1b, $\delta_1 \ll \delta_{II} \ll 1$). This assumption derives from the fact that the effective properties of simple laminates can be computed exactly, and hence, by treating the iterated laminate as a simple laminate with the embedded laminate replaced by a

homogeneous material with the effective properties of the embedded laminate, the effective properties of the iterated laminate can also be computed exactly. An equivalent way of stating this result is that, under the assumption of widely separated length scales for the different layers composing the iterated laminate, the fields will be essentially constant within each elemental layer, provided that the boundary conditions applied to the laminate are uniform. This feature, greatly simplifies the computation of effective properties, thereby making sequentially laminated materials very useful constructions.

In this and in the next two paragraphs, I review some of the important results concerning linear laminates. I first note that the effective energy function of a simple laminate (which, as mentioned previously, can be computed exactly) lies within and attains (for specific orientations of the applied fields) the Wiener bounds. Thus, at least in the linear case, the Wiener bounds on the effective energy-function of arbitrarily anisotropic linear composites are known to be sharp (i.e. they can be attained by specific microstructures). In §4a, I demonstrate that the same result holds for the nonlinear Wiener bounds.

In the context of two-phase linear dielectrics, it is known that only iterated laminates of rank greater than, or equal to, the dimension of the underlying physical space ($d = 2$, or 3) can have isotropic properties. These isotropic laminates are obtained by choosing the relative volume fractions and the layering directions of each of the embedded laminates in such a way that the effective dielectric tensor of the iterated laminate is isotropic, while the absolute volume fractions of the constituent phases remain fixed. For details, the reader is referred to Tartar (1985), where it is additionally demonstrated that the result of this calculation is independent of the rank of the iterated laminate (as long as it is greater than d), and equal to the corresponding Hashin-Shtrikman bounds. The lower bound is obtained by letting the phase with the highest dielectric constant occupy one of the two phases in the (innermost) rank-I laminate, and by choosing the other phase (with the lower dielectric constant) to play the role of the homogeneous phase at each of the other layering operations. This yields a composite material with the phase with the highest dielectric constant serving as the inclusion phase, and the other phase acting as the matrix phase (for a picture of this construction in two dimensions, let phase 3 be the same as phase 2 in figure 1b). Intuitively, such a construction should produce a material with a low effective dielectric constant. In fact, it yields a material with the lowest possible dielectric constant since, as mentioned above, it attains the H-S lower bound. Conversely, the upper bound for the effective dielectric constant is obtained by exchanging the roles of the two phases in the above construction.

One source of potential concern in connection with sequentially laminated materials is the fact that the resulting inclusions are 'flat', whereas in practice the inclusions are often equiaxed. However, this concern can be easily dispelled by noting that iterated laminates can be used to model arbitrarily close the properties of any two-phase microstructure (Milton 1986). For instance, the so-called composite-spheres (concentric spheres) model of Hashin-Shtrikman (1962) (with inclusions of spherical shape) possesses exactly the same effective properties as an isotropic iterated laminate with the same volume fractions (i.e. they both saturate the Hashin-Shtrikman bounds for isotropic microstructures). Similarly, Tartar (1985) proposed a confocal ellipsoidal microstructure to demonstrate the optimality of the anisotropic Hashin-Shtrikman bounds developed by Murat & Tartar (1985) and Lurie & Cherkov (1984, 1986). Thus, a one-to-one correspondence may be

established between the confocal ellipsoidal and sequentially laminated microstructures in this case also. It may initially seem that the concentric spheres (or confocal ellipsoids) microgeometry is to be preferred over the iterated laminate construction, but in fact the sequentially laminated microgeometry has several advantages over the composite-spheres microgeometry. First, the sequentially laminated microstructure involves a finite number of length scales in contrast with the composite-spheres microstructure which involves an infinite number of length scales (because the composite spheres must be chosen to cover all sizes to fill space). Second, the laminated microstructure, unlike the composite-spheres microstructure, generalizes to other more complex systems such as elasticity, and to nonlinear systems, as we see in §4b.

As discussed previously, the main advantage of sequentially laminated composites is that, when subject to uniform boundary conditions, the fields are piecewise constant within the composite (whether the phases in the composite are linear or nonlinear), except in small boundary layer regions at the interfaces separating laminates of different rank. However, the 'widely separated length scales' hypothesis makes the effect of these boundary layer regions negligible on the effective behaviour of the laminate. For example, the assumption that $\delta_1 \ll \delta_{11} \ll 1$, in the two-dimensional microstructure of figure 1b, leads to different constant fields in each of the regions labelled 1, 2 and 3 (even if there are only two phases). For a rigorous treatment of this issue in the linear case, the reader is referred to Tartar (1985). Because of this feature of sequentially laminated materials, it is useful to state the following corollaries of Theorems 2.1 and 2.2.

Corollary 4.1. Suppose that we have an n -phase nonlinear composite with local energy function w characterized by (3.1), additionally satisfying the hypotheses of Theorem 2.1, such that the distribution of fields within the composite is piecewise constant (with perhaps different constants in different portions of the same phase). We assume that the distribution of fields is given by the characteristic functions $\tilde{\chi}^{(s)}$ ($s = 1, \dots, m$), such that, for example,

$$E(x) = \sum_{s=1}^m \tilde{\chi}^{(s)}(x) E^{(s)},$$

with each $E^{(s)}$ constant. In general, the characteristic functions $\tilde{\chi}^{(s)}$ and the characteristic functions defining the microstructure of the composite $\chi^{(r)}$ ($r = 1, \dots, n$) are different ($m \geq n$). Then, the effective energy function of the nonlinear composite may be expressed in the form

$$\tilde{W}(\bar{E}) = \sup_{\tilde{\chi}_0 \geq 0} \left\{ \tilde{W}_0(\bar{E}) - \sum_{s=1}^m \tilde{\rho}^{(s)} \rho^{(s)}(\tilde{\epsilon}_0^{(s)}) \right\}, \quad (4.1)$$

where $\tilde{W}_0$ is the effective energy function of a linear comparison composite with m phases of dielectric constant $\tilde{\epsilon}_0^{(s)}$ in volume fractions $\tilde{\rho}^{(s)}$, such that the dielectric coefficient of the comparison composite is given by

$$\tilde{\epsilon}_0(x) = \sum_{s=1}^m \tilde{\chi}^{(s)}(x) \tilde{\epsilon}_0^{(s)},$$

and where the functions $\rho^{(s)}$ are obtained by specializing relation (2.23) to the s th region.

For example, in the rank II laminate of figure 1b, the linear comparison composite is made up of three phases, denoted 1, 2 and 3, even if the corresponding nonlinear composite is made up of only two phases (nonlinear phases 2 and 3 are equal).

Phil Trans R Soc Lond A (1992)

Proof. This result is obtained directly from Theorem 2.1 by noticing that, because the fields are piecewise constant with distribution given by characteristic functions $\chi^{(r)}$, the piecewise constant distribution of comparison dielectric coefficients (with the same characteristic functions) is the exact solution for $\epsilon_0(x)$ in the variational principle (2.24). To see this, note that, within a given phase (characterized by some $\chi^{(r)}$), if E is constant (say $E^{(r)}$) over some subregion of the given phase (characterized by $\tilde{\chi}^{(s)}$) then the optimal $\epsilon_0(x)$ in relation (2.24) for the local energy function w is constant over that subregion (say $\tilde{\epsilon}_0^{(s)}$). Therefore the integrals in (2.24) may be evaluated exactly, and expression (4.1) results.

Corollary 4.2. Suppose that we have an n -phase nonlinear composite with local complementary energy function w^* , characterized by the dual version of (3.1), and additionally satisfying the hypotheses of Theorem 2.2, such that the distribution of fields within the composite is piecewise constant (with perhaps a different constant in different portions of the same phase). We assume that the distribution of fields is given by the characteristic functions $\tilde{\chi}^{(s)}$ ($s = 1, \dots, m$) ($m \geq n$), such that, for example,

$$D(x) = \sum_{s=1}^m \tilde{\chi}^{(s)}(x) D^{(s)},$$

with $D^{(s)}$ constant. Then, the effective complementary energy function of the nonlinear composite may be expressed in the form

$$\tilde{V}(\bar{D}) = \inf_{\tilde{\epsilon}_0 \geq 0} \left\{ \tilde{V}_0(\bar{D}) + \sum_{s=1}^m \tilde{\rho}^{(s)} \rho^{(s)}(\tilde{\epsilon}_0^{(s)}) \right\}, \quad (4.2)$$

where $\tilde{V}_0$ is the effective complementary energy function of a linear comparison composite with m phases of dielectric constant $\tilde{\epsilon}_0^{(s)}$ in volume fractions $\tilde{\rho}^{(s)}$, such that the dielectric coefficient of the comparison composite is given by

$$\tilde{\epsilon}_0(x) = \sum_{s=1}^m \tilde{\chi}^{(s)}(x) \tilde{\epsilon}_0^{(s)},$$

and where the functions $\rho^{(s)}$ are obtained by specializing relation (2.23) to the s th region.

Proof. The demonstration of this result is analogous to that of the previous result.

(a) Simple laminates

In this subsection, I determine the effective energy functions of simple (one layering direction) nonlinear laminates by applying the well known corresponding results for linear laminates to the above corollaries of the variational principles of §2c. This calculation has been discussed previously by Ponte Castañeda (1992) for the case of two phase (rank I) laminates; here, I investigate n phase laminates made up of nonlinear isotropic phases with potentials $\phi^{(r)}$ in volume fractions $\epsilon^{(r)}$, distributed randomly in layers perpendicular to a fixed orientation $\mathbf{n}$. The corresponding linear comparison laminate with dielectric constants $\tilde{\epsilon}_0^{(s)}$ in volume fractions $\tilde{\rho}^{(s)}$ has anisotropic effective dielectric tensor

$$\tilde{\epsilon}_0 = \left(\sum_{r=1}^n \epsilon^{(r)} \tilde{\epsilon}_0^{(r)} \right) (I - \mathbf{n} \otimes \mathbf{n}) + \left(\sum_{r=1}^n \frac{\epsilon^{(r)}}{\tilde{\epsilon}_0^{(r)}} \right) \mathbf{n} \otimes \mathbf{n}, \quad (4.3)$$

where $\mathbf{n}$ is the lamination direction, and $\otimes$ denotes the tensor product of two vectors.

Phil Trans R Soc Lond A (1992)

Thus, according to (4.1), the effective energy function of the nonlinear laminate may be computed from the relation

$$\tilde{W}(\mathbf{E}) = \sup_{\epsilon_0^{(n)} > 0} \left\{ \frac{1}{2} (\tilde{\mathbf{E}}_0 \cdot \mathbf{E}) \cdot \mathbf{E} - \sum_{r=1}^n \epsilon_0^{(r), (n)} (\epsilon_0^{(n)}) \right\}, \quad (4.4)$$

where $\tilde{\mathbf{E}}_0$ is given by (4.1), and where I have used the fact that the fields for a laminated composite material (even if it is nonlinear), subject to uniform boundary conditions, are constant within each phase (i.e. $\tilde{\chi}^{(n)} = \chi^{(n)}$). Then, making use of the identity (3.7), and interchanging the sup and the inf by invoking the appropriate version of the saddle point theorem, I arrive at

$$\tilde{W}(\mathbf{E}) = \inf_{\epsilon_0^{(n)} > 0} \left\{ \sum_{r=1}^n \epsilon_0^{(r)} \sup_{\epsilon_0^{(n)} > 0} \left\{ \frac{1}{2} \epsilon_0^{(n)} [\mathbf{E}^2 - (\mathbf{E} \cdot \mathbf{n})^2 + (1 - \omega^{(n)})^2 (\mathbf{E} \cdot \mathbf{n})^2] - \epsilon_0^{(n)} (\epsilon_0^{(n)}) \right\} \right\}, \quad (4.5)$$

which by means of (3.10) finally reduces to

$$\tilde{W}(\mathbf{E}) = \inf_{\epsilon_0^{(n)} > 0} \left\{ \sum_{r=1}^n \epsilon_0^{(r)} \phi^{(n)} \left[\lambda \left\{ \mathbf{E}^2 - (\mathbf{E} \cdot \mathbf{n})^2 + (1 - \omega^{(n)})^2 (\mathbf{E} \cdot \mathbf{n})^2 \right\} \right] \right\}. \quad (4.6)$$

It is not difficult to see that the maximum value of $\tilde{W}$ in the above expression is obtained when $\mathbf{E}$ is perpendicular to $\mathbf{n}$ yielding the Wiener upper bound $\tilde{W}_{w+}$, as given by (3.28). Conversely, the minimum value of $\tilde{W}$ is obtained whenever $\mathbf{E}$ is parallel to $\mathbf{n}$ yielding the Wiener lower bound $\tilde{W}_{w-}$, as given by (3.11).

This demonstrates that the Wiener bounds on the effective energy function are sharp within the class of nonlinear anisotropic composite materials with prescribed volume fractions, and that these bounds are attained by simple rank-I laminates. Thus, for anisotropic composites, the linear and nonlinear theories are similar. As we see in the next subsection, the same is not true for nonlinear isotropic composites.

(b) Iterated laminates

To motivate the study of nonlinear iterated laminates, I first consider the corresponding linear problem in some detail. For the most part, we will restrict our attention to the case of two-phase composites. It is well known that rank- d laminates with orthogonal layering directions attain the isotropic Hashin-Shtrikman bounds for the effective dielectric constant of two-phase materials (in d dimensions). For example, a two-phase, orthogonal rank-II laminate in two dimensions is depicted in figure 1b. We note that phase 1 occupies relative volume fraction ϵ^I in the (embedded) rank-I laminate and overall volume fraction $\epsilon^{(I)} = \epsilon^I \epsilon^{II}$ in the composite, where ϵ^{II} is the volume fraction occupied by the rank-I laminate within the rank-II laminate. The rest of the composite is made up of phase 2 (we are letting phase 3 be equal to phase 2 in figure 1b). Its effective dielectric tensor is obtained by first computing the effective dielectric tensor of the embedded rank-I laminate with layering orientation $\mathbf{n}_1$, and then using this result in the computation of the effective dielectric tensor of the rank-II laminate with layering direction $\mathbf{n}_2$ (where $\mathbf{n}_2$ is orthogonal to $\mathbf{n}_1$). Thus the dielectric constants in the $\mathbf{n}_1$ and $\mathbf{n}_2$ (principal) directions of the rank-II laminate are, respectively,

$$\tilde{\epsilon}_1^{II} = \epsilon^{II} \tilde{\epsilon}_1^I + (1 - \epsilon^{II}) \epsilon^{(2)} \quad \text{and} \quad \tilde{\epsilon}_2^{II} = [\epsilon^{II} (\tilde{\epsilon}_2^I)^{-1} + (1 - \epsilon^{II}) (\epsilon^{(2)})^{-1}]^{-1}, \quad (4.7)$$

where $\tilde{\epsilon}_1^I$ and $\tilde{\epsilon}_2^I$ are the effective dielectric constants of the rank-I laminate in the $\mathbf{n}_1$

and $\mathbf{n}_2$ directions. Then, the condition for isotropy is obtained by choosing ϵ^I (ϵ^{II}) in turn given by $\epsilon^{(I)} = \epsilon^I \epsilon^{II}$ such that $\tilde{\epsilon}_1^{II} = \tilde{\epsilon}_2^{II} = \tilde{\epsilon}$. The result of this calculation for the effective dielectric constant of the rank-II isotropic laminate is

$$\tilde{\epsilon} = \left(\sum_{r=1}^2 \frac{\epsilon^{(r)}}{\epsilon_0^{(r)} + \epsilon^{(2)}} \right)^{-1} - \epsilon^{(2)}. \quad (4.8)$$

This result is in agreement with the H-S lower bound given by (3.17) for $d=2$, if we assume that $\epsilon^{(I)} > \epsilon^{(2)}$. Otherwise, the above expression would yield the corresponding H-S upper bound. Thus, the isotropic laminate with the largest dielectric constant as the inclusion phase (and the smallest as the matrix phase) leads to the lowest effective dielectric constant for the composite, and vice versa.

Clearly, this procedure generalizes to three dimensions by considering a rank-III laminate, obtained by layering the rank-II laminate in volume fraction ϵ^{III} again with phase 2 in direction $\mathbf{n}_3$ (orthogonal to $\mathbf{n}_1$ and $\mathbf{n}_2$). Then, the inclusion phase (I) occupies volume fractions ϵ^I and $\epsilon^I \epsilon^{II}$ in the embedded rank-I and rank-II laminates, respectively, and volume fraction $\epsilon^{(I)} = \epsilon^I \epsilon^{II} \epsilon^{III}$ in the rank-III laminate. The condition of isotropy in this case leads to two equations to be solved for ϵ^I and ϵ^{II} . Again, the result of this calculation is in agreement with the H-S bounds (3.17) and (3.27). I emphasize that, within the class of linear, isotropic iterated laminates, there exist a large (in fact, infinite) number of laminates of different ranks attaining the H-S bounds. The rank- d laminates with orthogonal layering directions, discussed above, are the simplest such composites.

The above discussion suggests that the same identification may be possible between the nonlinear H-S bounds given in §3 and nonlinear rank- d laminates. However, this identification fails even in the simplest non-trivial case ($d=2$). To see this, note that the effective energy function of the two phase, nonlinear rank-II laminate with the same microstructure as the corresponding linear, isotropic rank-II laminate may be expressed in the form

$$\tilde{W}_L(\mathbf{E}) = \sup_{\epsilon_0^{(n)}, \epsilon_0^{(2)} > 0} \{ \tilde{W}_0^{(2)}(\mathbf{E}) - \epsilon_0^{(I), (n)} (\epsilon_0^{(I)}) - (\epsilon^{II} - \epsilon_0^{(I)}) \epsilon^{(2)} (\epsilon_0^{(2)}) - (1 - \epsilon^{II}) \epsilon^{(2)} (\epsilon_0^{(2)}) \}, \quad (4.9)$$

where $\tilde{W}_0^{(2)}(\mathbf{E})$ is the effective energy function of a three-phase, linear rank-II laminate (see figure 1b), with comparison dielectric constants $\epsilon_0^{(1)}, \epsilon_0^{(2)}, \epsilon_0^{(3)}$, and where ϵ^{II} has been specified previously. Note that $\tilde{W}_0^{(2)}(\mathbf{E})$ is not, in general, isotropic (unless $\epsilon_0^{(2)} = \epsilon_0^{(3)}$), and therefore $\tilde{W}_L$ is also not isotropic. The reason in this case for the three distinct dielectric constants for the linear comparison material, even though we are dealing with a two phase nonlinear material, is again that for the nonlinear rank-II laminate the fields will be different constants in phase 2, depending on whether we are dealing with the portion of phase 2 that is embedded within the rank-I laminate (in volume fraction $\epsilon^I \epsilon^{II} - \epsilon_0^{(I)}$), or we are referring to the portion of phase 2 in the rank-II laminate that is not part of the rank-I laminate (the volume fraction of this portion of phase 2 is $1 - \epsilon^{II}$). Thus to obtain an exact result for the effective energy function of the nonlinear laminate, a three-dimensional optimization is required (see Corollary 4.1).

On the other hand, if instead of distinguishing between the two portions of the nonlinear phase 2 subject to different constant fields, and carrying out the optimization over the three independent comparison dielectric constants, we carry out the optimization over two independent variables $\epsilon_0^{(1)}, \epsilon_0^{(2)}$ (as suggested by (3.2) of

Corollary 3.1), a lower bound approximation for the effective energy function of the two-phase nonlinear laminate is obtained, namely,

$$\bar{W}_1(\bar{\mathbf{E}}) \geq \sup_{\epsilon_0^1, \epsilon_0^2 \geq 0} \{ \bar{W}_0^{(2)}(\bar{\mathbf{E}}) - \epsilon_0^1 \epsilon_0^2 (\epsilon_0^1) - (1 - \epsilon_0^1) \epsilon_0^2 (\epsilon_0^2) \}, \quad (4.10)$$

where $\bar{W}_0^{(2)}(\bar{\mathbf{E}})$ corresponds to the effective energy function of the two-phase, isotropic linear rank-II laminate described at the beginning of this subsection (where the two linear phases are made to correspond exactly to the two nonlinear phases). Note that this result also follows directly from the exact result (4.9) by letting $\epsilon_0^{(3)} = \epsilon_0^{(2)}$; the inequality arising because the sup over the smaller set in (4.10) leads to a result that is in general smaller than the exact result of (4.9) over the larger set.

However, more importantly, note that, as demonstrated at the beginning of this subsection (see (4.8)), $\bar{W}_0^{(2)}(\bar{\mathbf{E}})$ corresponds exactly to the Hashin-Shtrikman bounds for two-phase, isotropic linear composites, and hence the right-hand side of inequality (4.10) corresponds to the Hashin-Shtrikman bounds (and estimates) for two-phase, isotropic nonlinear composites. Thus, in general, we do not have equality between the nonlinear H-S bounds, and the nonlinear rank-II laminates. These observations apply also to the three-dimensional composites; in this case, as we will see, the exact computation of the nonlinear rank-III laminate involves a four-dimensional optimization, whereas the corresponding evaluation of the nonlinear H-S bounds of §3 involve only a two-dimensional optimization.

Thus we have reached the important conclusion that, in general, the H-S bounds (and estimates) of the previous section for two-phase nonlinear composites are not attained by rank- d iterated laminates with the same microstructure as the corresponding optimal linear laminates, and hence the nonlinear H-S bounds are probably not sharp. In fact, as we have seen, the nonlinear rank- d iterated laminates with the same microstructure as the corresponding optimal linear laminates are not even isotropic in general. This motivates the question of whether rank- d iterated laminates with different microstructures may be designed for overall isotropy in the nonlinear context. In general, we expect that the answer to this question is negative since it is known that d planes of symmetry are not sufficient to ensure overall isotropy in d dimensions. However, in the balance of this section, I show that nonlinear 'isotropic' rank- d (iterated) laminates may be constructed in some restricted sense. I also compute their properties, and in §5 compare them with the nonlinear H-S bounds and estimates of §3, for the special case of two-phase composites, where one of the phases has either zero, or infinite dielectric constant. The results for these extreme situations (we expect that the differences will be proportional to the contrast between the phases) within the class of two-phase nonlinear isotropic composites are expected to serve as a test of the conjecture that the results for the 'isotropic' rank- d laminates are not far from the actual (unknown) extremal effective properties (or optimal bounds) of two-phase nonlinear isotropic composites.

To determine the effective energy functions of the 'isotropic' nonlinear laminates, we first develop a simpler form for the effective energy functions of general nonlinear iterated laminates (i.e. simpler than (4.9)). Thus, to compute the effective energy function of nonlinear rank- d laminates ($d = 2, 3$), with layering directions $\mathbf{n}_1, \dots, \mathbf{n}_d$, we begin by specifying the result of §4a to the rank-I laminate depicted in figure 1a. Denoting by ϵ' the volume fraction of phase 1 with potential $\phi^{(1)}$ in the rank-I laminate, and noting that $1 - \epsilon'$ is the corresponding volume fraction of phase 2 with

potential $\phi^{(2)}$, we find that the effective energy function of the nonlinear, rank-I laminate is given by

$$\bar{W}_1(\bar{\mathbf{E}}) = \inf_{\omega_I^{(1)}, \omega_I^{(2)}} \{ \epsilon' \phi^{(1)}(\mathbf{s}^{(1)}) + (1 - \epsilon') \phi^{(2)}(\mathbf{s}^{(2)}) \}, \quad (4.11)$$

where $\omega_I = \epsilon' \omega_I^{(1)} + (1 - \epsilon') \omega_I^{(2)}$, and where

$$\begin{aligned} \mathbf{s}^{(1)} &= \sqrt{|\mathbf{E}^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2 + (1 - \omega_I^{(1)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2|}, \\ \mathbf{s}^{(2)} &= \sqrt{|\mathbf{E}^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2 + (1 - \omega_I^{(2)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2|}. \end{aligned}$$

Next, we consider the rank-II laminate obtained by mixing layers of the rank-I laminate with layers of a third phase characterized by energy function $\phi^{(3)}$, in relative (to the rank-II laminate) volume fractions ϵ'' and $1 - \epsilon''$, respectively. The new lamination direction $\mathbf{n}_2$ is orthogonal to $\mathbf{n}_1$, as shown in figure 1b. Then, applying Corollary 4.1 to the resulting microstructure (a three-phase material with constant fields over each isotropic phase), we arrive, via (4.9) (with thus far arbitrary ϵ''), to the following energy function for the nonlinear rank-II laminate in dimension $d \geq 2$, namely,

$$\bar{W}_2(\bar{\mathbf{E}}) = \inf_{\omega_{II}^{(1)}, \omega_{II}^{(2)}, \omega_{II}^{(3)}} \{ \epsilon'' \epsilon' \phi^{(1)}(\mathbf{s}^{(1)}) + \epsilon'' (1 - \epsilon') \phi^{(2)}(\mathbf{s}^{(2)}) + (1 - \epsilon'') \phi^{(3)}(\mathbf{s}^{(3)}) \}, \quad (4.12)$$

where $\omega_{II} = \epsilon'' \omega_{II}^{(1)} + (1 - \epsilon'') \omega_{II}^{(2)}$, and where

$$\begin{aligned} \mathbf{s}^{(1)} &= \sqrt{|\mathbf{E}^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_2)^2 + (1 - \omega_{II}^{(1)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2 + (1 - \omega_{II}^{(1)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_2)^2|}, \\ \mathbf{s}^{(2)} &= \sqrt{|\mathbf{E}^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_2)^2 + (1 - \omega_{II}^{(2)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_1)^2 + (1 - \omega_{II}^{(2)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_2)^2|}, \\ \mathbf{s}^{(3)} &= \sqrt{|\mathbf{E}^2 - (\bar{\mathbf{E}} \cdot \mathbf{n}_2)^2 + (1 - \omega_{II}^{(3)})^2 (\bar{\mathbf{E}} \cdot \mathbf{n}_2)^2|}. \end{aligned}$$

with $\epsilon'' \epsilon' = \epsilon^{(1)}$. I emphasize that we cannot make use of Corollary 4.1 to treat the rank-II laminate as a simple laminate made up of the rank-I laminate (with effective energy function given by (4.11)) and phase 3, the reason being that the rank-I laminate is anisotropic and Corollary 4.1 does not hold. Instead, we must treat the rank-II laminate as a three-phase composite; in this case, we are assured of the isotropy of all three phases and may take advantage of Corollary 4.1.

A corresponding result may be obtained for a two-phase, nonlinear rank-II laminate by letting $\phi^{(3)} = \phi^{(2)}$ in (4.12). As discussed previously, even in this case, the result for $\bar{W}_2$ is generally anisotropic, but in two dimensions ($d = 2$) it may be used to obtain an isotropic result, for each value of ϵ' , by an appropriate choice of ϵ'' (not the fixed choice that makes the corresponding linear rank-II laminate isotropic). This choice is accomplished by requiring that ϵ'' ($0 \leq \epsilon'' \leq 1$) and $\epsilon'_1 = \bar{\mathbf{E}} \cdot \mathbf{n}_1$ satisfy the relations determined by

$$\frac{\partial \bar{W}_2}{\partial \epsilon'_1} \bigg|_{\epsilon'_1, \epsilon'_2, \epsilon'_3} = 0 \quad \text{and} \quad \frac{\partial \bar{W}_2}{\partial \epsilon''} = 0, \quad (4.13)$$

where the derivative in the first relation is subject to the constraint $\epsilon'_1 + \epsilon'_2 + \epsilon'_3 = \mathbf{E}^2$, and that in the second assumes that $\mathbf{E}$ is fixed. These conditions follow by performing a Taylor series expansion of (4.12) in ϵ'' and ϵ'_1 and requiring that the expansion yield the same result for any choice of ϵ'' and $\mathbf{E}$. Physically, this corresponds to selecting a microstructure (by choosing ϵ'') with fixed overall volume fractions of the phases for each value of $\mathbf{E}$ ensuring that $\bar{W}_2$ is independent of the direction of

$\bar{E}$. Thus, the resulting energy function, $\bar{W}_{II}^*$, is isotropic. However, $\bar{W}_{II}^*$ does not correspond to a fixed microstructure, but rather to a family of (anisotropic) microstructures: one for each value of applied electric field. Therefore, $\bar{W}_{II}^*$ should be thought of as the effective energy function of an approximation to the class of nonlinear isotropic microstructures. From now on, for simplicity, I refer to the result $\bar{W}_{II}^*$, defined by (4.12) together with (4.13), as the effective energy function of the 'isotropic' nonlinear laminate. Similar comments will apply for rank-III laminates in three dimensions, which are discussed next.

The effective energy function of a nonlinear rank-III laminate is obtained by computing the effective behaviour of a simple laminate made up of layers of the rank-II laminate (just discussed) and of layers of a fourth phase with energy function $\phi^{(4)}$ in volume fractions c^{III} and $1 - c^{III}$, respectively. The new layering direction $\mathbf{n}_3$ is chosen to be orthogonal to both $\mathbf{n}_2$ and $\mathbf{n}_1$. Then, the effective energy function of the nonlinear rank III laminate may again be obtained by means of Corollary 4.1 (in this case, the fields are constant over each of the four phases), with the result that

$$\bar{W}_{III}(\bar{E}) = \inf_{\omega_1^0, \omega_2^0, \omega_3^0, \omega_4^0, \omega_5^0, \omega_6^0} \{ c^{III} c^{II} \phi^{(1)}(s^{(1)}) + c^{III} c^{II} (1 - c^I) \phi^{(2)}(s^{(2)}) + \dots + c^{III} (1 - c^{II}) \phi^{(3)}(s^{(3)}) + (1 - c^{III}) \phi^{(4)}(s^{(4)}) \}, \quad (4.14)$$

where $\bar{\omega}_{III} = c^{III} \omega_{III}^{(1)} + (1 - c^{III}) \omega_{III}^{(2)}$.

$$\begin{aligned} s^{(1)} &= \sqrt{|\bar{E}^2 - \bar{E}_1^2 - \bar{E}_2^2 - \bar{E}_3^2 + (1 - \omega_1^{(1)})^2 \bar{E}_1^2 + (1 - \omega_1^{(1)})^2 \bar{E}_2^2 + (1 - \omega_1^{(1)})^2 \bar{E}_3^2|}, \\ s^{(2)} &= \sqrt{|\bar{E}^2 - \bar{E}_1^2 - \bar{E}_2^2 - \bar{E}_3^2 + (1 - \omega_2^{(2)})^2 \bar{E}_1^2 + (1 - \omega_2^{(2)})^2 \bar{E}_2^2 + (1 - \omega_2^{(2)})^2 \bar{E}_3^2|}, \\ s^{(3)} &= \sqrt{|\bar{E}^2 - \bar{E}_1^2 - \bar{E}_2^2 - \bar{E}_3^2 + (1 - \omega_3^{(3)})^2 \bar{E}_1^2 + (1 - \omega_3^{(3)})^2 \bar{E}_2^2 + (1 - \omega_3^{(3)})^2 \bar{E}_3^2|}, \\ s^{(4)} &= \sqrt{|\bar{E}^2 - \bar{E}_1^2 - \bar{E}_2^2 + (1 - \omega_4^{(4)})^2 \bar{E}_3^2|}. \end{aligned}$$

and where I have made use of the notations $\bar{E}_i = \mathbf{E} \cdot \mathbf{n}_i$ ($i = 1, 2, 3$).

Once again, the effective energy function of a two-phase, nonlinear rank-III laminate may be obtained by letting $\phi^{(4)} = \phi^{(3)} = \phi^{(2)} = \phi^{(1)}$ in (4.14). Then, in three dimensions ($d = 3$), expression (4.14) for the rank-III laminate may be used to obtain an isotropic energy function by letting $c^{III} c^{II} c^I = c^{(0)}$, and choosing c^{II} and c^{III} ($0 \leq c^{II}, c^{III} \leq 1$), and $\bar{E}_1, \bar{E}_2, \bar{E}_3$ with $\bar{E}_1^2 + \bar{E}_2^2 + \bar{E}_3^2 = \bar{E}^2$ such that the relations implicit in

$$\frac{\partial \bar{W}_{III}}{\partial \bar{E}_1} |_{\bar{E}_1^2, \bar{E}_2^2, \bar{E}_3^2, c^I, c^{II}, c^{III}} = \frac{\partial \bar{W}_{III}}{\partial \bar{E}_2} |_{\bar{E}_1^2, \bar{E}_2^2, \bar{E}_3^2, c^I, c^{II}, c^{III}} = 0, \quad \frac{\partial \bar{W}_{III}}{\partial c^{II}} = \frac{\partial \bar{W}_{III}}{\partial c^{III}} = 0 \quad (4.15)$$

are satisfied. The resulting isotropic energy function $\bar{W}_{III}^*$, corresponding to a nonlinear 'isotropic' rank-III composite, may be used to model the behaviour of nonlinear isotropic composites in three dimensions. In the next section, I compare this and the previous results for the rank-II laminates with the bounds and estimates of §3b for isotropic nonlinear composites, with some special choices for their constituent phases.

5. Application to two-phase composites

To illustrate the power of the methods described in the previous two sections for determining bounds and estimates for the effective properties of nonlinear composites, I investigate in this section two simple, but important, examples. They both involve a nonlinear matrix with isotropic potential $\phi^{(2)} = \phi$ (subject to the

restrictions of Theorem 2.1), and an inclusion phase with either infinite dielectric constant, or alternatively, with zero dielectric constant. In the first case, we have that $\phi^{(1)} = 0$ if $E = 0$, or $\phi^{(1)} = \infty$ otherwise; and in the second, we have that $\phi^{(1)} = 0$ regardless of the value of E . Thus I compute bounds for these nonlinear composites applying the results of §3, and compare them with exact estimates for 'isotropic' laminates based on the results of §4.

(a) The nonlinear material with perfectly conducting inclusions

The first observation is that the upper bounds for this class of composite materials, corresponding to geometries with the unbounded dielectric material in the matrix phase, are unbounded. Thus we concentrate our efforts in determining lower bounds and minimal estimates for the effective properties of this class of nonlinear materials.

According to relation (3.12), the Wiener lower bound for this nonlinear material is given by

$$\bar{W}_W(\bar{E}) = (1 - c) \phi(\bar{E}/(1 - c)), \quad (5.1)$$

where I have used the fact that $\omega = (1 - c)^{-1}$ solves the infimum problem in (3.12). Here c is the volume fraction of the perfectly conducting inclusions. The corresponding H-S lower bound is obtained by specializing relation (3.24) and noting that once again $\omega = (1 - c)^{-1}$ solves the optimization problem. The result may be written in the form

$$\bar{W}_{HS}(\bar{E}) = (1 - c) \phi \left(\sqrt{\frac{1 + (d - 1)c^d}{1 - c}} \bar{E} \right). \quad (5.2)$$

Note that for $d > 1$, $\bar{W}_{HS} > \bar{W}_W$, and the H-S bound for isotropic materials is sharper than the Wiener bound for generally anisotropic materials (which also holds, in particular, for isotropic materials). Note that the above bounds hold for arbitrary nonlinear matrix satisfying the growth conditions mentioned at the end of §2c (superquadratic growth for ϕ). We note further that the above results for the H-S bounds can also be obtained by the method of Talbot & Willis (1985), although the specific results described above have apparently not yet appeared in the published literature. Analogous results in nonlinear infinitesimal elasticity have been given by Ponte Castañeda & Willis (1988) and improved by Ponte Castañeda (1991a, b). I do not consider the Beran bounds in this section, because they require more geometric information than the volume fractions of the phases.

Next, I obtain results for the nonlinear 'isotropic' laminates by considering the two- and three-dimensional laminates separately. The effective energy function of the two-dimensional 'isotropic' laminate is obtained directly from relations (4.12) and (4.13) with $\phi^{(2)} = \phi^{(3)} = \phi$, and by noticing that $\omega_1^{(0)} = 1$ and $\omega_2^{(0)} = 1$ solve the optimization problem in this case. This leads to the result that

$$\bar{W}_{II}^*(\bar{E}) = \text{stat}_{\substack{c^{II} \\ 0 \leq c^{II} \leq 1}} \text{stat}_{\substack{E_1, E_2 \\ E_1^2 + E_2^2 = \bar{E}^2}} \left\{ (c^{II} - c) \phi \left(\left(\frac{c^{II}}{c^{II} - c} \right) \bar{E}_1 \right) + (1 - c^{II}) \phi \left(\sqrt{E_1^2 + \left(\frac{E_2}{1 - c^{II}} \right)^2} \right) \right\}, \quad (5.3)$$

where I have rewritten the isotropy conditions (4.13) in the form of stationary optimization processes. In general, these optimizations will need to be carried out numerically, but we will see in §5c that further simplification can be achieved for special choices of ϕ . Note also that a quadratic choice for ϕ leads to the exact H-S lower bound for a two-phase, linear isotropic composite.

The corresponding three-dimensional result for the effective energy function of the nonlinear, isotropic rank-III laminate is obtained in a similar fashion from (4.14) and (4.15), and is given by

$$\begin{aligned} \bar{W}_{III}^1(\mathbf{E}) = & \operatorname{stat}_{\substack{c^{II}, c^{III} \\ 0 \leq c^{II}, c^{III} \leq 1}} \left\{ \operatorname{stat}_{\substack{\mathbf{E}_1, \mathbf{E}_2 \\ \mathbf{E}_1^2 + \mathbf{E}_2^2 = \mathbf{E}^2}} \left[\left(\frac{c^{III} c^{II}}{c^{III} c^{II} - c} \right) \mathbf{E}_1 \right] + \right. \\ & \left. \dots + c^{III} (1 - c^{II}) \phi \left[\sqrt{\mathbf{E}_1^2 + \left(\frac{\mathbf{E}_2}{1 - c^{II}} \right)^2} \right] + (1 - c^{III}) \phi \left[\sqrt{\mathbf{E}_1^2 + \mathbf{E}_2^2 + \left(\frac{\mathbf{E}_2}{1 - c^{III}} \right)^2} \right] \right\}. \end{aligned} \quad (5.4)$$

This result can also be shown to reduce to the corresponding linear H-S lower bound for a quadratic choice for ϕ .

(b) *The nonlinear material with perfectly insulating inclusions*

In this case, note that the lower bounds will of necessity be trivial because the choice of the perfect insulator as the matrix phase leads to perfectly insulating effective behaviour for the composite. Thus I attempt to obtain upper bounds, and maximal estimates for the effective energy function of this class of composite material. The Wiener upper bound, corresponding to arbitrarily anisotropic composites, is obtained from (3.26), which yields the result

$$\bar{W}_W(\mathbf{E}) = (1 - c) \phi(\mathbf{E}), \quad (5.5)$$

where c now stands for the volume of the perfectly insulating inclusions. The Hashin-Shtrikman upper 'estimate' (not a rigorous bound) is obtained from (3.21) with the min replaced by a max. The result is

$$\bar{W}_{HS}(\mathbf{E}) = (1 - c) \phi \left(\sqrt{\left(\frac{d-1}{d-1+c} \right) \mathbf{E}} \right). \quad (5.6)$$

Although, as already mentioned, we do not expect this result to yield an upper bound in general, it does reduce to the well-known H-S upper bound in the case of a linear matrix.

The exact estimate for the two-dimensional, nonlinear 'isotropic' laminate is obtained from expressions (4.12) and (4.13) with $\phi^{(2)} = \phi^{(3)} = \phi$. In this case, the result is not as explicit as the corresponding result in §5a, and takes the form

$$\bar{W}_{II}^1(\mathbf{E}) = \operatorname{stat}_{\substack{c^{II} \\ 0 \leq c^{II} \leq 1}} \left\{ \operatorname{stat}_{\substack{\mathbf{E}_1^2, \mathbf{E}_2^2 \\ \mathbf{E}_1^2 + \mathbf{E}_2^2 = \mathbf{E}^2}} \left[\inf_{\omega_{II}} \{ (c^{II} - c) \phi(s^{(2)}) + (1 - c^{II}) \phi(s^{(3)}) \} \right], \right. \quad (5.7)$$

where

$$\begin{aligned} s^{(2)} &= (1 + c^{II} \omega_{II}) \mathbf{E}_2, \\ s^{(3)} &= \sqrt{\mathbf{E}_1^2 + [1 - (1 - c^{II}) \omega_{II}]^2 \mathbf{E}_2^2}. \end{aligned}$$

Further simplification is available for special choices of ϕ , as we see in §5c. Also, this result reduces to the H-S upper bound in the limit of linear behaviour for the matrix.

The result for the effective energy function of the three-dimensional, nonlinear 'isotropic' laminate is obtained analogously, with the result that

$$\begin{aligned} \bar{W}_{III}^1(\mathbf{E}) = & \operatorname{stat}_{\substack{c^{II}, c^{III} \\ 0 \leq c^{II}, c^{III} \leq 1}} \left\{ \operatorname{stat}_{\substack{\mathbf{E}_1, \mathbf{E}_2 \\ \mathbf{E}_1^2 + \mathbf{E}_2^2 = \mathbf{E}^2}} \left[\inf_{\omega_{II}, \omega_{III}} \{ (c^{III} c^{II} - c) \phi(s^{(2)}) + \right. \right. \\ & \left. \left. \dots + c^{III} (1 - c^{II}) \phi(s^{(3)}) + (1 - c^{III}) \phi(s^{(4)}) \} \right], \right\} \quad (5.8) \end{aligned}$$

Phil Trans R Soc Lond A (1992)

$$\begin{aligned} s^{(2)} &= \sqrt{\{(1 + c^{III} \omega_{II})^2 \mathbf{E}_2^2 + [1 + c^{III} \omega_{III}]^2 \mathbf{E}_3^2\}}, \\ s^{(3)} &= \sqrt{\{\mathbf{E}_1^2 + [1 - (1 - c^{III}) \omega_{II}]^2 \mathbf{E}_2^2 + (1 + c^{III} \omega_{III})^2 \mathbf{E}_3^2\}}, \\ s^{(4)} &= \sqrt{\{\mathbf{E}_1^2 + \mathbf{E}_2^2 + [1 - (1 - c^{III}) \omega_{III}]^2 \mathbf{E}_3^2\}}. \end{aligned}$$

Again no further simplification of this result seems possible for general ϕ , but for the special case considered in the following subsection for pure-power behaviour for the matrix, a simpler form of the result is available.

(c) *Special results for pure-power materials*

In this subsection, I specialize the results for the two special types of nonlinear composites of §§5a and b by considering pure-power behaviour for the matrix material. This type of behaviour for the matrix, characterized by the energy-density function

$$\phi(\mathbf{E}) = (n + 1)^{-1} \epsilon \mathbf{E}^{n+1} \quad (5.9)$$

(where $n \geq 1$ is the power and ϵ is the nonlinear dielectric constant), has the advantage that it yields the same type of behaviour for the isotropic composite materials with perfectly conducting, or perfectly insulating, inclusions. Thus, for these two types of isotropic composites, we have that

$$\bar{W}(\mathbf{E}) = (n + 1)^{-1} \epsilon \mathbf{E}^{n+1}, \quad (5.10)$$

where $\bar{\epsilon}$ denotes the effective nonlinear dielectric constant of the composite. For the anisotropic composites, the form of the effective energy will be different in general, but at least the Wiener bounds will also be of the form (5.10). We can then characterize the behaviour of the Wiener, Hashin-Shtrikman and 'isotropic' laminates for the class of composites studied in §§5a and b in terms of this effective nonlinear dielectric constant, appropriately normalized by the dielectric constant of the matrix material (i.e. $\bar{\epsilon}/\epsilon$).

The results for the bounds and estimates of two-dimensional composites are not essentially different from the results for three-dimensional composites. For this reason, I only present results here for the three-dimensional composites. I begin by depicting the results for the Wiener lower bound, the H-S lower bound and the 'isotropic' laminate estimate, as given by relations (5.1), (5.2) and (5.4), respectively, for the nonlinear material with perfectly conducting inclusions. These results, expressed in terms of the effective nonlinear dielectric constant, are given respectively by

$$\begin{aligned} \bar{\epsilon}_{HS}/\epsilon &= (1 + 2c)^{(n+1)/2} / (1 - c)^n, \\ \bar{\epsilon}_{II}/\epsilon &= \sup_{\substack{0 \leq c \leq 1 \\ \forall \mathbf{E} \in \mathcal{E}_1}} \left\{ (1 - y)^{2n/(n+1)} + \left(\frac{2 - y}{y} \right)^{(n+1)/(n+1)} \left\{ (xy - c)^{2n/(n+1)} \right\} \right. \\ & \quad \left. \times \left(\frac{2 - x}{x} \right)^{(n+1)/(n+1)} + \{(1 - x)y\}^{2n/(n+1)} \right\}^{(1)/(n+1)}. \end{aligned} \quad (5.11)$$

Note that the expressions for the bounds are remarkably simple. On the other hand, the expression for the exact estimate for the 'isotropic' laminate involves a two-dimensional optimization, which must be solved numerically. It is interesting to note that $(\bar{\epsilon}_{II}/\epsilon)^{1/n} \rightarrow (1 - 8c + 12c^2 - 6c^3 + c^4)^{-1/4}$ as $n \rightarrow \infty$, which is different from, but close to, $(\bar{\epsilon}_{HS}/\epsilon)^{1/n}$ in the limit as $n \rightarrow \infty$.

Phil Trans R Soc Lond A (1992)

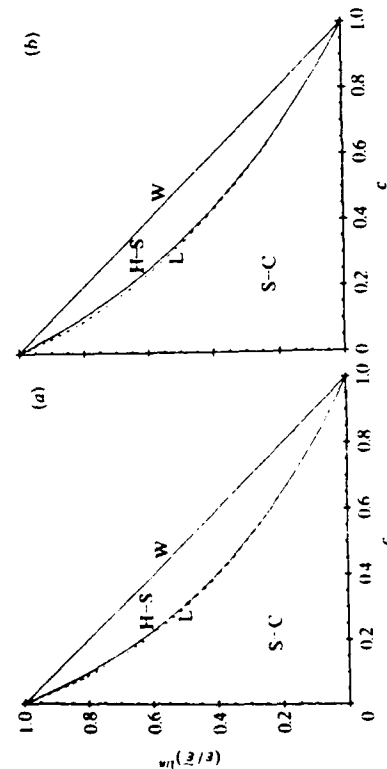


Figure 2. Plot of the effective nonlinear dielectric constant as a function of the volume fraction of perfectly conducting inclusions for (a) a weakly nonlinear matrix ($n = 3$) and (b) a strongly nonlinear matrix ($n = 10$).

The results of these computations for the effective dielectric constant of the nonlinear 'isotropic' laminate as functions of the volume fraction c of the perfectly conducting inclusions for two values of n (3 and 10, corresponding to a weakly nonlinear and a strongly nonlinear material, respectively) are given in figure 2a and b (they are denoted by L). In these plots, we have also included for comparison the results for the bounds (W and H-S), as well as self-consistent (S-C) estimates obtained in the same way as the nonlinear H-S bounds, but making use of the linear self-consistent estimate instead of the linear H-S bounds in relation (3.3). This self-consistent result takes the form

$$\hat{\epsilon}_{sc}/\epsilon = (1-c)^{-n/2}(1-3c)^{-n/2} \quad (5.12)$$

All of the above results are depicted in terms of the reciprocal of the effective dielectric constant to an appropriate power for plotting convenience. Thus the lower bounds appear as upper bounds in these plots. By comparing the Wiener and H-S bounds, we observe that the H-S bounds are significantly tighter than the Wiener bounds. This is not surprising since the Wiener bounds must apply for generally anisotropic materials, which is a larger class of materials than the class of isotropic materials for which the H-S bounds must hold. Recall, however, that the Wiener bounds were shown to be sharp within the class of arbitrarily anisotropic composites. (Comparing the H-S bounds with the exact estimates for the 'isotropic' laminates (1), and observing that they are very close, even for the larger value of n , is suggestive that the H-S bounds, although probably not optimal, are close to the optimal bounds. Such optimal bounds would be expected to lie between the exact estimates for the 'isotropic' laminates, which are approximations to actual microstructures within the class of two phase isotropic materials, and the H-S bounds, which are rigorous bounds in this case. Finally, we note that the self-consistent estimates lie below the bounds, as they should, and that they are close to the bounds for small volume fractions of the perfectly conducting inclusions, but diverge away from the bounds for moderate volume fractions reaching an early percolation limit at $c = \frac{1}{3}$.)

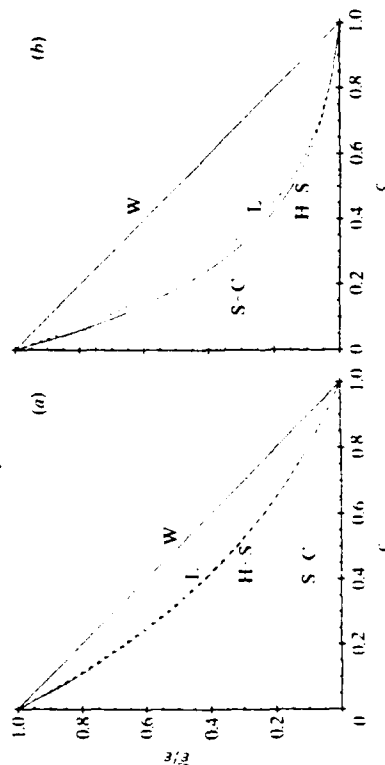


Figure 3. Plot of the effective nonlinear dielectric constant as a function of the volume fraction of perfectly insulating inclusions for (a) a weakly nonlinear matrix ($n = 3$) and (b) a strongly nonlinear matrix ($n = 10$).

Next, we consider the corresponding results for the nonlinear material with perfectly insulating inclusions in three space dimensions. These results for the Wiener upper bound, the H-S 'upper estimate', the self-consistent estimate and the exact estimate for the 'isotropic' laminate are given, respectively, by the relations

$$\left. \begin{aligned} \hat{\epsilon}_{Wc}/\epsilon &= (1-c), \\ \hat{\epsilon}_{HSc}/\epsilon &= (1-c)/(1+\frac{1}{2}c)^{n+1/2}, \\ \hat{\epsilon}_{sc}/\epsilon &= (1-\frac{3}{2}c)^{n+1/2}/(1-c)^{n+1/2}, \\ \hat{\epsilon}_{IL} &= \sup_{\substack{0 \leq x, y \leq 1 \\ xy \geq c}} \left\{ \frac{[(1-x)y + (1-y)x]p^{n+1/2}}{[(xy-c)^{n+1/2} + (1-x)y + (1-y)x]p^{n+1/2}} \right\} \end{aligned} \right\} \quad (5.13)$$

where p solves the quadratic equation

$$\frac{1}{2}(1-y)/(1-x)p^2 - \frac{1}{2}xy + 2(1-y)p + (1-x)(1-y) = 0,$$

and

$$q = \left(\frac{xy}{xy-c} \right) \left(\frac{1-x}{2-x} \right).$$

These results are plotted as functions of the volume fraction of inclusions c in figure 3a and b for two values of n (3 and 10). We observe that, as expected, the H-S 'upper estimates' for the isotropic composite lie well below the Wiener bounds for the arbitrarily anisotropic composites. On the other hand, we find that the exact estimates for the nonlinear 'isotropic' laminates (L) lie above the H-S 'upper estimates'. This verifies our expectation, proposed in §4, that the H-S 'upper estimates' are probably not in general upper bounds. This is because the 'isotropic' laminates correspond to (approximations of) specific microstructures within the class of isotropic composite materials, and if the H-S estimates were rigorous bounds for this class of composite materials, they would have to lie above all possible isotropic microstructures, and, in particular, they would be expected to lie above the 'isotropic' laminates. Nevertheless, we find that the effective dielectric constants of

the 'isotropic' laminates are not far from the H-S 'upper estimates'. Thus, we anticipate that the optimal upper bounds for this class of materials would probably not be too distant from the 'isotropic' laminate results, and hence not too far from the H-S 'upper estimates'. Then, the H-S 'upper estimates', although not bounds, would provide simple estimates for the extremal properties of this class of nonlinear composites. We add that the self-consistent estimates agree with the H-S estimates for low values of ϵ , but they reach an early percolation limit at $\epsilon = \frac{1}{3}$.

Finally, we note that through the well-known connections between the H-S bounds and the Maxwell-Garnett approximations, and between the self-consistent and symmetric effective-medium estimates in the context of linear conductivity, we could equally well refer to our nonlinear H-S bounds as nonlinear Maxwell-Garnett estimates, and to the self-consistent estimates as nonlinear effective-medium estimates.

6. Concluding remarks

To summarize, in this paper, I have made use of the variational principles proposed by Ponte Castañeda (1992) (Theorems 2.1 and 2.2) to develop bounds and estimates for the effective properties of nonlinear dielectrics. These variational principles can generally be used in either one of two ways: they can be used approximately to yield bounds for whole classes of microstructures (and for specific microstructures), or they can be used exactly to determine the effective properties of certain special microstructures. Prior work by the author, in different physical contexts (Ponte Castañeda 1991*a, b*), has made use of the approximate versions of the new variational principles to determine bounds for the effective properties of nonlinear composites. The present work is the first to consider the exact use of the variational principles in the computation of the effective properties of specific microstructures.

In §3, which deals with the approximate use of the variational principles to obtain bounds (Corollaries 3.1 and 3.2), I was able to recover the classical bounds of Wiener for generally anisotropic nonlinear composites. More importantly, I also obtained bounds of the Hashin-Shtrikman type for nonlinear isotropic composites by direct implementation of the corresponding linear Hashin-Shtrikman bounds into the new variational principles. Bounds of the Hashin-Shtrikman type for nonlinear dielectrics have also been proposed by Willis (1986) and by Talbot & Willis (1985), starting from an appropriate generalization of the Hashin-Shtrikman variational principles for nonlinear problems. However, the new method has the distinct advantage that it is not limited to bounds of the Hashin-Shtrikman type, and can be used to yield other bounds (and estimates), such as higher-order bounds (and self-consistent estimates), by direct application of the corresponding higher-order linear bounds (and estimates). Thus I was able to obtain, for the first time, bounds of the Beran type for two-phase, nonlinear isotropic composites. On the other hand, note that the method of Talbot & Willis has the advantage that it applies to anisotropic behaviour for the phases, whereas the new method, as developed thus far, applies only to isotropic phases. However, generalizations of the new method to anisotropic behaviour are certainly possible, and these will be explored elsewhere. One weakness of both methods is that the bounds generated are only one-sided. Additionally, we note that the final form of the expressions for the nonlinear Wiener and Hashin-Shtrikman bounds and estimates obtained in this work is new and simpler than previously available forms.

Phil. Trans. R. Soc. Lond. A (1992)

In §§4 and 5, we studied the application of the exact versions of the new variational principles (Corollaries 4.1 and 4.2) to compute exact estimates for the effective properties of special classes of nonlinear composite materials. These special materials, called sequentially laminated materials, are obtained as their name implies by a sequence of lamination procedures, and have the advantage that their effective behaviour can be controlled to a large extent by appropriate choices of the lamination directions, and relative volume fractions of the phases within each elemental layer. Thus, in particular, we show that a simple laminate leads to the most anisotropic composite possible within the class of arbitrarily anisotropic nonlinear composites with prescribed volume fractions, in the sense that the effective energy of the simple laminate attains the Wiener bounds (for special choices of the applied electric field). Analogous, but not quite as strong, results are obtained for the class of isotropic composites, by considering 'isotropic' laminates (in the sense described in §4*b*). In this case, we found in §5*c* that the 'isotropic' laminates do not quite attain, but lie below, the nonlinear Hashin-Shtrikman lower bounds for isotropic composites. On the other hand, we also verified that the Hashin-Shtrikman 'upper estimates' cannot be rigorous upper bounds in general (since they do not encompass the nonlinear 'isotropic' laminates), but nevertheless they are probably not far from the optimal upper bounds. It is expected that sequentially laminated microstructures, which have been found in recent years to play an important role in the linear theories, will also continue to play an important role in the development of the nonlinear theories. They have many advantages over other standard types of models because their effective properties can be computed exactly, and also because they can be given generalizations in other fields, where other models may be too difficult to apply.

This research was supported in part by the National Science Foundation/Materials Research Laboratory program at the University of Pennsylvania under Grant no. 91-20608 and in part by the Air Force Office of Scientific Research under Grant no. 91-0161.

Appendix

In this appendix, I give a simple proof of the identity (3.7) used throughout the body of the paper. I begin by introducing a Lagrange multiplier to account for the zero-average constraint on the optimization variables, $\omega^{(n)}$. Thus,

$$\inf_{\omega^{(n)}} \left\{ \sum_{i=1}^n \epsilon^{(i)} \epsilon^{(i)} (1 - \omega^{(n)})^2 \right\} = \inf_{\omega^{(n)}} \left\{ \sum_{i=1}^n \epsilon^{(i)} \epsilon^{(i)} (1 - \omega^{(n)})^2 + \sup_{\lambda} \left\{ \lambda \sum_{i=1}^n \epsilon^{(i)} \omega^{(i)} \right\} \right\}, \quad (\text{A } 1)$$

and, by duality, we have

$$\inf_{\omega^{(n)}} \left\{ \sum_{i=1}^n \epsilon^{(i)} \epsilon^{(i)} (1 - \omega^{(n)})^2 \right\} = \sup_{\lambda} \left\{ \inf_{\omega^{(n)}} \left\{ \sum_{i=1}^n \epsilon^{(i)} \epsilon^{(i)} (1 - \omega^{(n)})^2 + \lambda \sum_{i=1}^n \epsilon^{(i)} \omega^{(i)} \right\} \right\}. \quad (\text{A } 2)$$

The infimum over the $\omega^{(n)}$ is satisfied by

$$\omega^{(n)} = 1 - \lambda / 2\epsilon^{(n)}, \quad (\text{A } 3)$$

which finally leads to

$$\inf_{\omega^{(n)}} \left\{ \sum_{i=1}^n \epsilon^{(i)} \epsilon^{(i)} (1 - \omega^{(n)})^2 \right\} = \sup_{\lambda} \left\{ \sum_{i=1}^n \epsilon^{(i)} \left\{ \lambda - \frac{\lambda^2}{4\epsilon^{(i)}} \right\} \right\} = \sup_{\lambda} \left\{ \lambda - \frac{\lambda^2}{4} \sum_{i=1}^n \frac{\epsilon^{(i)}}{\epsilon^{(i)}} \right\} = \left(\sum_{i=1}^n \frac{\epsilon^{(i)}}{\epsilon^{(i)}} \right)^2. \quad (\text{A } 4)$$

Phil. Trans. R. Soc. Lond. A (1992)

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Received 30 August 1991; revised 9 March 1992; accepted 7 April 1992