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TOUGHENING, STRENGTHENING, AND SYNERGISM

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THEORY OF CERAMIC/ZIRCONIA COMPOSITES: TOUGHENING, STRENGTHENING, AND SYNERGISM

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ABSTRACT

A review is given of recent theoretical results concerning toughening and strengthening in brittle ceramics reinforced with dilatantly transforming zirconia particles. Small-scale analysis of "long" dominant cracks provides resistance curves that exhibit peak fracture toughnesses for finite amounts of crack growth. Similar results for the tensile fracture strengths of "short" cracks have implications for transformation strengthening. The interaction due to the presence of *both* transforming particles and ductile metal particles can result in synergistic reinforcing effects over a parametric range of material properties. Keywords; Ceramic Materials, Zirconium Alloys, Particle reinforcements, Mechanical Properties, Crystal Structure, Transformation toughening, Transformation Strengthening, Synergi

INTRODUCTION

Small zirconia particle reinforcements can produce substantial increases in the toughnesses of normally brittle ceramic materials by undergoing a martensitic phase transformation from a tetragonal to monoclinic crystal structure. The critical Mode-I fracture toughnesses, K_{IC}, of "transformation-toughened" composites can exceed 15 MPa$\sqrt{m}$, while the values of the unreinforced matrix ceramics are typically ≈ 2 - 4 MPa$\sqrt{m}$[1].

This paper reviews results for resistances to crack growth in ceramic/zirconia systems. The first two sections consider, respectively, toughening and strengthening due to the growth of pre-existing cracks in zirconia reinforced ceramics. The final portion treats toughness increases in ceramics reinforced with *both* phase-transforming zirconia particles and ductile metal particles.

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TRANSFORMATION TOUGHENING

Transformation toughening has been most frequently studied in the applied mechanics literature by use of the "supercritical" transformation model[2]. The "stress-free" transformation strain of the unconstrained zirconia particles consists of a dilatation of about 4% and a shear strain of about 16%. However, the toughening effects of randomly oriented or twinned transformation shear strains have often been neglected. In the "supercritical" model, the region of matrix ceramic and transformed particles surrounding an advancing crack tip is modeled as an elastic continuum that has undergone a permanent "stress-free" transformation strain of strength $c_t\theta_p^T$, where c_t is the volume fraction of tetragonal particles (typically .3-.5) and θ_p^T is the particle dilatation (approximately .04). The transformation is assumed to occur completely, that is "supercritically", when the mean stress $\sigma_m = \sigma_{kk}/3$ reaches the critical value σ_m^c. This model is used throughout this paper and is applied first to steady-state cracks.

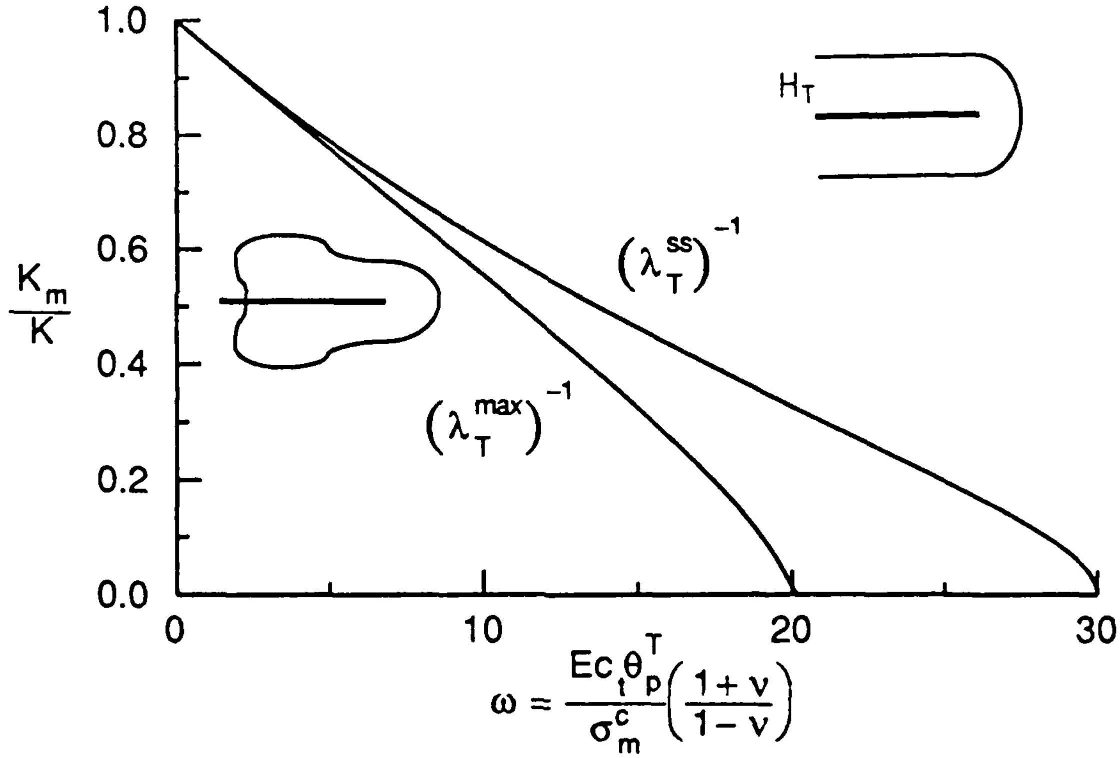

Fig. 1 Reciprocal toughness ratios versus the toughening parameter ω.

Steady-state crack growth, shown in the insert of Figure 1, has been considered in several studies[2,3]. The crack faces are bordered by small-scale transformed regions of height H_T, and the asymptotic near-tip and remote stresses obey Mode-I K-fields with the respective magnitudes K_{tip} and K. Along the exterior of the leading edge of the transformed region, the mean stress due to the combined effects of the applied $K=K_{ss}$ and the transformations themselves has just reached σ_m^c, while K_{tip} is held at

K_m, the critical stress-intensity factor for fracture of the matrix ceramic. An important *transformation-toughening* parameter and a plane-strain characteristic length[4] are provided by

$$\omega = \frac{E c_t \theta_p^T}{\sigma_m^c}\left(\frac{1+\nu}{1-\nu}\right) \tag{1}$$

and

$$L = \frac{2}{9\pi}\left[\frac{K_m(1+\nu)}{\sigma_m^c}\right]^2 \tag{2}$$

(In general, (1) and (2) cannot be evaluated ab initio because values of σ_m^c for constrained zirconia inclusions are very uncertain.) The complete results[4] for the steady-state toughness ratio $\lambda_T^{ss} = K_{ss}/K_m$, shown by the outer line in Figure 1, a plot of $(\lambda_T^{ss})^{-1}$ versus the toughening parameter ω, confirmed the discovery[5] of "lock-up" (i.e. divergence of the toughness ratio) for $\omega = \omega_c \approx 30$. The height ratio H_T/L is also a function of the parameter ω that diverges for ω_c, but is not shown in this paper. Estimates of ω and L have been made by fitting experimental measurements to the steady-state results and give predictions in the ranges $0 \le \omega \le 15$ and $L \approx 2$ - $15\ \mu m$.

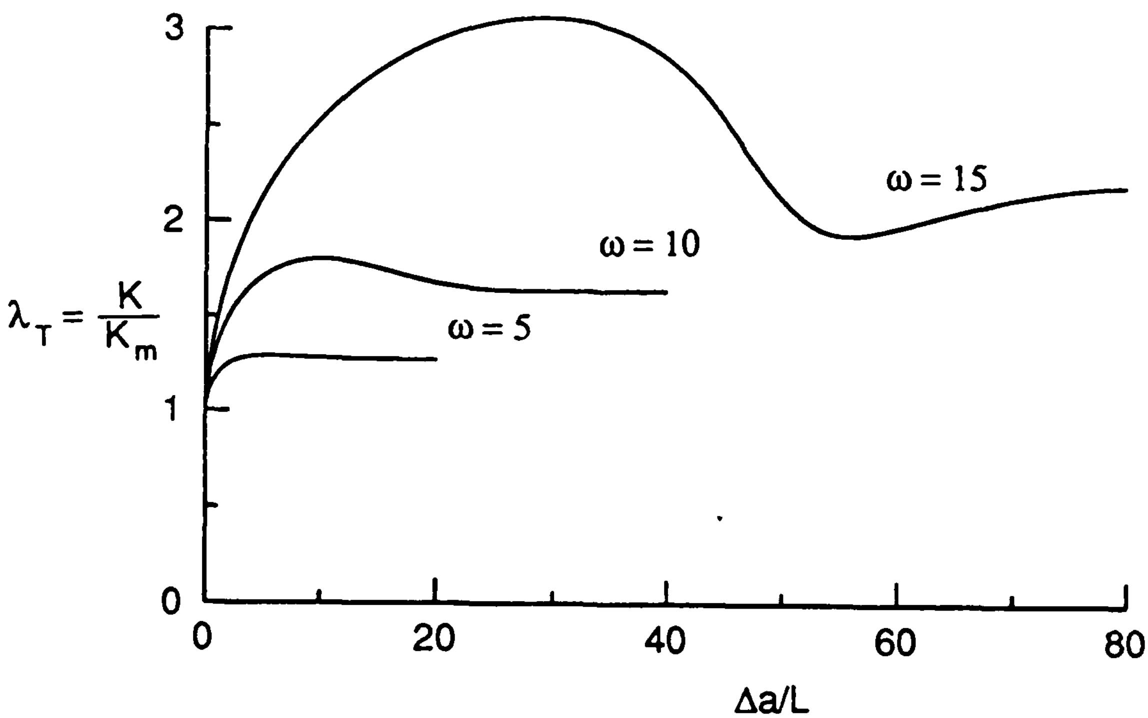

Fig. 2 Toughness ratios versus normalized crack growth; various ω.

More recently, the transient growth of precut "long" (i.e. semi-infinite) cracks in supercritical ceramics has been considered[6]. As a stationary precut crack is subjected to a remote "applied" K, a region of transformed material develops around the tip according to the σ_m^c criterion. The initial transformed region provides no toughening[2], so that crack-growth initiation occurs when the "applied" $K = K_{tip} = K_m$. As the tip advances, "fresh" material ahead of the tip is transformed and previously transformed material is left behind bordering the crack faces. Throughout crack growth, the combined effects of the "applied" K and the transformations themselves maintain σ_m at σ_m^c along a varying portion of the leading edge of the transformed region while K_{tip} is held at K_m. It was discovered that the height of the transformed region broadens substantially just after the onset of growth before narrowing back down to the steady-state level, as has been shown schematically in Figure 1. Plots of the instantaneous toughness ratio $\lambda_T = K/K_m$ versus the normalized extension $\Delta a/L$ (i.e. R-curves) are shown in Figure 2 for various values of the parameter ω. In conjunction with the zone broadening, each λ_T reaches a peak value λ_T^{max}, which exceeds the steady-state level λ_T^{ss}, at finite amounts of crack growth. The λ_T's then asymptotically approach the respective levels of λ_T^{ss}. The full results for $(\lambda_T^{max})^{-1}$ as a function of the parameter ω are given the inner curve of Figure 1. Surprisingly, "lock-up" of the growing cracks occurs for the value $\omega \approx 20.2$. Thus, for $0 \le \omega \le 15$, peak toughnesses are expected to exceed the corresponding steady-state values by up to 50%. It is noteworthy that some experimental measurements of R-curves have indeed shown peaks[7], although conclusive corroboration of zone broadening is still awaited.

TRANSFORMATION STRENGTHENING

The presence of phase-transforming zirconia particles is also predicted to improve the tensile fracture strengths of ceramics containing finite cracks. A recent study[8] has examined the growth of an isolated pre-existing finite crack in a "supercritical" ceramic subjected to a remote applied tensile-stress σ. The enhanced strengths are conveniently normalized by the fracture strength of the *unreinforced* ceramic, σ_0, which is given by

$$\sigma = \sigma_0 = \frac{K_m}{\sqrt{\pi a}} \tag{3}$$

where K_m is the critical stress-intensity factor for fracture of the matrix ceramic and 2a is the initial crack length. When transformations are included in the analysis, the parameter ω and the characteristic length L re-emerge along with the *crack-length* parameter a/L. We discuss briefly the finite crack results, beginning with crack-growth initiation.

As the remote tensile stress σ is applied, the high stresses near the crack tips at $\pm a$, given asymptotically by Mode-I K-fields, cause surrounding material to transform supercritically according to the σ_m^c criterion. When σ reaches the special value σ_i, crack-growth initiation occurs at $K_{tip} = K_m$. The "initiation" stress ratio σ_i/σ and zone shape depend upon both the parameters ω and a/L. Surprisingly, σ_i/σ_0 is less than unity except for the limit of very "long" cracks (i.e. $a/L \to \infty$) so that the presence of the transformations *weakens* the composite with respect to crack-growth initiation. (In fact for very large values of ω, *self-cracking* can even occur as $\sigma_i/\sigma_0 \to 0$.) However for values of $0 \le \omega \le 15$, initiation strengths are expected to be reduced by at most 25% for even the smallest critical crack sizes (i.e. $a/L \approx 1$).

As the finite crack grows, "fresh" material ahead of the tips is transformed while regions of previously transformed material are left behind bordering the crack faces. Throughout growth, the combined effects of the "applied" stress σ and the transformations maintain σ_m at σ_m^c just outside a portion of the leading edges of the transformed regions while holding K_{tip} at the critical value K_m. Some calculated stress ratios σ/σ_0 are plotted versus the normalized tip advance $\Delta a/L$ in Figure 3 for $\omega = 10$ and various a/L. In the limit the parameter $a/L \to \infty$, the semi-infinite crack results for K/K_m apply. Consistent with peak reinforcing effects, each σ/σ_0 curve achieves a maximum value, σ_{max}/σ_0, at finite amounts of crack growth. However, as the parameter a/L approaches $O(1)$, the levels of σ_{max}/σ_0 drop dramatically. In fact, for the parametric ranges $\omega < 10$ and $a/L \approx 1$, σ_{max}/σ_0 does not even reach unity.

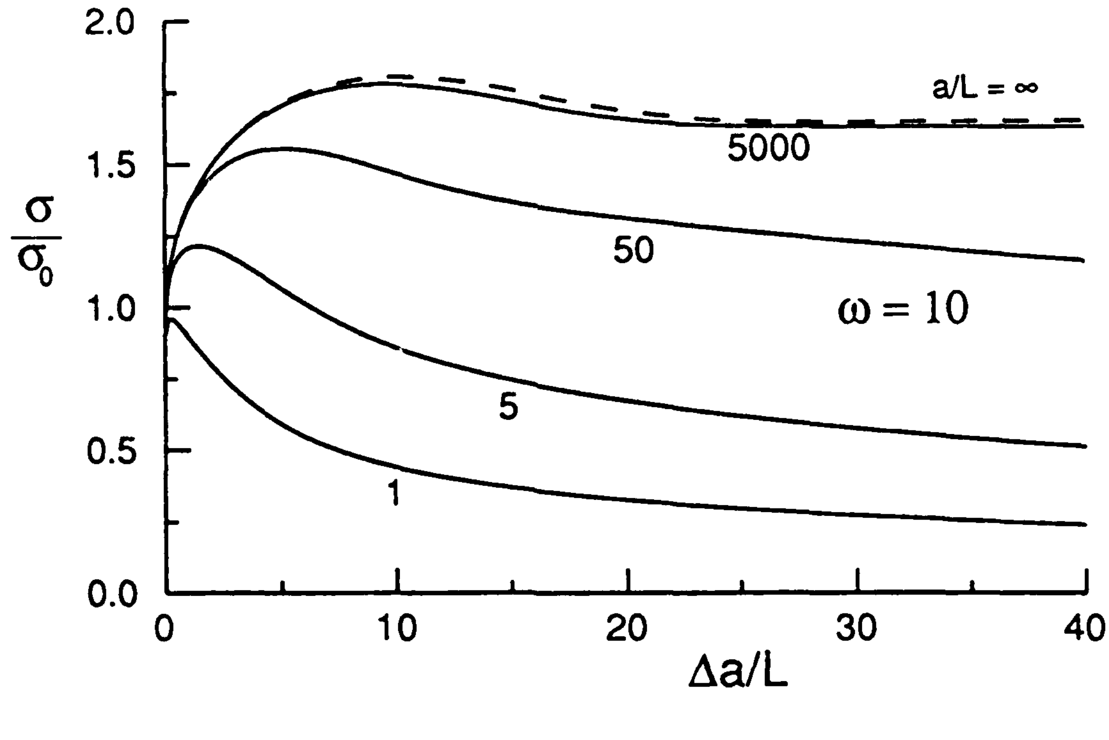

Fig. 3 Stress ratios versus normalized crack growth; $\omega = 10$, various a/L.

The finite crack results can be summarized succinctly in terms of a *transformation-strengthening* parameter

$$t = \frac{\omega}{3\pi}\sqrt{\frac{2a}{L}} = \frac{E c_t \theta_p^T \sqrt{a}}{K_m(1-\nu)} \tag{4}$$

which contains readily accessible quantities. Figure 4 shows plots of σ_{max}/σ_0 versus peak toughening ratios λ_T^{max} for various t. In the limit $t \to \infty$, for which the semi-infinite crack results apply, σ_{max}/σ_0 coincides with λ_T^{max}. Based on the reported values[1] E=200 GPa, c_t=.3, θ_p^T=.04, and K_m=3 MPa$\sqrt{m}$, critical crack sizes a = 1 - .01 mm. only correspond to t ≈ 3.5 - 35. Thus, the tensile fracture strengths of ceramic/zirconia systems containing small to moderate size critical cracks are expected to be improved by a factor of two at most.

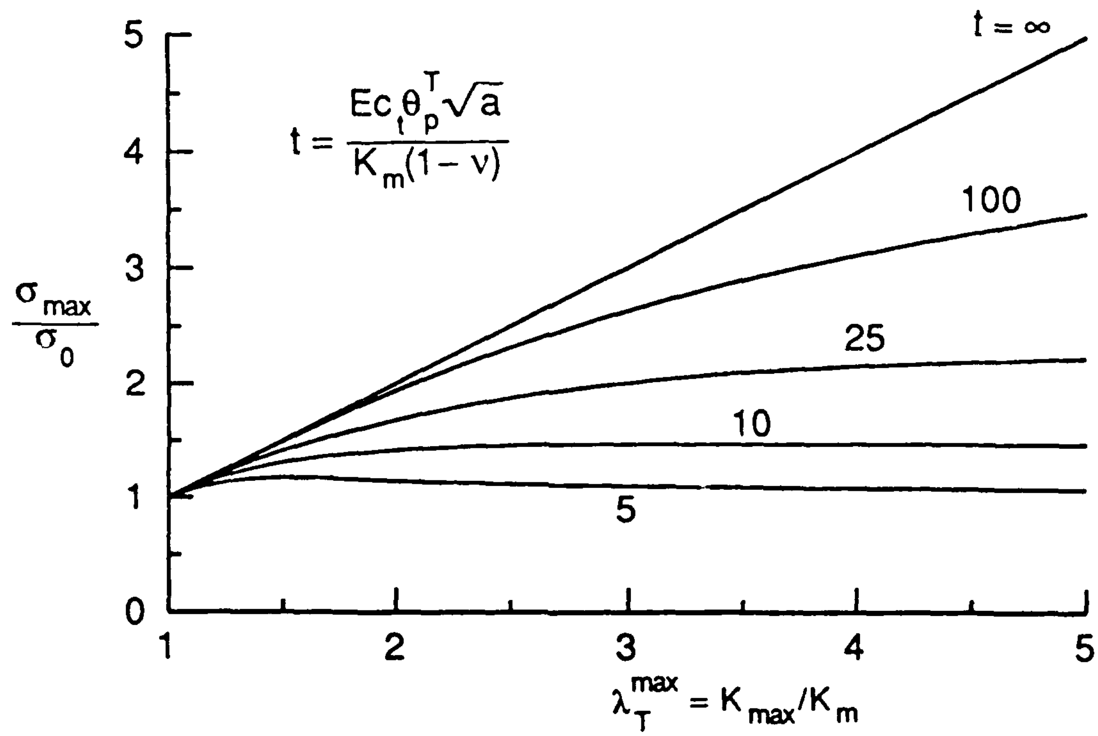

Fig. 4 Peak stress ratios versus peak toughness ratios; various t.

SYNERGISM

This section addresses transient crack growth in brittle ceramics reinforced with *both* phase-transforming zirconia particles and ductile metal particles. We begin with some preliminary remarks on "particulate" toughening due to just ductile metal particle reinforcements.

Under special conditions, it has been observed experimentally that ductile metal particle reinforcements can toughen normally brittle ceramic materials by bridging the faces directly behind an advancing crack tip[9]. This phenomena has been studied theoretically[10] by assuming bridging particles that obeyed a linear-elastic ideally-plastic deformation rule. This study invokes the simplified form of that analysis which neglects the elastic deformation of the particles and adopts a rigid ideally-plastic inclusion model with the "flow" stress S (estimated 5 -7 times the bulk metal yield stress). Additionally, the particles are assumed to rupture when the crack-opening displacement (COD) reaches the critical value $2v_f$.

During steady-state crack growth, the advancing tip is bridged by a small-scale particle strip of length l_p that exerts a uniform restraining traction c_pS, where c_p is the ductile-particle volume fraction. The enhanced toughness is expressed in terms of the *modified* toughness ratio

$$\Lambda_p^{ss} = \frac{K_{ss}}{K_m\sqrt{1-c_p}} \qquad (5)$$

where the steady-state "applied" $K = K_{ss}$ and K_m is the critical value for fracture in the matrix ceramic. (The square-root factor arises from the decrease in crack front due to the presence of the ductile particles at the tip.) An application of the J-integral provides the compact formula

$$\Lambda_p^{ss} = \left[1 + \frac{2\,ESc_pv_f}{K_m(1-\nu^2)(1-c_p)}\right]^{1/2} \qquad (6)$$

for steady plane-strain crack growth, where E and ν are the elastic constants of the composite. An alternative calculation of the stress-intensity factor provides the relation

$$\Lambda_p^{ss} = 1 + \frac{c_pS}{K_m}\sqrt{\frac{8l_p}{\pi}} \qquad (7)$$

between Λ_p^{ss} and the bridge length l_p. Experimental measurements[9] of (5) disagree with the predictions of (6) by about a factor of two, while satisfactory data to judge the adequacy of (7) has not yet been collected. Nevertheless, for the purposes of studying the interactions, the validity of both (6) and (7) is accepted for the remainder of this paper.

The interaction of zirconia/ductile-metal particle reinforcements during steady-state crack growth has recently been studied[11] by considering the self-similar advance of a crack which is bridged by a strip of "very" ductile particles and whose faces are bordered by small-scale wake regions of supercritically transformed material. In order to assess the overall toughness with respect to the effects of the individual mechanisms, the "component" toughness ratios Λ_p^{ss} and λ_T^{ss} due to the separate, isolated systems are taken as prescribed material constants. This approach is followed throughout this section.

The steady-state crack results are expressed in terms of an overall *modified* toughness ratio Λ_{ss}, given by the form of (5), which is a function of Λ_p^{ss}, λ_T^{ss} (and hence ω via Figure 1), and the "natural" coupling parameter

$$\rho = \frac{c_pS(1+\nu)}{\sigma_m^c} \qquad (8)$$

A more convenient alternative coupling parameter is provided by the combination

$$\eta = \frac{H_T(1-c_p)}{l_p} \tag{9}$$

where H_T and l_p are the "uncoupled" transformation wake height and bridge length. Analytical arguments were developed to show that the limiting values of Λ_{ss} are given by the formulas

$$\Lambda_{ss} = \lambda_T^{ss}\Lambda_p^{ss} \qquad \rho,\eta \to \infty \tag{10}$$

$$\Lambda_{ss} = \left[(\lambda_T^{ss})^2 + (\Lambda_p^{ss})^2 - 1\right]^{1/2} \qquad \rho,\eta \to 0 \tag{11}$$

Remarkably, it was observed that in some instances values of the parameters $\rho \approx 1$ and $\eta \approx .1$ yielded overall Λ_{ss}'s almost as high as those predicted by synergistic product rule (10). This is an important result since typical wake heights H_T are ≤ 50 μm, while bridge lengths l_p as long as 200 μm have been reported. Thus, synergistic interactions appear possible in mixtures of accessible separate systems.

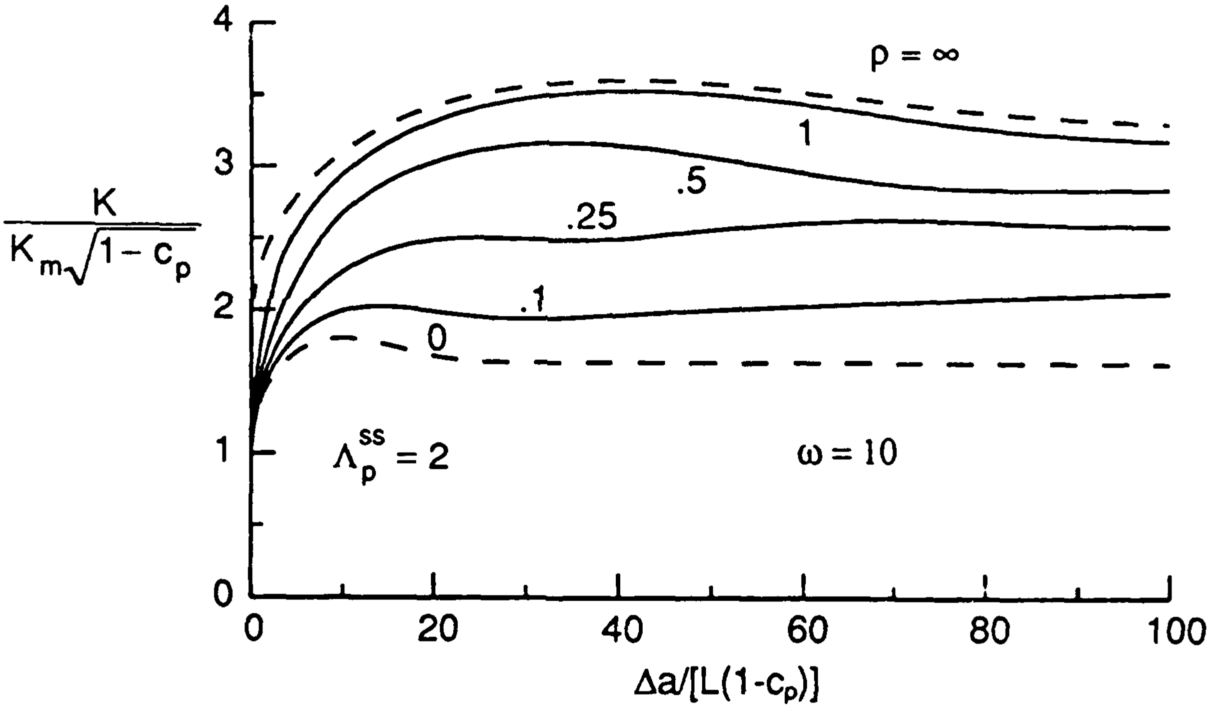

Fig. 5 Toughness ratios versus normalized crack growth; $\omega = 10$, $\Lambda_p^{ss} = 2$, various ρ.

More recently, transient interactions in the "combined" material have been studied[12] by modeling the growth of semi-infinite pre-existing cracks. This was accomplished by adapting the growing crack model described in the section on transformation toughening to include the effects of "very" ductile crack-bridging particles. The component toughness ratios λ_T^{max}, λ_T^{ss} (ω via Figure 1) and Λ_p^{ss}, along with the "natural" coupling parameter ρ were again taken as prescribed constants. The *modified* characteristic length $L_1 = L(1-c_p)$, where L is given by (2), and the overall instantaneous toughness ratio

$$\Lambda = \frac{K}{K_m\sqrt{1-c_p}} \tag{12}$$

emerged from the analysis. Plots of Λ versus $\Delta a/L_1$ are shown in Figure 5 for $\Lambda_p^{ss}=2$, $\omega = 10$ ($\lambda_T^{max}=1.8$), and a variety of ρ. The presence of the transformations causes each of the Λ's to reach a peak value Λ_{max} at finite amounts of crack advance. For "strong" interactions (i.e. large ρ), Λ_{max} can greatly exceed Λ_{ss} since the bridging particles provide substantial reinforcement just after the onset of growth. The additional applied stress necessary to overcome the bridging effects acts to expand the transformed region and thus amplify the peak in the transformation toughness component. Conversely, for "weak" interactions (i.e. $\rho \approx 0$), bridging effects only become important after the crack has grown through the peak in transformation toughness. As a result, each mechanism is close to its steady-state level when interactions take place, so that Λ_{max} just barely exceeds Λ_{ss}. The limiting values of Λ_{max} for strong interactions are given by the approximate formula

$$\Lambda_{max} \approx \lambda_T^{max}\Lambda_p^{ss} \qquad \rho, \eta \to \infty \tag{13}$$

while for the limit of vanishing interaction strength (i.e. $\rho,\eta \to 0$) $\Lambda_{max} = \Lambda_{ss}$ and the exact formula (11) remains in effect.

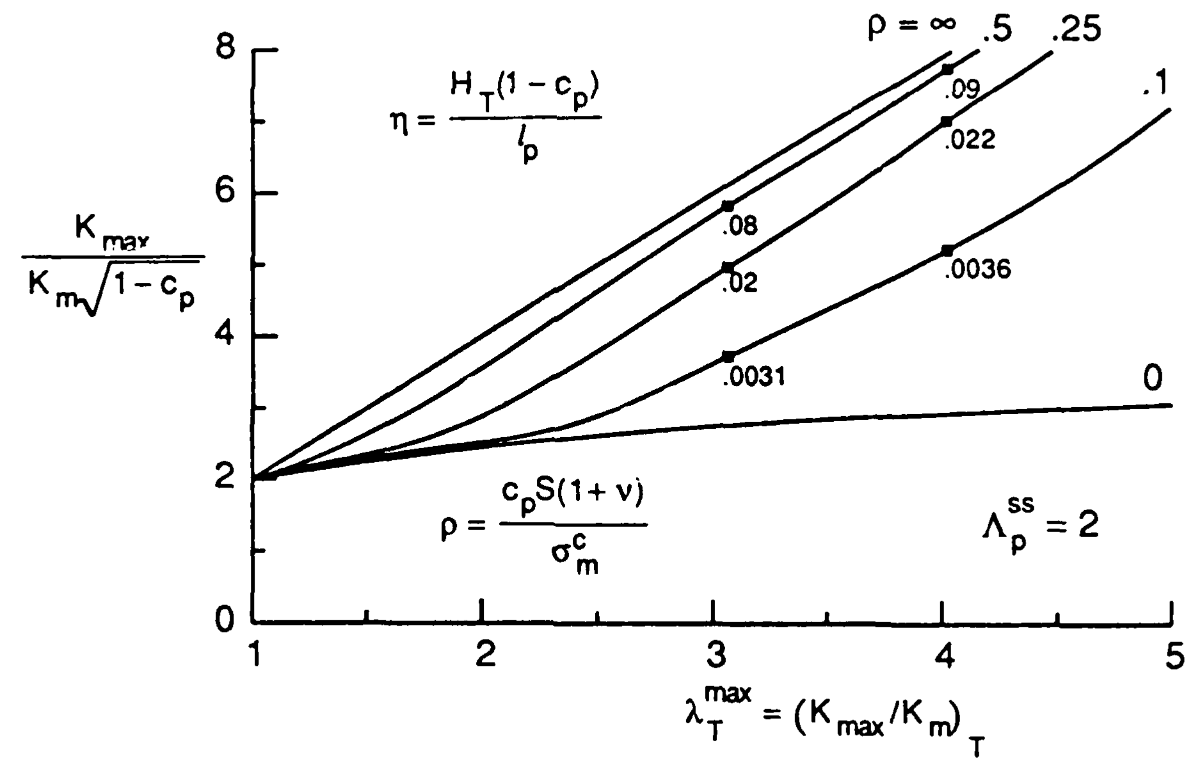

Fig. 6 Peak overall toughness ratios versus λ_T^{max}; $\Lambda_p^{ss} = 2$, various ρ and η.

We concluue with Figure 6 where Λ_{max} has been plotted versus λ_T^{max} for $\Lambda_p^{ss}=2$, and a variety of ρ. Some corresponding values of the alternative parameter η have also been shown. Importantly, peak toughening may occur for values of the parameter η smaller than the steady-state theory predicts. Thus, synergistic interactions have the allure of making combined reinforcement a potent toughening method. A similar study for synergism in strengthening of finite cracks has also been made by Stump (1990).

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