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THE DEVELOPMENT OF A CONSTITUTIVE RELATION
FOR
NEOPRENE BALLOON FILM

HAROLD ALEXANDER
Department of Aeronautics and Astronautics

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Aerospace Instrumentation Laboratory

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Harold Alexander

New York University
New York

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ABSTRACT

The physical and mechanical characteristics of neoprene film, a commonly used expandable-type balloon skin material, are presented for the purpose of developing constitutive equations that can be used in balloon analyses.

"Day" balloon film is found to be an almost perfectly elastic material through an extremely large range of deformations. A survey of previously proposed elastic constitutive equations for rubber-like materials is presented and all are found to be inadequate for neoprene. Consequently, a new constitutive relation based on the attempts of previous investigators and experiments performed by the author is proposed. It is shown to be a generalization of most earlier theories.

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THE DEVELOPMENT OF A CONSTITUTIVE RELATION
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NEOPRENE BALLOON FILM

by

Harold Alexander

I. INTRODUCTION

Today, almost two centuries after the first Montgolfier balloon flight, spurred by the space program, there is a renewed interest in high altitude balloons. As a result of recent improvements in manufacturing and flight techniques, balloons have found new applications in meteorological studies, cosmic ray studies, astronomy, space biology and the testing of new concepts in space vehicles. The balloon provides a stable, slow moving, high altitude platform serving a multitude of scientific purposes.

To reach high altitudes it is necessary to maintain a substantial amount of lift. High lift is attained by having a large balloon volume with the least weight of inflation gas and structure. Expandable-type balloons achieve a large volume by allowing the balloon skin to distend during ascent.

In all balloons, the balloon skin is not just a gas barrier, it is also the structure that supports the weight of the balloon and payload. The analysis of this structure is necessary for successful ballooning. For example, during the flight of an expandable-type balloon,

* This report constitutes part of a dissertation submitted by the author to New York University in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

the skin undergoes very large deformations and often ruptures before the balloon reaches float equilibrium. Elimination of this type of failure requires a knowledge of the mechanical properties of the balloon skin material through a wide range of extensions and temperatures as well as the analysis of balloons subjected to very large deformations (of the order of magnitude of the balloon radius).

In the present report, an elastic constitutive relation is proposed for neoprene, a commonly used balloon skin material. In subsequent reports, this constitutive relation will be utilized in the analysis of various balloon and balloon related problems.

II. MATERIAL PROPERTIES

Expandable-type balloons, due to the nature of their operation, must be made of highly extensible materials. Consequently, this type of balloon is usually produced from one of the high polymeric rubber-like materials known as elastomers. In this section, a short discussion will be presented on the physical and mechanical characteristics of elastomers.

1. PHYSICAL CHARACTERISTICS

Elastomers are composed of long, flexible chain molecules having weak intermolecular forces. These molecules are interconnected by primary valence bonds at suitable intervals so as to form a three dimensional network. Due to their structure, these chains have the ability to uncoil and recoil rapidly in an almost completely elastic manner through a rather large range of ambient temperatures [1].

The necessary cross-linking in the formation of a network of chain molecules is usually introduced through the process of vulcanization. This is a chemical reaction of the elastomer with some vulcanizing agent which results in the formation of the interconnecting bonds mentioned above. For natural and butyl rubbers the vulcanizing agent is sulfur. For neoprene (polychloroprene), the material given the greatest attention in this treatise due to its almost universal use in expandable-type balloons, the most often used vulcanizing agents are magnesium oxide and zinc oxide.

In addition to uncoiling and recoiling, the molecules of an elastomer often undergo a partial crystallization induced by stretching. This has a considerable bearing on the stress-strain curve and on the rupture and tear phenomena exhibited by the elastomer. The crystallization occurring upon stretching is proportional to the absolute temperature of the sample.

2. MECHANICAL CHARACTERISTICS

All elastomers are viscoelastic materials. However, depending upon the temperature and stress level, their time dependent component of response is of greater or lesser importance. As is pointed out by Tobolsky [2], there are four well-defined temperature regions of viscoelastic behavior (see Fig. 1).

- a) A low temperature glassy region (for neoprene, below approx. -100°F) in which the stress-strain ratio is very large. The creep effects are not very pronounced in this region so that one may speak of a quasi-static glassy state.
- b) A transition region (for neoprene, between approx. -70° and -100°F) in which the stress-strain ratio changes rapidly with time and temperature. The creep effects are produced by short-range motions of relatively small segments of the polymer molecules. At a set temperature in the transition region, the mechanical response of the material to stress can be considered to be time independent due to the very short time involved in the total response.

c) A quasi-static "rubbery plateau" region (for neoprene, between 170° and -70°F) in which the stress-strain ratio is comparatively low and changes quite slowly with time and temperature. At a set temperature in this region the response can be considered as time independent due to its quasi-static nature (long times of response).

d) A flow region (for neoprene, above 170°F) in which the stress-strain ratio changes very rapidly with time from a value comparable to that of the "rubbery plateau" region to zero. In this region, there is a true viscous flow occurring.

In regions a, b and c, a time independent elastic theory yields a fairly accurate result for the mechanical response of the material as long as one is not concerned with the creep effects in the transition region. However, any theory proposed, in order to be useful in the region of large deformations encountered in balloons, must be able to account for the strain hardening effect occurring due to partial crystallization.

The neoprene used in the production of expandable-type balloons is in region c at launch temperature. Since throughout a balloon flight, the temperature is lower than the launch temperature, it seems possible to analyze the balloon structure using an elastic theory. This turns out to be the case with "day" balloons, i.e. those used on day flights when the temperature of the balloon does not go into the glassy region. However, for night flights a different type of balloon material is used due to the fact that the skin material of a

"day" balloon becomes brittle at the cold temperature encountered during such a flight. "Night" balloons are produced by the addition of a plasticizer to the neoprene. This has the effect of transposing the film properties with respect to the above mentioned temperature regions. At launch temperature, a "night" balloon is on the boundary of regions c and d. Consequently, a viscoelastic law must be used in analyzing the flight of such a balloon.

Since "day" balloons have been shown to undergo only elastic deformations, this report will be devoted to the development of a suitable elastic constitutive relation for "day" balloon film.

III. ELASTIC BEHAVIOR

Over the past three decades a considerable amount of effort has been directed towards finding the constitutive relation for rubber and rubber-like materials. Hooke's law, the usually used constitutive relation for elastic materials, is not suitable for rubbers. Hooke's law may be considered as a linear approximation to a much more general stress-strain relationship and is usually valid only in a small range of deformations. Rubber-like materials usually experience large deformations under loading and the stress-strain relationship is not linear.

Most of the research on large deformation theories for highly extensible materials seems to have been performed in the United States and the United Kingdom. A large part of these results is described in Ref. [3]. Another "school" of large deformation elasticity has formed in the U.S.S.R. along the lines of the formulation developed by Kappus [4] and presented by Novozhilov [5], [6]. In the following, using the general theory as a guide, the constitutive relations previously proposed by various investigators are presented and critically analyzed with the aim in view of finding the constitutive relation for neoprene film. Since all of the previous theories are found to be inadequate, a new constitutive relation based on experiments performed by the author on neoprene "day" balloon film is proposed and is shown to be a generalization of most earlier theories.

Preliminary to this critical analysis and development of a new constitutive relation, the general constitutive relation is developed

along the lines of the theory presented by Novozhilov [5], [6]. During this development, some remarks are made about the definitions of stress and strain and how they fit into the formulation.

1. DEFINITION OF STRAIN

Consider a point of a body which, in the undeformed state, lies at (x, y, z) in a rectangular, cartesian coordinate system fixed in space (see Fig. 2). Suppose, that due to the deformation, the point (x, y, z) moves to $(x+u, y+v, z+w) = (\xi, \eta, \zeta)$. An element of the material in the undeformed state has length ds and direction cosines (l, m, n) . The same element in the deformed state has length ds' and direction cosines (l', m', n') . The direction cosines are defined,

$$l = \frac{dx}{ds} \quad ; \quad m = \frac{dy}{ds} \quad ; \quad n = \frac{dz}{ds}$$

and

$$l' = \frac{d\xi}{ds'} \quad ; \quad m' = \frac{d\eta}{ds'} \quad ; \quad n' = \frac{d\zeta}{ds'} \tag{1.1}$$

By the Pythagorean Theorem,

$$\begin{aligned} ds^2 &= dx^2 + dy^2 + dz^2 \\ ds'^2 &= d\xi^2 + d\eta^2 + d\zeta^2 \end{aligned} \tag{1.2}$$

and dividing the first of (1.2) by ds^2 and the second by ds'^2 yields, through the use of (1.1),

$$\begin{aligned} l^2 + m^2 + n^2 &= 1 \\ (l')^2 + (m')^2 + (n')^2 &= 1 \end{aligned} \tag{1.3}$$

The quantities ξ , η and ζ are functions of x , y and z . Using the chain rule, it follows that

$$\begin{aligned}
d\xi &= \xi_{,x} dx + \xi_{,y} dy + \xi_{,z} dz \\
dr &= r_{,x} dx + r_{,y} dy + r_{,z} dz \\
d\zeta &= \zeta_{,x} dx + \zeta_{,y} dy + \zeta_{,z} dz
\end{aligned}
\tag{1.4}$$

Dividing the first of equ. (1.4) by ds' yields,

$$\frac{d\xi}{ds'} = \ell' = \frac{ds}{ds'} (\xi_{,x} \ell + \xi_{,y} m + \xi_{,z} n)$$

Similarly, the following equations are obtained:

$$\frac{dr}{ds'} = m' = \frac{ds}{ds'} (r_{,x} \ell + r_{,y} m + r_{,z} n) \tag{1.5}$$

$$\frac{d\zeta}{ds'} = n' = \frac{ds}{ds'} (\zeta_{,x} \ell + \zeta_{,y} m + \zeta_{,z} n)$$

Substituting (1.5) into the second of (1.3) yields

$$\begin{aligned}
\left(\frac{ds'}{ds}\right)^2 &= (\xi_{,x}^2 + r_{,x}^2 + \zeta_{,x}^2) \ell^2 + (\xi_{,y}^2 + r_{,y}^2 + \zeta_{,y}^2) m^2 \\
&+ (\xi_{,z}^2 + r_{,z}^2 + \zeta_{,z}^2) n^2 + 2(\xi_{,y} \xi_{,z} + r_{,y} r_{,z} + \zeta_{,y} \zeta_{,z}) mn \\
&+ 2(\xi_{,z} \xi_{,x} + r_{,z} r_{,x} + \zeta_{,z} \zeta_{,x}) n\ell + 2(\xi_{,x} \xi_{,y} + r_{,x} r_{,y} + \zeta_{,x} \zeta_{,y}) \ell m
\end{aligned}
\tag{1.6}$$

The quantity ds'/ds is defined as the total extension of the original element. A constitutive relation is usually expressed as a relationship between stress and strain; where the strain is defined as some function of ds'/ds . There have been many strain definitions proposed. Some of the more common ones are presented below.

The Lagrangian Strain measure, given by Cauchy [7] is the one most used in the analysis of large elastic deformations. It can be expressed as,

$$\frac{1}{2} \left\{ \left(\frac{ds'}{ds} \right)^2 - 1 \right\} \quad (1.7)$$

Using equ. (1.6), and noting that

$$\xi = x+u, \quad \eta = y+v, \quad \zeta = z+w \quad (1.8)$$

yields

$$\begin{aligned} \epsilon^L = \frac{1}{2} \left\{ \left(\frac{ds'}{ds} \right)^2 - 1 \right\} &= \epsilon_{xx}^L \ell'^2 + \epsilon_{yy}^L m^2 + \epsilon_{zz}^L n^2 \\ &+ \epsilon_{xy}^L \ell'm + \epsilon_{xz}^L \ell'n + \epsilon_{yz}^L m'n \end{aligned} \quad (1.9)$$

where

$$\epsilon_{xx}^L = u_{,x} + \frac{1}{2}(u_{,x}^2 + v_{,x}^2 + w_{,x}^2), \text{ etc.}$$

$$\epsilon_{yz}^L = w_{,y} + v_{,z} + u_{,y}u_{,z} + v_{,y}v_{,z} + w_{,y}w_{,z}, \text{ etc.}$$

It was suggested by Coker and Filon [8] that the strain should be defined in terms of the displacements as a function of the deformed state of the body. This leads to the Almansi strain measure (sometimes called the Eulerian strain measure due to the fact that it is referred to an Eulerian coordinate system),

$$\begin{aligned} \epsilon^E = \frac{1}{2} \left\{ 1 - \left(\frac{ds}{ds'} \right)^2 \right\} &= \epsilon_{\xi\xi}^E (\ell')^2 + \epsilon_{\eta\eta}^E (m')^2 + \epsilon_{\zeta\zeta}^E (n')^2 \\ &+ \epsilon_{\xi\eta}^E \ell'm' + \epsilon_{\xi\zeta}^E \ell'n' + \epsilon_{\eta\zeta}^E m'n' \end{aligned} \quad (1.10)$$

where

$$\epsilon_{\xi\xi}^E = u_{,\xi} - \frac{1}{2}(u_{,\xi}^2 + v_{,\xi}^2 + w_{,\xi}^2), \text{ etc.}$$

$$\epsilon_{\eta\zeta}^E = v_{,\zeta} + w_{,\eta} - u_{,\eta}u_{,\zeta} - v_{,\eta}v_{,\zeta} - w_{,\eta}w_{,\zeta}, \text{ etc.} \quad (1.11)$$

There have been many other strain measures suggested [9]. For example:

Swainger's Measure:

$$\epsilon^s = 1 - \frac{ds}{ds'} \quad (1.12)$$

Kuhn's Measure:

$$\epsilon^k = \frac{1}{3} \left\{ \left(\frac{ds'}{ds} \right)^2 - \frac{ds}{ds'} \right\} \quad (1.13)$$

Simple Strain Measure: (stretch)

$$\epsilon^{\text{sim}} = \frac{ds'}{ds} - 1 \quad (1.14)$$

and the Ludwik-Hencky Strain Measure:

$$\epsilon^{\text{LH}} = n \frac{ds'}{ds} \quad (1.15)$$

The Ludwik-Hencky Strain Measure has become very popular in the large deformation theories of metallic solids. With these materials a linear relationship may often be assumed to exist between true stress and Ludwik-Hencky strain ([10], p. 76).

It can be seen from the above that the strain definition is really quite arbitrary and may often be chosen only to simplify the constitutive relation. For the purposes of this treatise, since it is planned to obtain a constitutive relation from experimental data, there is no immediately obvious preference for any one strain measure. Therefore, the fundamental quantity ds'/ds , the total extension, will be used.

Defining λ_t as the total extension, equ. (1.6) becomes,

$$\lambda_t^2 = \lambda_{xx}^2 + \lambda_{yy}^2 + \lambda_{zz}^2 + 2\lambda_{yz}^2 + 2\lambda_{zx}^2 + 2\lambda_{xy}^2 \quad (1.16)$$

where,

$$\begin{aligned}
 \lambda_{xx}^2 &= \xi_x^2 + \eta_x^2 + \zeta_x^2 = (1+u_x)^2 + (v_x)^2 + (w_x)^2 = 1 + 2\epsilon_{xx}^L \\
 \lambda_{yy}^2 &= \xi_y^2 + \eta_y^2 + \zeta_y^2 = (1+v_y)^2 + (u_y)^2 + (w_y)^2 = 1 + 2\epsilon_{yy}^L \\
 \lambda_{zz}^2 &= \xi_z^2 + \eta_z^2 + \zeta_z^2 = (1+w_z)^2 + (u_z)^2 + (v_z)^2 = 1 + 2\epsilon_{zz}^L \\
 \lambda_{yz}^2 &= \xi_y \xi_z + \eta_y \eta_z + \zeta_y \zeta_z = w_y v_z + u_y u_z + v_y v_z + w_y w_z = \epsilon_{yz}^L \\
 \lambda_{zx}^2 &= \xi_z \xi_x + \eta_z \eta_x + \zeta_z \zeta_x = w_x v_z + u_x u_z + v_x v_z + w_x w_z = \epsilon_{zx}^L \\
 \lambda_{xy}^2 &= \xi_x \xi_y + \eta_x \eta_y + \zeta_x \zeta_y = u_y v_x + u_x u_y + v_x v_y + w_x w_y = \epsilon_{xy}^L
 \end{aligned} \tag{1.17}$$

The square of the total extension, λ_t^2 , is a symmetric tensor. By diagonalizing λ_t^2 , (expressing it in terms of a principal axis system) it is possible to determine its three invariants. The diagonalization process leads to the following set of three homogeneous equations:

$$\begin{aligned}
 (\lambda_{xx}^2 - \lambda^2)l + \lambda_{xy}^2 m + \lambda_{zx}^2 n &= 0 \\
 \lambda_{xy}^2 l + (\lambda_{yy}^2 - \lambda^2)m + \lambda_{yz}^2 n &= 0 \\
 \lambda_{zx}^2 l + \lambda_{yz}^2 m + (\lambda_{zz}^2 - \lambda^2)n &= 0
 \end{aligned} \tag{1.18}$$

where λ^2 is the eigenvalue of λ_t^2 .

Since the trivial solution, $l=m=n=0$, violates the first of (1.3), equations (1.18) are satisfied only if

$$(\lambda^2)^3 - I_1(\lambda^2) + I_2\lambda^2 - I_3 = 0 \tag{1.19}$$

where

$$\begin{aligned}
 I_1 &= \lambda_{xx}^2 + \lambda_{yy}^2 + \lambda_{zz}^2 \\
 I_2 &= \lambda_{xx}^2 \lambda_{yy}^2 + \lambda_{xx}^2 \lambda_{zz}^2 + \lambda_{yy}^2 \lambda_{zz}^2 - \lambda_{xy}^4 - \lambda_{xz}^4 - \lambda_{yz}^4 \\
 I_3 &= \lambda_{xx}^2 \lambda_{yy}^2 \lambda_{zz}^2 - \lambda_{xx}^2 \lambda_{yz}^4 - \lambda_{yy}^2 \lambda_{xz}^4 - \lambda_{zz}^2 \lambda_{xy}^4 + 2 \lambda_{xy}^2 \lambda_{xz}^2 \lambda_{yz}^2
 \end{aligned} \tag{1.20}$$

The roots of equ. (1.19), λ_1 , λ_2 and λ_3 , are the principal extensions. They are independent of the orientation of the cartesian axis system. The coefficients, I_1 , I_2 and I_3 , are, therefore, also independent of the axis system orientation and are the invariants of the deformation. In terms of the principal extensions, they become

$$\begin{aligned}
 I_1 &= \lambda_1^2 + \lambda_2^2 + \lambda_3^2 \\
 I_2 &= \lambda_1^2 \lambda_2^2 + \lambda_2^2 \lambda_3^2 + \lambda_3^2 \lambda_1^2 \\
 I_3 &= \lambda_1^2 \lambda_2^2 \lambda_3^2
 \end{aligned} \tag{1.21}$$

It can be shown [6], that the invariants of the Lagrangian strain, ϵ^L , are:

$$\begin{aligned}
 I_1^L &= \epsilon_{xx}^L + \epsilon_{yy}^L + \epsilon_{zz}^L \\
 I_2^L &= \epsilon_{xx}^L \epsilon_{yy}^L + \epsilon_{yy}^L \epsilon_{zz}^L + \epsilon_{zz}^L \epsilon_{xx}^L - \frac{1}{4} \left[(\epsilon_{xy}^L)^2 + (\epsilon_{xz}^L)^2 + (\epsilon_{yz}^L)^2 \right] \\
 I_3^L &= \epsilon_{xx}^L \epsilon_{yy}^L \epsilon_{zz}^L - \frac{1}{4} \left[\epsilon_{xx}^L (\epsilon_{yz}^L)^2 + \epsilon_{yy}^L (\epsilon_{xz}^L)^2 + \epsilon_{zz}^L (\epsilon_{xy}^L)^2 - \epsilon_{xy}^L \epsilon_{xz}^L \epsilon_{yz}^L \right]
 \end{aligned} \tag{1.22}$$

Since both the I_1^L 's and the I_1 's are invariants of the same deformation they should be dependent. This can be proven by relating

the terms of ϵ^L to those of λ_t^2 through (1.17) and then equating like terms in (1.20) and (1.22), yielding

$$\begin{aligned} I_1^L &= \frac{(I_1 - 3)}{2} \\ I_2^L &= \frac{1}{2} [(I_2 - 3) - (I_1 - 3)] \\ I_3^L &= \frac{1}{8} [(I_3 - 1) - (I_2 - 3) + (I_1 - 3)] \end{aligned} \quad (1.23)$$

2. THE STRAIN ENERGY DENSITY AND THE RESULTING CONSTITUTIVE RELATION

Throughout this report it is assumed that the material is elastic, homogeneous and isotropic in the undeformed state.

For an elastic material, the work required to deform the body, which is equal to the internal strain energy, is a function of only the total deformation. This can be expressed as a strain energy per unit undeformed volume (strain energy density),

$$W = W(\lambda_{xx}, \lambda_{yy}, \lambda_{zz}, \lambda_{xy}, \lambda_{yz}, \lambda_{zx}) \quad (2.1)$$

Due to the assumption of initial isotropy, there is a further constraint on the form of the strain energy density. W must be independent of a coordinate rotation. It is therefore convenient to express W in terms of the three independent strain invariants, I_1 , I_2 and I_3 , which are unchanged by a coordinate rotation.

$$W = W(I_1(\lambda_{xx}, \dots, \lambda_{zx}), I_2(\lambda_{xx}, \dots, \lambda_{zx}), I_3(\lambda_{xx}, \dots, \lambda_{zx})) \quad (2.2)$$

The differential change in the strain energy density caused by a differential change in displacements is derived by Novozhilov ([6], chapt. III) in terms of the Lagrangian strain as

$$dW = \sigma_{xx}^* d\epsilon_{xx}^L + \sigma_{yy}^* d\epsilon_{yy}^L + \sigma_{zz}^* d\epsilon_{zz}^L + \sigma_{xy}^* d\epsilon_{xy}^L + \sigma_{yz}^* d\epsilon_{yz}^L + \sigma_{zx}^* d\epsilon_{zx}^L \quad (2.3)$$

where σ_{ij}^* , the "generalized stresses" referred to the dimensions of an element of volume before the deformation, are defined,

$$\sigma_{ij}^* = \frac{S_i^*}{S_i} \frac{\sigma_{ij}}{1+E_j} \quad (\text{no summation}) \quad (2.4)$$

with,

$$E_j = \lambda_{jj} - 1 \quad (\text{no summation}) \quad (2.5)$$

and

$$\frac{S_i^*}{S_i} = \sqrt{\lambda_{i+1, i+1}^2 \lambda_{i-1, i-1}^2 - \lambda_{i-1, i+1}^4} \quad (2.6)$$

The indices of (2.6) are cyclic (1,2,3,1,2,3,...) and σ_{ij} are the true stresses referred to the deformed element.

By integrating equ. (2.3) the work performed in the deformation of an initially rectangular elementary parallelepiped with the sides dx, dy, dz (referred to its volume in the undeformed state) can be expressed as,

$$W = \int_{\epsilon_{ij}=0}^L (c_{xx}^* d\epsilon_{xx}^L + \dots + c_{zx}^* d\epsilon_{zx}^L) \quad (2.7)$$

Since the material is ideally elastic, the integral (2.7) should be independent of the path of deformation and the integrand a total

differential. This leads to the relationship,

$$\sigma_{ij}^* = \frac{\partial W}{\partial \epsilon_{ij}^L} \quad (2.8)$$

Eliminating S_i^*/S_i and E_j from (2.4) through the use of (2.5) and (2.6) yields,

$$\sigma_{ij}^* = \frac{\sqrt{\lambda_{i+1,i+1}^2 \lambda_{i-1,i-1}^2 - \lambda_{i-1,i+1}^4}}{\lambda_{ij}} \sigma_{ij} \quad (2.9)$$

Since ϵ^L is a symmetric tensor, equ. (2.8) implies that

$$\sigma_{ij}^* = \sigma_{ji}^* \quad (2.10)$$

However, in view of equ. (2.10), equ. (2.9) implies that

$$\sigma_{ij} \neq \sigma_{ji} \quad (2.11)$$

a. Constitutive Relation for a Compressible Material

Through the use of the chain rule, equ. (2.8) can be rewritten

$$\sigma_{ij}^* = \frac{\partial W}{\partial \lambda_{ij}} \frac{d\lambda_{ij}}{d\epsilon_{ij}^L} \quad (2.12)$$

since for set values of i and j , it can be seen from (1.17) that $\epsilon_{ij}^L = f(\lambda_{ij})$ only. Differentiation of (1.17) yields

$$\frac{d\lambda_{ij}}{d\epsilon_{ij}^L} = \frac{1}{\lambda_{ij}(2-\delta_{ij})} \quad (2.13)$$

Again through the use of the chain rule, $\partial W/\partial \lambda_{ij}$ can be put in the form,

$$\frac{\partial W}{\partial \lambda_{ij}} = \frac{\partial W}{\partial I_1} \frac{\partial I_1}{\partial \lambda_{ij}} + \frac{\partial W}{\partial I_2} \frac{\partial I_2}{\partial \lambda_{ij}} + \frac{\partial W}{\partial I_3} \frac{\partial I_3}{\partial \lambda_{ij}} \quad (2.14)$$

The terms, $\partial I_k / \partial \lambda_{ij}$, can be evaluated by differentiating (1.20) with respect to λ_{ij} ,

$$\begin{aligned} \frac{\partial I_1}{\partial \lambda_{ij}} &= 2\lambda_{ij} \delta_{ij} \\ \frac{\partial I_2}{\partial \lambda_{ij}} &= 2\lambda_{ij} \left\{ \delta_{ij} (\lambda_{i+1, j+1}^2 + \lambda_{i-1, j-1}^2 + 2\lambda_{ij}^2) - 2\lambda_{ij}^2 \right\} \end{aligned} \quad (2.15)$$

$$\begin{aligned} \frac{\partial I_3}{\partial \lambda_{ij}} &= 2\lambda_{ij} \left\{ \delta_{ij} (-\lambda_{i+1, j+1}^4 + \lambda_{i-1, j-1}^4 - \lambda_{i-1, j+1}^4 + 2\lambda_{ij}^2 \lambda_{i-1, j+1}^2) \right. \\ &\quad \left. + 2\lambda_{i+1, j+1}^2 \lambda_{i-1, j-1}^2 - 2\lambda_{ij}^2 \lambda_{i-1, j+1}^2 \right\} \end{aligned}$$

Therefore, using equations (2.13), (2.14) and (2.15) to eliminate $d\lambda_{ij}/d\epsilon_{ij}^L$ and $\partial W / \partial \lambda_{ij}$ from equ. (2.12), the constitutive equation in terms of the strain energy density function for a general compressible solid becomes,

$$\begin{aligned} c_{ij}^* &= (1 + \delta_{ij}) \left\{ \delta_{ij} \left[\frac{\partial W}{\partial I_1} + \left(\delta_{ij} (\lambda_{i+1, j+1}^2 + \lambda_{i-1, j-1}^2 + 2\lambda_{ij}^2) - 2\lambda_{ij}^2 \right) \frac{\partial W}{\partial I_2} \right. \right. \\ &\quad \left. \left. + \left(\delta_{ij} (2\lambda_{ij}^2 \lambda_{i-1, j+1}^2 - \lambda_{i+1, j+1}^2 \lambda_{i-1, j-1}^2 - \lambda_{i-1, j+1}^4) \right) \right. \right. \\ &\quad \left. \left. + 2\lambda_{i+1, j+1}^2 \lambda_{i-1, j-1}^2 - 2\lambda_{ij}^2 \lambda_{i-1, j+1}^2 \right] \frac{\partial W}{\partial I_3} \right\} \end{aligned} \quad (2.16)$$

For the determination of valid forms of the strain energy density function, it will be sufficient to consider only pure homogeneous straining. This type of deformation causes no rotation of the principal

axes of stress and extension. However, the obtained form of W can later be applied to non-homogeneous straining since a rotation of principal axes does not affect the form of the strain energy density function.

The above considerations allow considerable simplification of the constitutive equations (2.16). The inequality (2.11) is now untrue. The true stress tensor, σ_{ij} , becomes symmetric, since areas that were mutually perpendicular in the undeformed state always remain perpendicular. Since there is no rotation of the principal axes due to the deformation, it is sufficiently general to formulate the constitutive equations in terms of principal stresses and extensions; σ_i and λ_i , respectively. Equ. (2.9) becomes

$$\sigma_i = \frac{\lambda_i^2}{\lambda_i \lambda_{i+1} \lambda_{i-1}} \sigma_i^* \quad (2.17)$$

and the constitutive equation (2.16) can be written

$$\sigma_i = \frac{2\lambda_i^2}{\lambda_i \lambda_{i+1} \lambda_{i-1}} \left[\frac{\partial W}{\partial I_1} + (\lambda_{i+1}^2 + \lambda_{i-1}^2) \frac{\partial W}{\partial I_2} + (\lambda_{i+1}^2 \lambda_{i-1}^2) \frac{\partial W}{\partial I_3} \right] \quad (2.18)$$

or, noting the definitions of the invariants (1.21),

$$\sigma_i = \frac{2}{\sqrt{I_3}} \left[\lambda_i^2 \frac{\partial W}{\partial I_1} - \frac{I_3}{\lambda_i^2} \frac{\partial W}{\partial I_2} + I_3 \frac{\partial W}{\partial I_3} + I_3 \frac{\partial W}{\partial I_3} \right] \quad (2.19)$$

b. Constitutive Relation for an Incompressible Material

A material is considered incompressible if upon straining the volume of every element of the material remains constant. This property seems to be exhibited in most high polymeric materials above the glassy state transition. This assumption of volumetric

constancy can be expressed mathematically by the relationship,

$$\frac{\text{final volume}}{\text{initial volume}} = \sqrt{I_3} = 1 \quad (2.20)$$

Consequently, I_3 is no longer variable and equ. (2.2) becomes

$$W = W(I_1(\lambda_1, \lambda_2, \lambda_3), I_2(\lambda_1, \lambda_2, \lambda_3)) \quad (2.21)$$

where λ_1, λ_2 and λ_3 are no longer independent variables, since they are related through the constraint equation (2.20).

If the material is incompressible, there is no deformation caused by a hydrostatic loading. Therefore, the strain energy is indeterminate by a term that will result from pure hydrostatic stress. This indeterminacy can be circumvented by expressing the constitutive relations in terms of the stress deviator, the only part of the stress that causes deformation.

The stress deviator is defined,

$$s_{ij} = \sigma_{ij} - \frac{1}{3} \delta_{ij} \sigma_{ii} \quad (2.22)$$

where

$$\sigma_{ii} = \sum_{i=1}^3 \sigma_{ij} \quad i=j \quad (2.23)$$

Therefore, equ. (2.22) can be rewritten in a system of principal axes as

$$s_i = \frac{1}{3} (2\sigma_i - \sigma_{i-1} - \sigma_{i+1}) \quad (2.24)$$

The constitutive equation (2.19) becomes, in terms of the stress deviator,

$$s_i = \frac{2}{3I_3} \left\{ (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) \frac{\partial W}{\partial I_1} - \left(\frac{2}{\lambda_i^2} - \frac{1}{\lambda_{i+1}^2} - \frac{1}{\lambda_{i-1}^2} \right) I_3 \frac{\partial W}{\partial I_2} \right\} \quad (2.25)$$

and applying the condition of incompressibility (2.20), eqn. (2.25) becomes

$$s_i = \frac{2}{3} \left\{ (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) \frac{\partial W}{\partial I_1} + \left(\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2 \right) \frac{\partial W}{\partial I_2} \right\} \quad (2.26)$$

Equations (2.26) are the constitutive equations for an initially isotropic, elastic, incompressible material under homogeneous straining. They can be shown to be identical to those derived by Rivlin ([11], equ. (6.5)). For small strains, (2.26) should reduce to Hooke's law for an incompressible material, which can be written as

$$s_i = \frac{2}{3} E e_i \quad (2.27)$$

where e_i , the strain deviator, is defined as

$$e_i = \epsilon_i^L + \frac{1}{3} e \quad (2.28)$$

and

$$e = \sum_{i=1}^3 \epsilon_i^L = I_1^L \quad (2.29)$$

For small strains, $I_1^L = 0$ and therefore (2.27) becomes,

$$s_i = \frac{2}{3} E \epsilon_i^L \quad (2.30)$$

Equ. (2.26) can be rewritten,

$$s_i = \frac{2}{3} \left\{ (3\lambda_i^2 - I_1) \frac{\partial W}{\partial I_1} + (I_2 - 3\lambda_{i-1}^2 \lambda_{i+1}^2) \frac{\partial W}{\partial I_2} \right\} \quad (2.31)$$

or through the use of (1.23) and (1.17),

$$s_i = \frac{2}{3} \left\{ \left[3(1+2\epsilon_i^L) - (2I_1^L + 3) \right] \frac{\partial W}{\partial I_1} + \left[(2I_2^L + 2I_1^L + 3) - 3(1+2\epsilon_{i-1}^L)(1+2\epsilon_{i+1}^L) \right] \frac{\partial W}{\partial I_2} \right\} \quad (2.32)$$

As the strains are made small, terms of $O(\epsilon^L)$ can be neglected and the incompressibility condition becomes,

$$I_1^L = 0 \quad (2.33)$$

Therefore, equ. (2.32) becomes,

$$s_i = \frac{2}{3} \left\{ 6 \left(\frac{\partial W}{\partial I_1} \right)_{\epsilon^L=0} + \left(\frac{\partial W}{\partial I_2} \right)_{\epsilon^L=0} \right\} \epsilon_i^L \quad (2.34)$$

Comparing (2.34) with Hooke's law (2.30), the condition that they are identical for small strains can be written,

$$\left. \frac{\partial W}{\partial I_1} \right|_{I_1=I_2=0} + \left. \frac{\partial W}{\partial I_2} \right|_{I_1=I_2=0} = \frac{E}{6} \quad (2.35)$$

where E, the elastic modulus, should now be thought of as the slope of the uniaxial stress versus strain curve at zero strain,

$$E = \left. \frac{\partial \sigma}{\partial \epsilon} \right|_{\epsilon^L=0} \quad (2.36)$$

Equation (2.35) establishes one condition for determining the strain energy density function. It seemed that an additional condition

could be obtained by equating the strain energy of the Hooke's law formulation with that of (2.26) at small strains. This, however, is not a valid procedure. This can be readily seen if one considers the uniaxial σ^* versus ϵ^L curve for Hooke's law and for a non-linear law, (Fig. 3). While it is true that the slopes of these curves approach the same value as $\epsilon^L \rightarrow 0$, it is certainly not true that the areas under the curves (the strain energies) are equal unless the curvature of the non-linear curve is zero as $\epsilon^L \rightarrow 0$.

Any additional conditions for determining the strain energy density function must be found by matching with experimental data.

3. DISCUSSION OF STRAIN ENERGY DENSITIES PROPOSED IN THE PAST

Upon an examination of the conditions imposed on the form of W for an incompressible material, as were stated in the previous section, it was concluded by Treloar ([12], p. 154) that "the most general form of the stored-energy function for an incompressible isotropic elastic material may be expressed as the sum of a series of terms involving powers of $(I_1 - 3)$ and $(I_2 - 3)$ ".

$$W = \sum_{i=0, j=0}^{\infty} C_{ij} (I_1 - 3)^i (I_2 - 3)^j \quad (3.1)$$

where $(I_1 - 3)$ and $(I_2 - 3)$ vanish for zero strain and $C_{00} = 0$ to insure that $W = 0$ at zero strain.

In this section, equ. (3.1) will be used to present some of the proposed strain energy densities. In this presentation, the ranges of validity and the shortcomings of each theory are critically dis-

cussed. It is shown that W as given in (3.1) seems to be far too restrictive.

a. Statistical Theory

Treloar [13] found, by applying Gaussian statistics to a simple model of a network of long chain molecules, that W can be expressed as

$$W = C_{10} (I_1 - 3) \quad (3.2)$$

This form would seem to be a reasonable first order approximation to (3.1). Using (3.2) the constitutive relation (2.26) becomes,

$$s_i = \frac{2}{3} C_{10} (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) \quad (3.3)$$

Physical significance can be given to the constant, C_{10} , if it is considered that for small strains, (3.3) should reduce to Hooke's law for an incompressible material. Applying equ. (2.35) yields,

$$C_{10} = \frac{E}{6}$$

In a subsequent report [14], Treloar tried to verify this theory with experiments performed on various rubbers. By a proper choice of the constant, C_{10} , he was able to obtain reasonably good correlation between experiments and theory in a small range of deformations for uniaxial and equi-biaxial tension of a sheet (see Figs. 4 and 5). However, it became quite apparent from an examination of the experimental results that the theory did not yield good correlation with experiments at the large strains encountered with neoprene balloons.

b. Mooney Theory

Mooney [15] noted that in the simple shear of a cuboid of rubber,

the shear stress was directly proportional to the shear angle. On the basis of this observation, he hypothesized a stress-strain law that is equivalent to assuming a strain energy density function of the type:

$$W = C_{10} (I_1 - 3) + C_{01} (I_2 - 3) \quad (3.5)$$

Using (3.5), the constitutive relation (2.26) becomes

$$s_i = \frac{2}{3} \left\{ \left(2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2 \right) C_{10} + \left(\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2 \right) C_{01} \right\} \quad (3.6)$$

If, as with the Statistical theory, (3.6) reduces to Hooke's law as the strains are made infinitesimal, then equ. (2.35) yields

$$C_{10} + C_{01} = \frac{E}{6} \quad (3.7)$$

Condition (3.7) together with a matching procedure with experimental data determine the values of C_{10} and C_{01} for any particular type of material.

The Mooney form for the strain energy density function has been used by many theoretical investigators in analyzing problems in large deformation elasticity, since it is a higher order theory that still maintains a certain amount of mathematical simplicity. However, as early as 1951 [16] (also [12] p. 164, [17] p. 299), it was pointed out that although for uniaxial extension and simple shear the Mooney assumption is not a bad approximation for $\lambda < 3$, it gives very poor correlation with biaxial tension experiments and experiments in pure shear.

c. Three Term Theory

Both the Statistical and the Mooney theories are successively higher order expansions of equ. (3.1) which is truncated at an arbitrary point. It would seem that there should be some logical method by which one could determine where to truncate this series. Since equ. (3.1) is an expansion in λ_i , it would seem that retaining only terms of the same order in λ_i would be a consistent method of truncation.

If only terms of $O(\lambda_i^4)$ are retained in (3.1), the strain energy density becomes,

$$W = C_{10} (I_1 - 3) + C_{20} (I_1 - 3)^2 + C_{01} (I_2 - 3) \quad * \quad (3.8)$$

where $(I_1 - 3) = O(\lambda_i^2)$, $(I_1 - 3)^2 = O(\lambda_i^4)$, $(I_2 - 3) = O(\lambda_i^4)$. The constitutive equations (2.26) become

$$s_i = \frac{2}{3} \left((2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) [C_{10} + 2C_{20} (I_1 - 3)] + (\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2) C_{01} \right) \quad (3.9)$$

Applying condition (2.35) at small strains yields

$$C_{10} + C_{01} = \frac{E}{6} \quad (3.10)$$

Treloar [14] performed experiments on thin sheets of an 8% sulphur rubber subjected to uniaxial and equi-biaxial tension. The Statistical theory predicts the results of these experiments with a fair degree of accuracy in a small range of deformations with a proper

* It is of interest to note that Isihara, Mashitsume and Tatibana [18] found this same form for W using network theory with a non-gaussian statistics.

choice of the constant C_{10} . The Mooney theory predicts the results of the uniaxial experiment better than the Statistical theory but is not as accurate for the biaxial experiment. In order to determine the validity of equ. (3.8), the constants C_{10} , C_{20} and C_{01} were evaluated by comparison with the above mentioned biaxial experiments. The results are compared with the predictions of the Mooney and Statistical theories.

For equi-biaxial tension of a thin sheet in the 1 and 2 directions, it is assumed that a state of plane stress exists. Therefore,

$$\sigma_1 = \sigma_2 = \sigma_b ; \sigma_3 = 0 ; \lambda_1 = \lambda_2 = \lambda_b \quad (3.11)$$

and by the incompressibility condition (2.20)

$$\lambda_3 = \frac{1}{\lambda_1 \lambda_2} = \frac{1}{\lambda_b^2} \quad (3.12)$$

The constitutive relation (3.9) becomes,

$$\sigma_b = 2 \left(\lambda_b^2 - \frac{1}{\lambda_b^4} \right) \left\{ C_{10} + 2C_{20} \left(2\lambda_b^2 + \frac{1}{\lambda_b^4} - 3 \right) + C_{01} \lambda_b^2 \right\} \quad (3.13)$$

The condition at small strains (2.35) can be used to eliminate one of the constants from (3.13). The remaining two can be evaluated by choosing two experimental points σ_{b_1} , λ_{b_1} and σ_{b_2} , λ_{b_2} and expressing (3.13) as two simultaneous linear algebraic equations in the two remaining unknown constants.

$$\begin{aligned} 2 \left(2\lambda_{b_1}^2 + \frac{1}{\lambda_{b_1}^4} - 3 \right) C_{20} + \left(\lambda_{b_1}^2 - 1 \right) C_{01} &= \frac{\sigma_{b_1}}{2 \left(\lambda_{b_1}^2 - \frac{1}{\lambda_{b_1}^4} \right)} - \frac{E}{6} \\ 2 \left(2\lambda_{b_2}^2 + \frac{1}{\lambda_{b_2}^4} - 3 \right) C_{20} + \left(\lambda_{b_2}^2 - 1 \right) C_{01} &= \frac{\sigma_{b_2}}{2 \left(\lambda_{b_2}^2 - \frac{1}{\lambda_{b_2}^4} \right)} - \frac{E}{6} \end{aligned} \quad (3.14)$$

Equations (3.14) were solved, yielding the values; $C_{10} = 8.08 \text{ Kg/cm}^2$, $C_{20} = 1.035 \text{ Kg/cm}^2$ and $C_{01} = -4.133 \text{ Kg/cm}^2$. Figure 4 clearly shows that with these values of the constants, the three term theory gives fair correlation with experiments.

For the uniaxial stretching of a thin strip, it is assumed that

$$\sigma_2 = \sigma_3 = 0 \quad ; \quad \sigma_1 = \sigma_u \quad ; \quad \lambda_1 = \lambda_u \quad (3.15)$$

and by considerations of isotropy and the incompressibility condition (2.20)

$$\lambda_2 = \lambda_3 = \frac{1}{\sqrt{\lambda_u}} \quad (3.16)$$

Therefore, the constitutive relation (3.9) becomes,

$$\sigma_u = 2 \left(\lambda_u^2 - \frac{1}{\lambda_u} \right) \left\{ C_{10} + 2C_{20} \left(\lambda_u^2 + \frac{2}{\lambda_u} - 3 \right) + C_{01} \frac{1}{\lambda_u} \right\} \quad (3.17)$$

If the theoretical curve obtained by evaluating (3.17) is compared with the experimental results, the Statistical theory and the Mooney theory (Fig. 5); it is obvious that the three term theory is by far a much poorer representation than either of the other two.

Obviously, the three term theory strain energy density equ. (3.8), is not as good an approximation, for all types of deformation, as those of the Statistical (3.2) or Mooney (3.5) theories. If (3.1) is re-examined it is realized that an expansion is being performed in powers of λ_1 , a quantity greater than 1. Convergence cannot be proven for a series expansion of this form. It is therefore impossible to decide a priori which terms should be included and which terms elimi-

nated. This question must be resolved experimentally. (see [5], p. 118).

d. Biderman Theory

Biderman [19] has suggested a W of the form,

$$W = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3 + C_{01}(I_2 - 3) \quad (3.18)$$

where higher order terms in I_1 are retained while terms of higher than first order in I_2 are neglected. This leads to constitutive equations of the form,

$$s_i = \frac{2}{3} \left\{ \left(2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2 \right) \left[C_{10} + 2C_{20}(I_1 - 3) + 3C_{30}(I_1 - 3)^2 \right] + \left(\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2 \right) C_{01} \right\} \quad (3.19)$$

Using experiments performed on an 8% sulfur rubber, the same material used by Treloar [14], Biderman [19] evaluated C_{10}, C_{20}, C_{30} and C_{01} obtaining the values; $C_{10} = 1.9 \text{ Kg/cm}^2$, $C_{20} = -0.019 \text{ Kg/cm}^2$, $C_{30} = 4.6 \times 10^{-4} \text{ Kg/cm}^2$ and $C_{01} = 0.1 \text{ Kg/cm}^2$. Very good correlation is exhibited between experiment and theory for uniaxial tension, uniaxial compression and plane strain of a thin sheet (pure shear). However, if equ. (3.19) is evaluated for the equi-biaxial stressing of a thin sheet, a comparison with the experiments of Treloar shows poor correlation (Fig. 4), indicating that this form for the strain energy density function does not seem to be valid for all types of deformations.

e. Thomas' Theory

In an attempt to obtain a more accurate form for the strain energy density function based on a statistical theory, Thomas [20]

proposed for the form of W ,

$$W = \frac{G}{\lambda_1 \lambda_2 \sqrt{C}} F(k, \alpha) \quad (3.20)$$

where

$$C = 1 - \frac{\lambda_2^2}{\lambda_1^2}$$

F is the elliptic integral of the first kind

$$k = \frac{D}{C} ; \quad \sin \alpha = \sqrt{C}$$

and

$$D = 1 - \frac{\lambda_2^2}{\lambda_1^2}$$

This form for the strain energy density function does give better correlation with experiments than the Statistical theory derived by Treloar. It is quite elegant in that it is derived from a consideration of the microscopic structure. However, its complicated form makes it unsuitable for analytical analysis. Consequently, it has seldom been used.

f. Rivlin-Saunders Theory

Recognizing the shortcomings of the various proposed forms of the strain energy density function, Rivlin and Saunders [16] performed a very complete series of experiments using two different types of vulcanized rubbers. They found, assuming incompressibility, that $\partial W / \partial I_1$ is substantially a constant (a function of neither I_1 nor I_2) and that $\partial W / \partial I_2$ is independent of I_1 but varies with I_2 . Therefore, a strain energy density function was proposed in the form,

$$W = C_1 (I_1 - 3) + f(I_2 - 3) \quad (3.21)$$

where the function of $I_2 - 3$ is to be determined by experimentation.

With this form of W , the constitutive relations (2.26) become,

$$s_i = \frac{2}{3} \left\{ (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) C_1 + \left(\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2 \right) \frac{\partial f}{\partial I_2} \right\} \quad (3.22)$$

Equations (3.21) and (3.22) are more complicated than their counterparts for the Mooney and Statistical theories. However, it seems that if a reasonably simple form can be found for the function $f(I_2 - 3)$, this form will be amenable to analytical solutions for a wide range of deformations and stress configurations. To the author's knowledge, this possibility has been exploited only once in the past. Gent and Thomas [21] proposed a form

$$\frac{\partial f(I_2 - 3)}{\partial I_2} = \frac{W_2}{I_2} \quad (W_2 = \text{constant}) \quad (3.23)$$

which was suggested as an empirical approximation to the theory of Thomas [20] described in the previous section.

The finding of a form for $f(I_2 - 3)$ certainly deserves further research since it has been found by various investigators that equ. (3.21) seems to have wide spread application in specifying the elastic response of many different high polymeric materials [22], [23].

4. EXTENSION OF THE RIVLIN-SAUNDERS THEORY

To express the strain energy density function in the form (3.21) it is necessary to evaluate C_1 and $f(I_2 - 3)$. It will be shown in the following, that $f(I_2 - 3)$ can be expressed as a simple function that has the same basic form for many different natural and synthetic rubbers.

One of the basic shortcomings of many of the previously presented theories is that when the stress field changes, the chosen form of W often becomes a poor approximation. Since it is the purpose of this treatise to solve problems related to balloon analysis, it is desirable to have a theory that is accurate for all membrane problems, regardless of the biaxial stress ratio. Consequently, it was decided to evaluate C_1 and $f(I_2-3)$ in the following manner.

For uniaxial stretching of a thin strip, relations (3.15) and (3.16) are applied to the constitutive equations (2.26) yielding

$$\sigma_u = 2\left(\lambda_u^2 - \frac{1}{\lambda_u}\right)\left(C_1 + \frac{1}{\lambda_u} \frac{\partial f}{\partial I_2}\right) \quad (4.1)$$

Equ. (4.1) can be rearranged to solve for $\partial f / \partial I_2$ as

$$\left(\frac{\partial f}{\partial I_2}\right) = \frac{\sigma_u}{2\left(\lambda_u^2 - \frac{1}{\lambda_u}\right)} - C_1 \lambda_u \quad (4.2)$$

and by (1.21) the second invariant can be expressed as

$$I_{2u} = 2\lambda_u + \frac{1}{\lambda_u^2} \quad (4.3)$$

For equi-biaxial tension of a thin sheet in the 1 and 2 directions, relations (3.11) and (3.12) are applied to the constitutive equations (2.26) yielding

$$\sigma_b = 2\left(\lambda_b^2 - \frac{1}{\lambda_b}\right)\left(C_1 + \lambda_b^2 \frac{\partial f}{\partial I_2}\right) \quad (4.4)$$

As with the uniaxial case, a rearrangement of (4.4) results in

$$\left(\frac{\partial f}{\partial I_2}\right)_b = \frac{C_2 b}{2\left(\lambda_b^4 - \frac{1}{\lambda_b^2}\right)} - \frac{C_3}{\lambda_b^2} \quad (4.5)$$

and by equ. (1.21), the second invariant can be expressed as

$$I_{2b} = \lambda_b^4 + \frac{2}{\lambda_b^2} \quad (4.6)$$

If (3.21) is a valid expression for W , then for some value of C_1 , the curve of $(\partial f/\partial I_2)_b$ versus I_{2b} should be identical with $(\partial f/\partial I_2)_u$ versus I_{2u} .

Computer programs were written for evaluating the curves of $(\partial f/\partial I_2)_u$ versus I_{2u} and $(\partial f/\partial I_2)_b$ versus I_{2b} for a large range of values of C_1 . Using the results of a series of experiments performed by Rivlin and Saunders ([16] p. 274) on thin sheets of a natural vulcanized rubber designated as type B, it was found that for $C_1 = 1.18 \text{ Kg/cm}^2$ * the experimental points follow the same basic curve. (Fig. 6). This curve is qualitatively the same as is presented in [16] and [22]. It was found that a transposed hyperbola gave an excellent fit to this curve (Fig. 6), yielding

$$\frac{\partial f}{\partial I_2} = \frac{C_2}{(I_2 - 3) + \gamma} + C_3 \quad (4.7)$$

with $C_2 = 0.979 \text{ Kg/cm}^2$, $\gamma = 1.519$ and $C_3 = 0.055$. Integrating equ. (4.7) and substituting the result into (3.21) leads to a strain energy density of the form,

$$W = C_1 (I_1 - 3) + C_2 \ln \left[(I_2 - 3) + \gamma \right] + C_3 (I_2 - 3) + C_5 \quad (4.8)$$

* Rivlin and Saunders found a different value of C_1 (1.28 Kg/cm^2). However, they mixed the results of two different types of rubber (A and B). As is evidenced by Fig. 7, at $C_1 = 1.28 \text{ Kg/cm}^2$, $(\partial f/\partial I_2)_b$ does not coincide with $(\partial f/\partial I_2)_u$.

The constant C_5 , can be eliminated by prescribing that $W=0$ for zero strain,

$$0 = C_5 \ln(v) + C_5 \quad (4.9)$$

Therefore,

$$C_5 = -C_5 \ln(v) \quad (4.10)$$

and equ. (4.8) becomes

$$W = C_1 (I_1 - 3) + C_2 \ln\left(\frac{(I_2 - 3) + \gamma}{\gamma}\right) + C_3 (I_2 - 3) \quad (4.11)$$

The constitutive relations (3.22) become,

$$s_i = \frac{2}{3} \left\{ \left(2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2 \right) C_1 + \left(\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2 \right) \left(\frac{C_2}{(I_2 - 3) + \gamma} + C_3 \right) \right\} \quad (4.12)$$

Using (4.12) to solve the problems of uniaxial and equi-biaxial tension as above, the theoretical solution can be compared with the experimental results (Figs. 8 and 9). As a means of comparison, the Statistical and Mooney theory results are presented on the same figures. Quite obviously, this form of the strain energy density function yields much more accurate results than any of the preceding theories.

The above procedure was also applied to the experiments of Treloar [14]. The constants were evaluated as: $C_1 = 1.50 \text{ Kg/cm}^2$, $C_2 = 1.23 \text{ Kg/cm}^2$, $\gamma = 1.785$ and $C_3 = 0.055 \text{ Kg/cm}^2$. Again using equ. (4.12) to solve the problems of uniaxial and equi-biaxial tension, the resulting stress-extension curves are presented on Figs. 4 and 5. For equi-biaxial tension there is excellent correlation with experiments in all ranges of deformation. For uniaxial tension the theoretical curve falls below

the experimental points for values of λ_u larger than 5. This is due to severe deformational anisotropy that cannot be represented within the framework of the assumptions of this theory.

The right hand side of equ. (4.12) can be expressed as a dimensionless function of extension multiplied by an initial elastic modulus. Defining as the shear modulus

$$\mu = \frac{E}{3} \quad (4.13)$$

equ. (2.35) can be expressed as

$$2(C_1 + \frac{C_2}{\gamma} + C_3) = \mu \quad (4.14)$$

The constants C_1 , C_2 and C_3 can be non-dimensionalized in the form,

$$\bar{C}_i = \frac{2C_i}{\mu} \quad i=1,2,3 \quad (4.15)$$

Therefore, the constitutive relation (4.12) becomes,

$$s_i = \frac{\mu}{3} \left\{ (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) \bar{C}_1 + (\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2) \left(\frac{\bar{C}_2}{(I_e - 3) + \gamma} + \bar{C}_3 \right) \right\} \quad (4.16)$$

and the strain energy density function can be expressed

$$W = \frac{\mu}{2} \left\{ \bar{C}_1 (I_1 - 3) + \bar{C}_2 \ln \left(\frac{(I_e - 3) + \gamma}{\gamma} \right) + \bar{C}_3 (I_e - 3) \right\} \quad (4.17)$$

5. HART-SMITH THEORY

After the conclusion of the research described in the preceding sections, it was discovered by the author that another theory for the elastic response of rubber-like materials has been proposed by Hart-Smith

[24], [25]. Recognizing the shortcomings of the previously presented theories, Hart-Smith decided to elaborate upon the strain energy of the theory of Gent and Thomas [21] by assuming their form of $f(I_e - 3)$, equ. (3.23), and finding a form of $\partial W / \partial I_1$ different from the constant G_1 . Using the experiments of Treloar [14], it was found that up to a value of $I_1 = 12$, $\partial W / \partial I_1$ is essentially a constant. For $I_1 > 12$, $\partial W / \partial I_1$ increases with increasing I_1 . It was assumed therefore that the partial derivatives of the strain energy density with respect to the strain invariants could be expressed in the form

$$\frac{\partial W}{\partial I_1} = G e^{k_1 (I_1 - 3)^2} ; \frac{\partial W}{\partial I_e} = \frac{G k_2}{I_e} \quad (5.1)$$

These are referred to as the exponential-hyperbolic elasticity parameters. Using equ. (5.1), the strain energy density function becomes,

$$W = G \left\{ \int e^{k_1 (I_1 - 3)^2} dI_1 + k_2 \ln \left(\frac{I_e}{3} \right) \right\} \quad (5.2)$$

which yields the constitutive relations,

$$s_i = \frac{2}{3} G \left\{ (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) e^{k_1 (I_1 - 3)^2} + \right. \\ \left. + (\lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2) \frac{k_2}{I_e} \right\} \quad (5.3)$$

Using Treloar's experiments, the constants of equ. (5.3) were evaluated as: $G = 1.60 \text{ Kg/cm}^2$, $k_1 = 0.00028$ and $k_2 = 1.2$. The problems of uniaxial and equi-biaxial tension were solved and the resulting

stress- extension curves are presented on Figures 4 and 5. For the equi-biaxial tension experiment, the theory seems to be very accurate up to $\lambda_b = 2.6$. From there on it falls below the experimental points. However, for very large strains it seems to follow the trend of the experimental results much better than the results from the extended Rivlin-Saunders theory. For the uniaxial experiments the Hart-Smith theory is obviously superior to any of the other theories considered. Even though it predicts values a slight bit above the experimental values for most of the range of deformations, in the upper range it obviously yields much better correlation with experiments than all previous theories.

The reasons for the superiority, as well as the deficiencies, of the Hart-Smith theory can be readily explained through a discussion of Figs. 10 and 11. In these figures, $\partial W/\partial I_1$, $\partial W/\partial I_2$ and their characteristic combinations are plotted versus extension for the uniaxial and equi-biaxial loading situations along with the experimental values obtained by Treloar. An examination of Fig. 10 indicates that due to the more complicated form of $\partial W/\partial I_2$ from the extended Rivlin-Saunders theory, it yields a better correlation with experiments than the Hart-Smith theory up to moderate values of deformation. However, in the range of large deformations ($\lambda_b > 3.5$), the Hart-Smith theory due to the more complicated form of $\partial W/\partial I_1$ shows better correlation with the general trend of the experimental points. An examination of Fig. 11, the uniaxial case, brings one to exactly the same conclusions as those reached for the biaxial case.

It would seem quite logical at this juncture to propose a strain energy density that would yield the form of $\partial W/\partial I_1$ of the Hart-Smith theory and the form of $\partial W/\partial I_2$ of the extended Rivlin-Saunders theory. Such a strain energy density would probably yield a more accurate representation of the material considered than any of the above theories. However, its complicated form and the necessity of evaluating five material constants would almost definitely negate the small gain in accuracy over the Hart-Smith theory for the 8% sulfur rubber. It will be shown in the next section, however, that for neoprene balloon film a combined theory is necessary.

6. EVALUATION OF THE CONSTANTS OF THE EXTENDED RIVLIN-SAUNDERS CONSTITUTIVE RELATION FOR NEOPRENE BALLOON FILM AND THE DEVELOPMENT OF A COMBINED EXTENDED RIVLIN-SAUNDERS HART-SMITH CONSTITUTIVE RELATION

In the preceding sections the theory of large elastic deformations was developed and forms for the strain-energy density function were found that seem to represent very well the response characteristics of natural rubbers. It remains now to discover whether or not these laws will apply with similar accuracy to a commonly used meteorological balloon material. For this purpose, experiments were conducted with spherical neoprene day balloons. These balloons, made of unplasticized neoprene had nominal undeformed diameters of nineteen inches and nominal skin thickness of 5 mils.

a. Uniaxial Tension Experiments

In order to determine the mechanical properties of the neoprene film in uniaxial tension, strips six inches long by one-half inch wide

were prepared. Their thicknesses were measured with a micrometer calipers. Bench marks, two inches apart, were drawn on the central region of each sample. This test area was sufficiently small to eliminate end condition effects without loss of experimental accuracy. The samples were hung from a rigid upper support with dead load weights hung from the lower clamp. The weight was varied from zero to one hundred and twenty grams with measurements of length, ℓ , width, b , and thickness, h , made at each weight. Also, each sample was loaded twice to determine if results were reproducible and if the material is elastic. The material was found to exhibit an almost perfectly elastic response.

From these measurements it was possible to determine the three principle extensions, which can be expressed as

$$\lambda_u = \lambda_1 = \frac{\ell}{\ell_0}, \lambda_2 = \frac{h}{h_0}, \lambda_3 = \frac{b}{b_0} \quad (6.1)$$

where the $()_0$ quantities correspond to the undeformed state. The assumption of incompressibility can be stated as

$$I_3 = \lambda_1 \lambda_2 \lambda_3 = 1 \quad (6.2)$$

The validity of equ. (6.2) for this material is verified by the experimental results from one of the strips (Fig. 12).

The true stress, σ_u , is defined, through force equilibrium in the 1 direction, as

$$\sigma_u = \frac{F}{A} \quad (6.3)$$

where F is the uniaxial force and

$$A = bh \quad (6.4)$$

the deformed cross-sectional area of the strip.

Eliminating A through the use of (6.2) and (6.1), (6.3) becomes

$$\sigma_u = \frac{F}{b_o h_o} \lambda_u \quad (6.5)$$

Using (6.1) and (6.5), the experimental results can be represented as points on a curve of σ_u versus λ_u (Fig. 13).

b. Equi-Biaxial Tension Experiments

The equi-biaxial tension response characteristics were determined by inflating the spherical balloons. Crosses were drawn on four different sectors of the balloon. At each inflation pressure the lengths of the cross branches were recorded with a vernier calipers. The circumferential extension can be defined

$$\lambda_b = \frac{s}{s_o} \quad (6.6)$$

where s is the arc length of the cross branch on the deformed balloon.

The circumferential stress, σ_b , can be found by consideration of the force equilibrium equation in the radial direction,

$$\sigma_b = \frac{pR}{2h} \quad (6.7)$$

where R is the deformed radius of the sphere, h is the deformed thickness and p is the pressure difference across the skin. Practically, it is extremely difficult to measure the skin thickness during an experiment. The need for this measurement is eliminated by the incompressibility condition (2.20) which can be expressed as

$$\left(\frac{s}{s_o}\right)^2 = \frac{h_o}{h} \quad (6.8)$$

Equ. (6.7) can then be rewritten,

$$\sigma_b = \frac{pR_o}{2h_o} \frac{R}{R_o} \left(\frac{s}{S_o}\right)^2 \quad (6.9)$$

If the balloon exhibits homogeneous straining,

$$\frac{R}{R_o} = \frac{s}{s_o} \quad (6.10)$$

However, this was not the case. It was necessary to measure the radius separately.

Experiments were performed on two different spheres, each with four equally spaced crosses, yielding eight sets of experimental results. These were averaged and are presented as points of a curve of σ_b versus λ_b (Fig. 14).

c. Application of the Extended Rivlin-Saunders Theory

As was done for natural vulcanized rubber in section 4, $(\partial f/\partial I_2)_b$ versus I_{2b} and $(\partial f/\partial I_2)_u$ versus I_{2u} were evaluated for a large range of values of C_1 . It was found that for $C_1 = 17.00$ psi the experimental points follow the same basic curve (Fig. 15). Fitting equ. (4.7) to this curve yielded the values of the constants as: $C_2 = 19.85$ psi, $\gamma = .735$ and $C_3 = 1.0$ psi.

As with the natural rubbers, equ. (4.12) was used to analyze the situations of uniaxial and equi-biaxial tension. These theoretical solutions are presented in Figs. 13 and 14. The correlation between theory and experiments is quite good for $\lambda_b < 3.5$ and $\lambda_u < 6$ indicating that this theory yields an accurate representation of this particular balloon material in that range of extension. For very large extensions, it is obvious that this theory is not valid. However, an examination of Fig. 15 indicates that Hart-Smith's theory will yield very poor results in the region of moderate strain. (note: the points plotted

in Fig. 15 are valid up to $I_2 - 3 \approx 50$ for the biaxial experiments and $I_2 - 3 \approx 5$ for the uniaxial experiments for correlation with Hart-Smith's theory, since $\partial W / \partial I_1$ remains approximately constant in these ranges of deformation).

d. Combined Extended Rivlin-Saunders, Hart-Smith Theory

It appears that a strain energy density function yielding the $\partial W / \partial I_1$ of the Hart-Smith theory and the $\partial W / \partial I_2$ of the Extended Rivlin-Saunders theory will be most accurate in specifying the response of neoprene balloon film. Such a strain energy density function is:

$$W = C_1 \int e^{k(I_1 - 3)^2} dI_1 + C_2 \ln\left(\frac{(I_2 - 3) + \gamma}{\gamma}\right) + C_3 (I_2 - 3) \quad (6.11)$$

or in dimensionless form,

$$W = \frac{\mu}{2} \left\{ \bar{C}_1 \int e^{k(I_1 - 3)^2} dI_1 + \bar{C}_2 \ln\left(\frac{(I_2 - 3) + \gamma}{\gamma}\right) + \bar{C}_3 (I_2 - 3) \right\} \quad (6.12)$$

The constitutive relation becomes,

$$s_i = \frac{\mu}{3} \left\{ (2\lambda_i^2 - \lambda_{i-1}^2 - \lambda_{i+1}^2) \bar{C}_1 e^{k(I_1 - 3)^2} + \lambda_i^2 \lambda_{i-1}^2 + \lambda_i^2 \lambda_{i+1}^2 - 2\lambda_{i-1}^2 \lambda_{i+1}^2 \left(\frac{\bar{C}_2}{(I_2 - 3) + \gamma} + \bar{C}_3 \right) \right\} \quad (6.13)$$

Equations (6.11-6.13) reduce to those of the Extended Rivlin-Saunders theory for $k = 0$ and to those of the Hart-Smith theory for $\gamma = 3$ and $C_3 = 0$. Consequently, the combined theory is a more general theory than any previous theory and the results of all previous theories can

be obtained from it through a proper choice of constants.

Using the values of C_1 , C_2 , C_3 and γ obtained in the previous section along with a value of $k = 0.00015$, this theory was applied to the experiments presented in Figs. 13 and 14. There is excellent correlation with experiments for all ranges of deformation.

IV. CONCLUSIONS

To analyze any engineering structure it is necessary to have three sets of equations; the equilibrium equations, the kinematical equations and the constitutive equations. The first two sets are easily obtained and were previously available. However, the constitutive equations for neoprene were not previously developed for the deformation range needed for the analysis of balloons.

In this report, the suggested constitutive relation, equ. (6.13), has been shown to yield an excellent representation of the stress-strain characteristics of neoprene balloon film for a large range of deformations. Using (6.13) coupled with the appropriate equilibrium and kinematical equations, various balloon structures can now be analyzed. These analyses will be presented in forthcoming reports.

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APPENDIX

DEFINITION OF SYMBOLS

The symbols used in this report are defined below

<u>Symbol</u>	<u>Definition</u>	<u>Dimension</u>
x, y, z	Coordinates of a point of the undeformed body	length
ξ, η, ζ	Coordinates of a point of the deformed body	length
u, v, w	Displacement components	length
ds	Length of an undeformed line element	length
ds'	Length of a deformed line element	length
l, m, n	Direction cosines of undeformed line element	
l', m', n'	Direction cosines of deformed line element	
ϵ^L	Lagrangian strain measure	
ϵ^E	Almansi (or Eulerian) strain measure	
ϵ^s	Swainger strain measure	
ϵ^k	Kuhn strain measure	
ϵ^{sim}	Simple strain measure (stretch)	
ϵ^{LH}	Ludwik-Hencky (or logarithmic) strain measure	
λ_t	Total extension	
λ_{ij}	Components of total extension	
I_1, I_2, I_3	Invariants of λ_t	
I_1^L, I_2^L, I_3^L	Invariants of ϵ^L	
W	Strain energy density function	force per unit area

<u>Symbol</u>	<u>Definition</u>	<u>Dimension</u>
σ_{ij}^*	Generalized stresses	force per unit area
σ_{ij}	True stresses	force per unit area
δ_{ij}	Kroneker delta	
s_i	Principal stress deviator	force per unit area
e_i	Principal lagrangian strain deviator	
E	Elastic modulus	force per unit area
$()_u$	Uniaxial quantities	
$()_b$	Equi-biaxial quantities	
μ	Shear modulus	force per unit area
l	Length of uniaxial strip	length
h	Deformed thickness of uniaxial strip and spherical balloon	length
$()_o$	Undeformed quantities	
b	Width of uniaxial strip	length
F	Force applied to uniaxial strip	force
A	Deformed cross-sectional area of uniaxial strip	area
s	Arc length or deformed spherical balloon	length
R	Deformed radius of spherical balloon	length
P	Pressure difference across the balloon skin	force per unit area
C_1, C_2, C_3	Material constants of combined constitutive relation	force per unit area
γ, k	Material constants of combined constitutive relation	

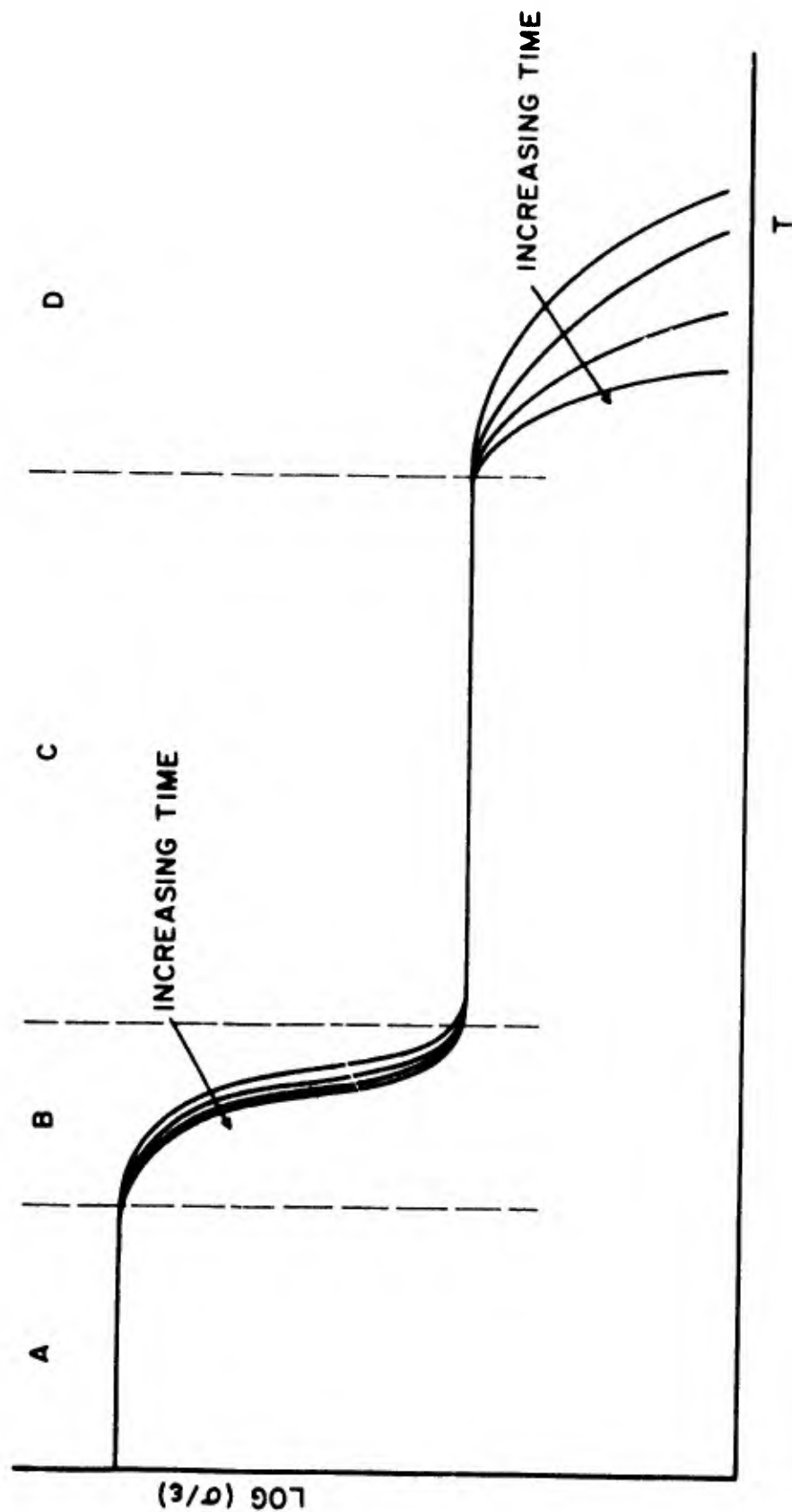


FIG. 1

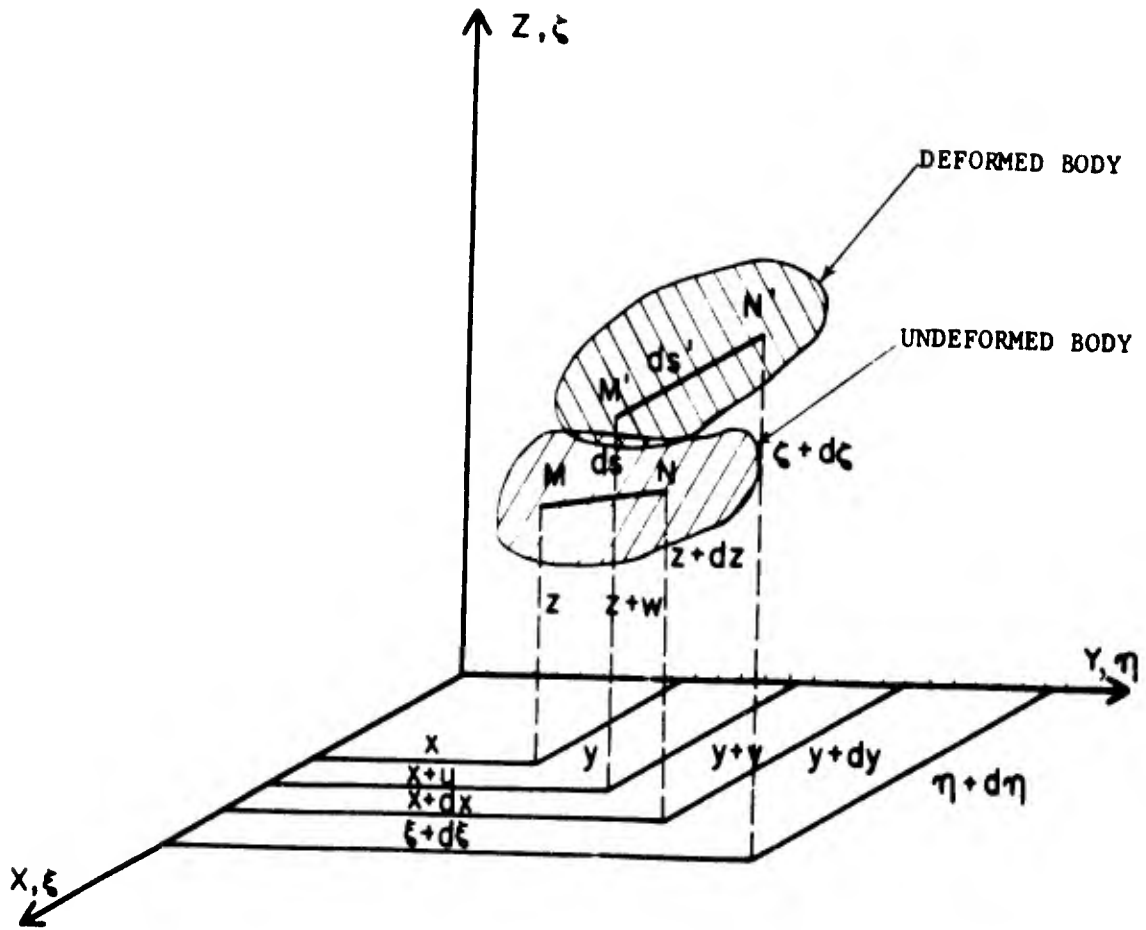


FIG. 2

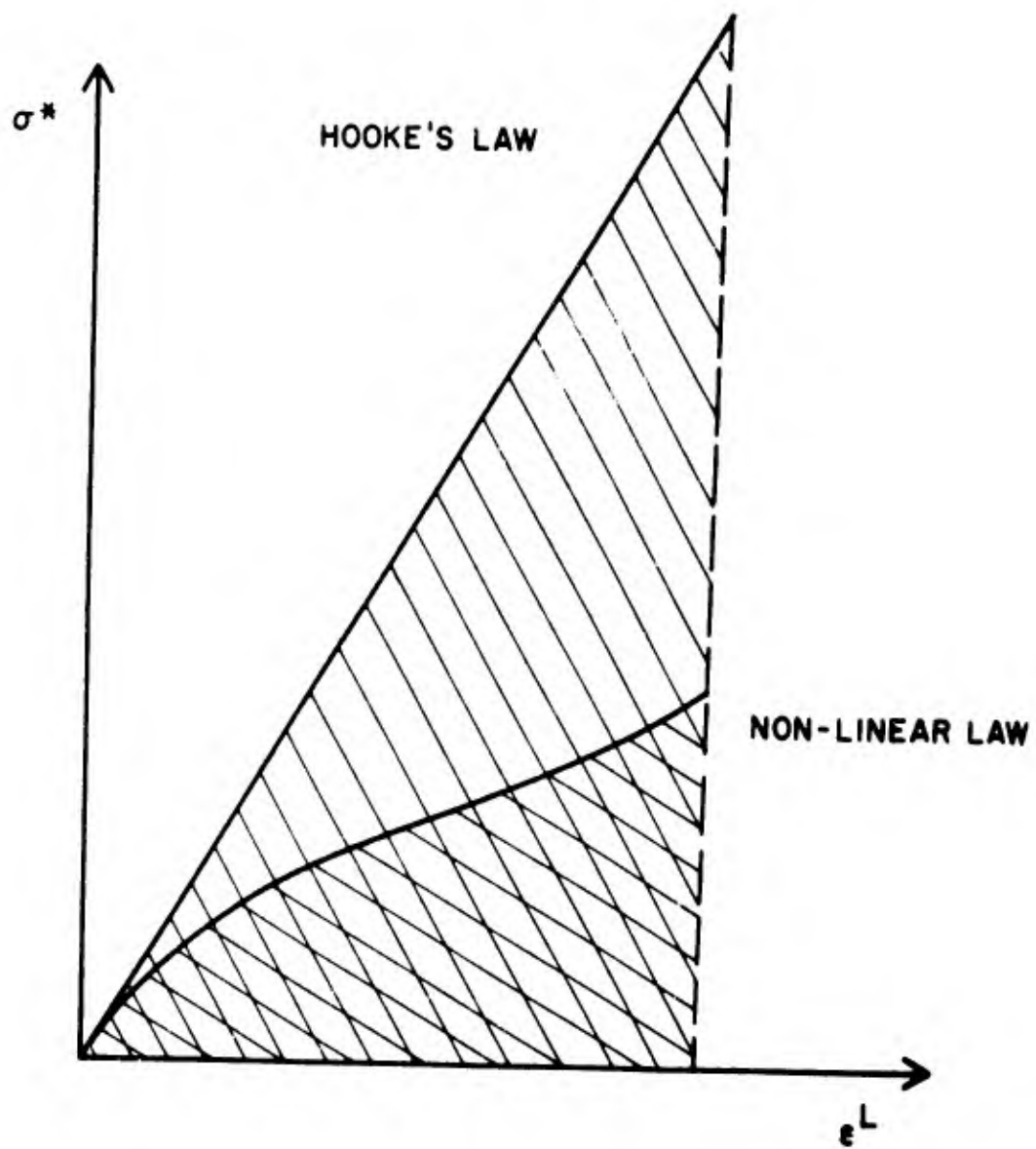


FIG. 3

EQUI-BIAxIAL TENSION OF A THIN SHEET
8% SULPHUR RUBBER

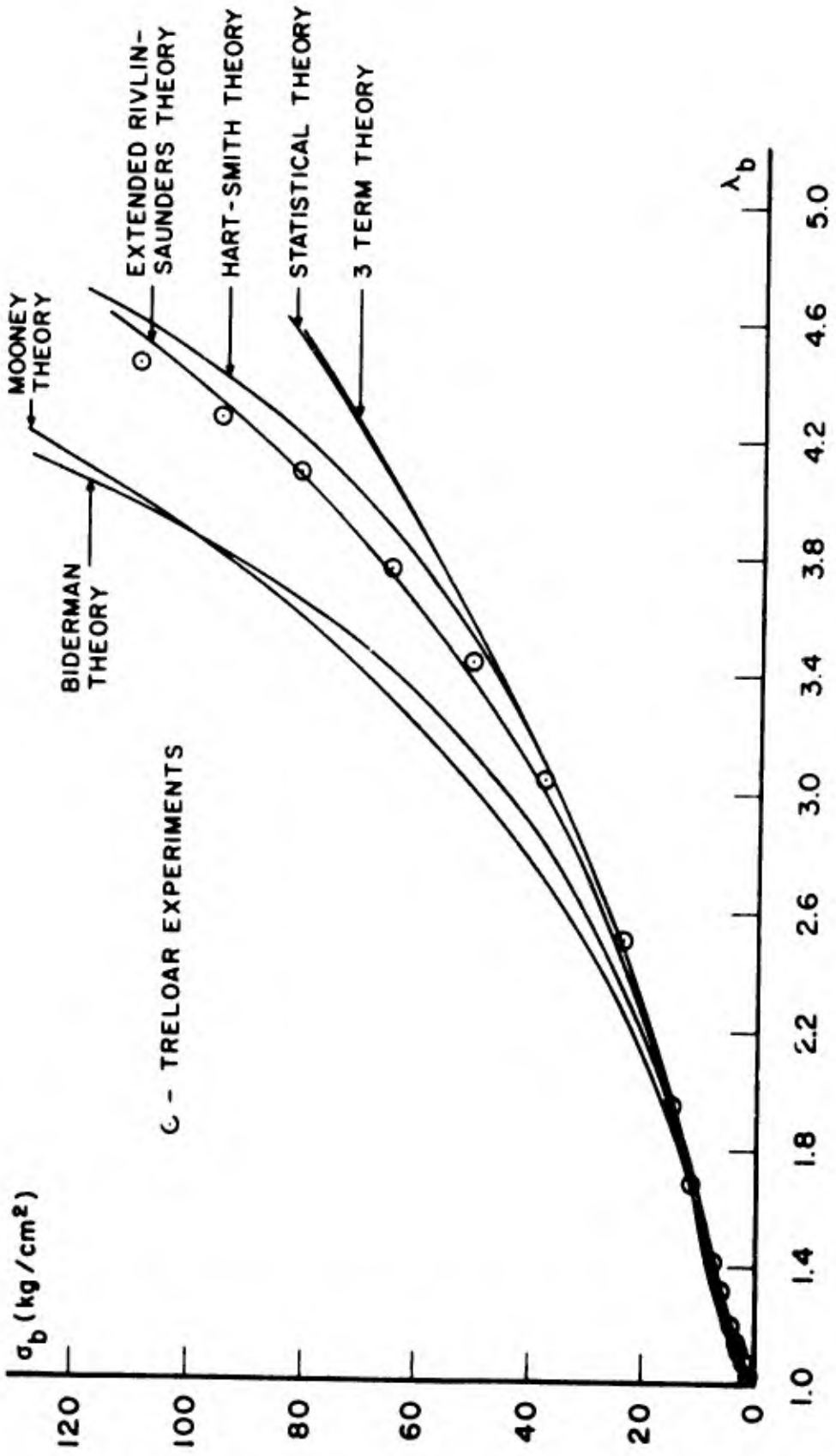


FIG. 4

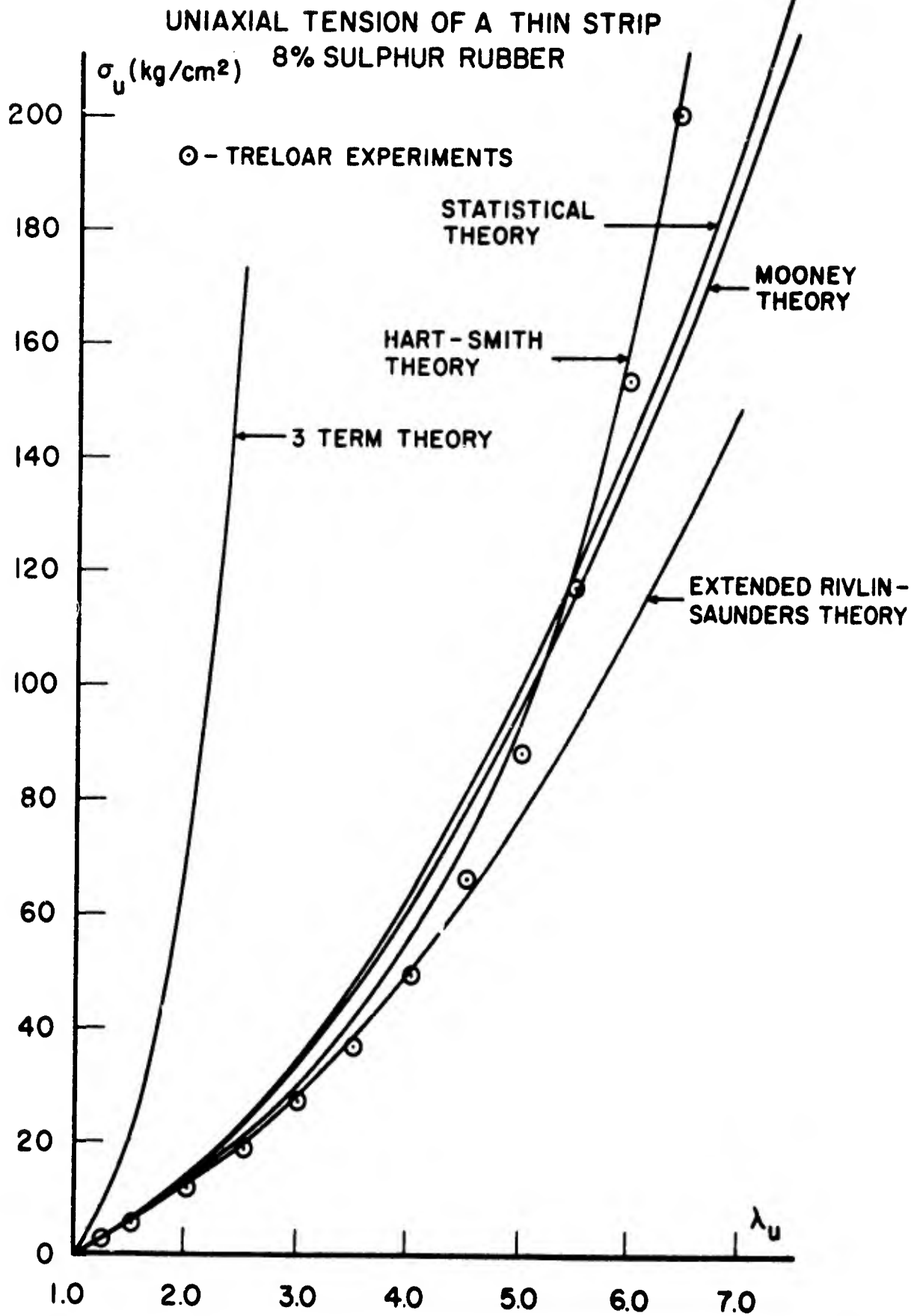


FIG.5

RIVLIN TYPE B RUBBER

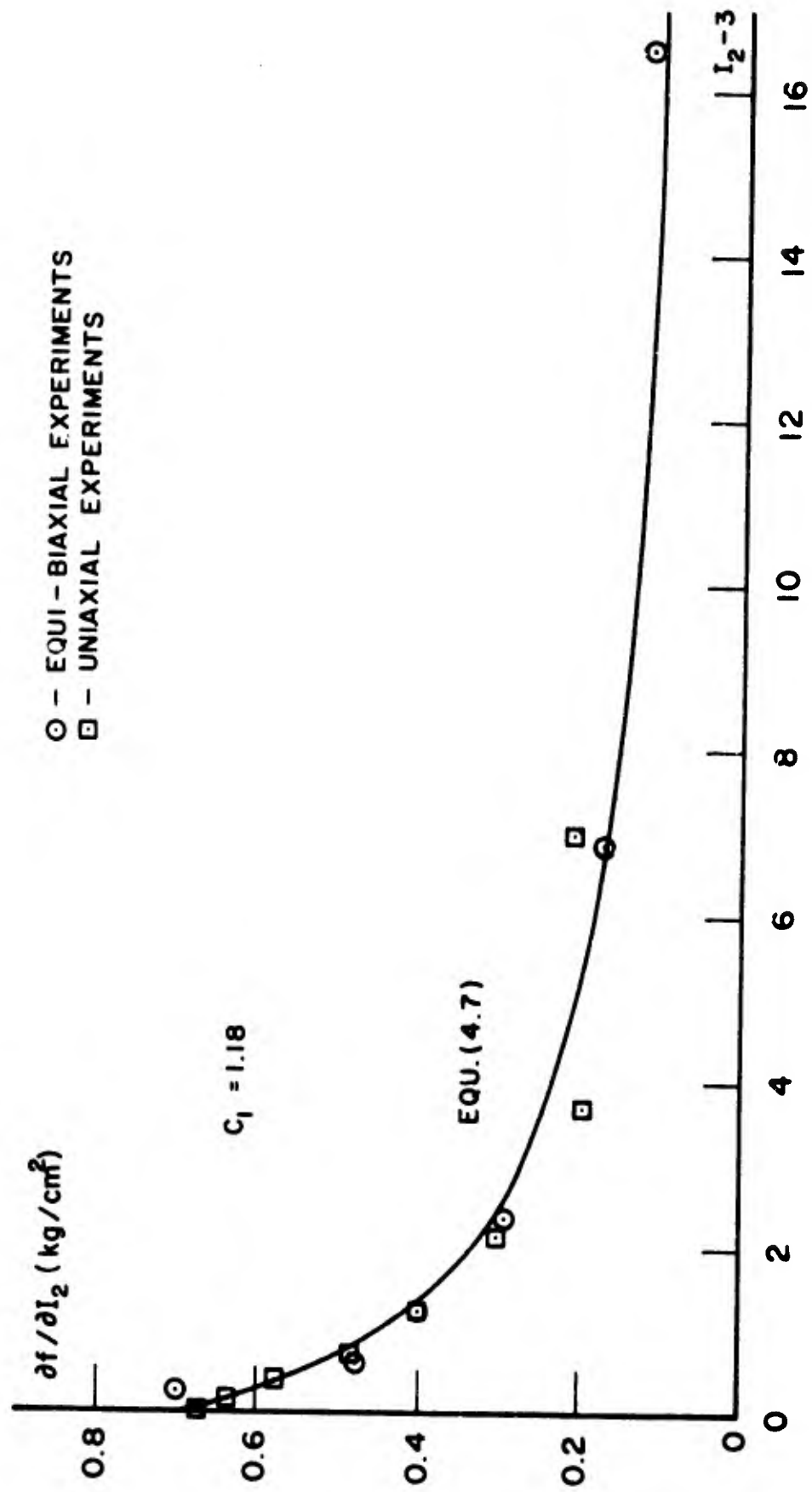


FIG. 6

RIVLIN TYPE B RUBBER

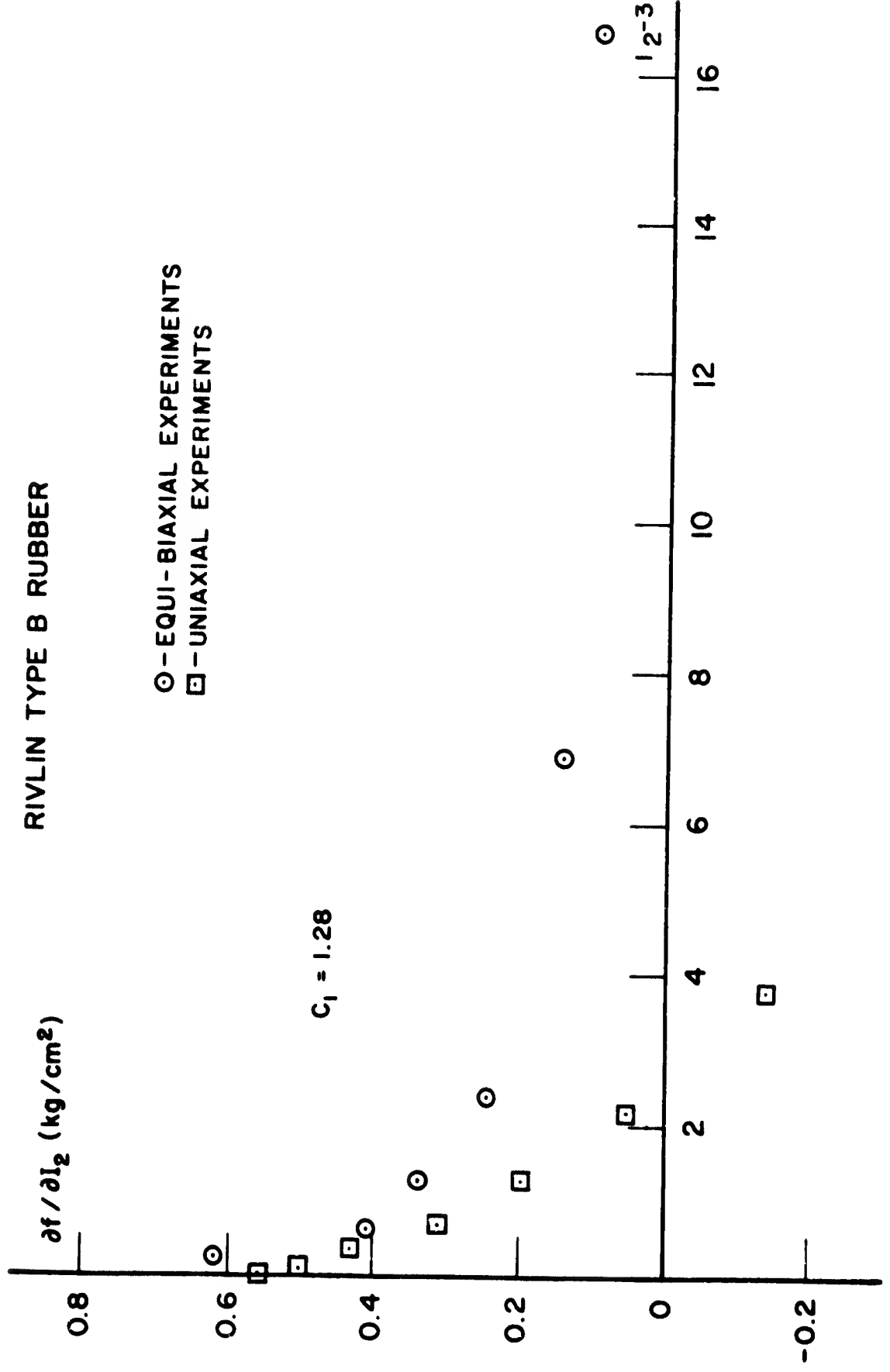


FIG.7

EQUI-BIAxIAL TENSION OF A THIN SHEET
RIVLIN TYPE B RUBBER

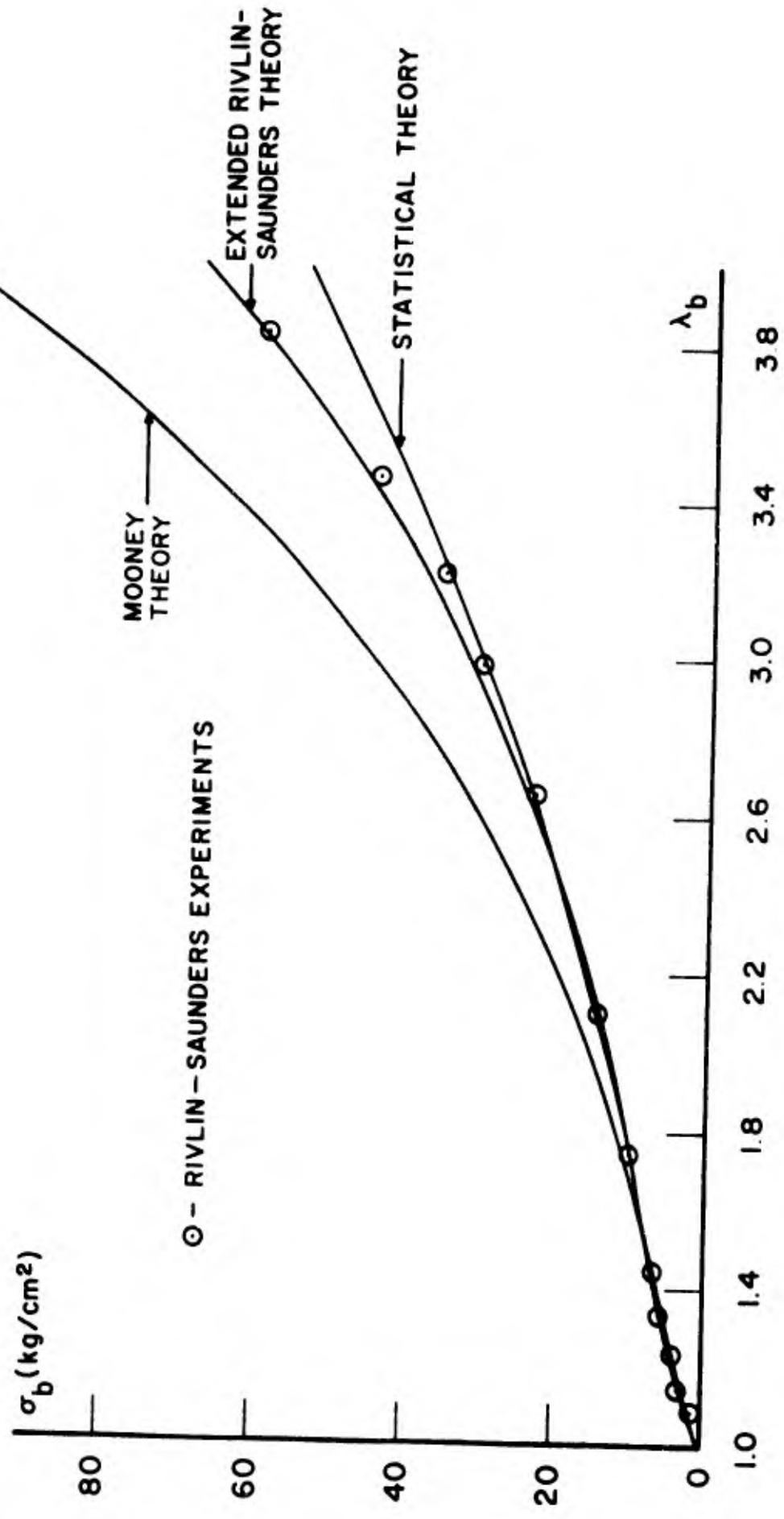


FIG. 8

UNIAXIAL TENSION OF A THIN STRIP
RIVLIN TYPE B RUBBER

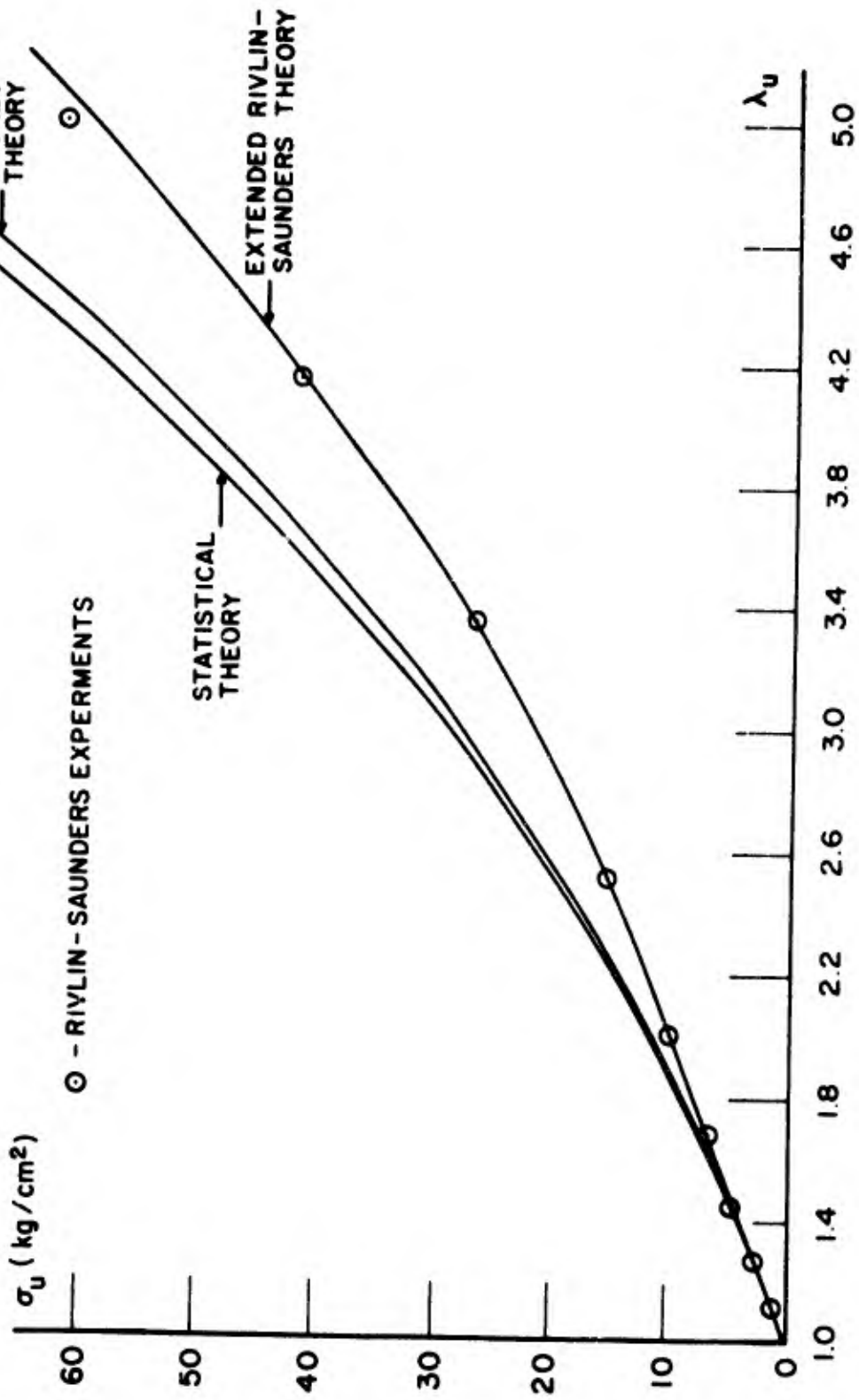


FIG. 9

EQUI-BIAXIAL TENSION OF A THIN SHEET
8% SULPHUR RUBBER

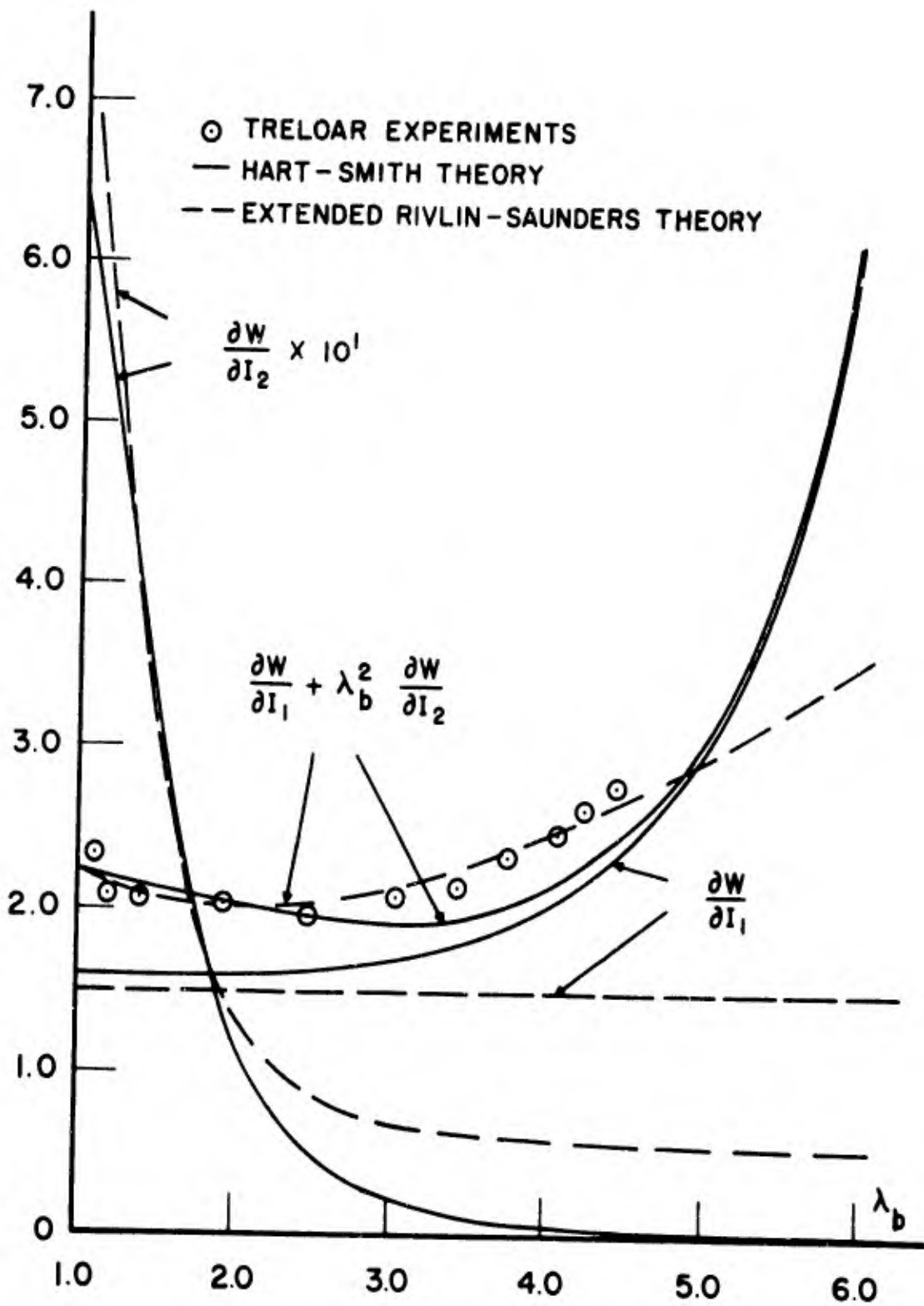


FIG. 10

UNIAXIAL TENSION OF A THIN STRIP
8% SULPHUR RUBBER

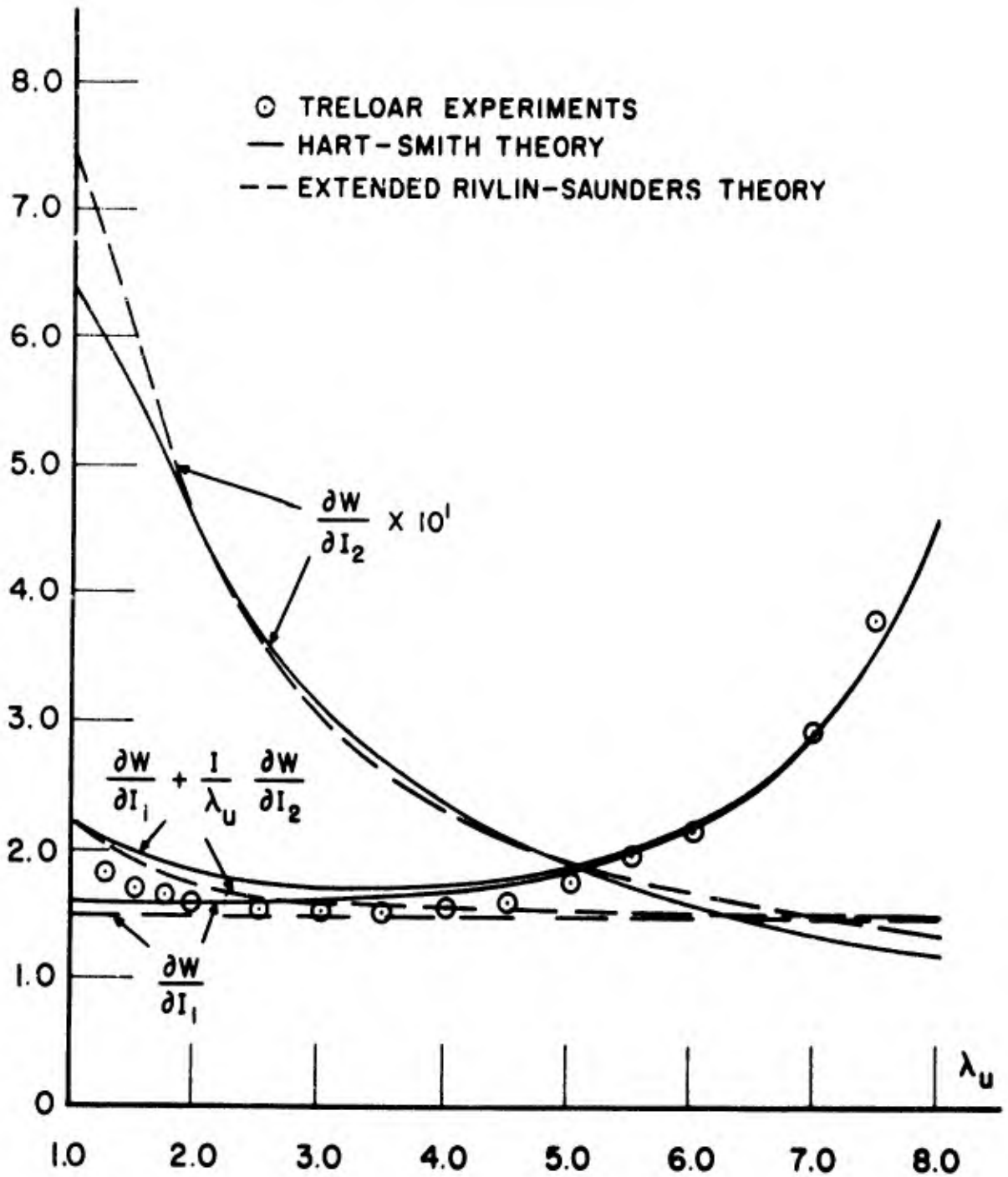


FIG. 11

NEOPRENE BALLOON FILM

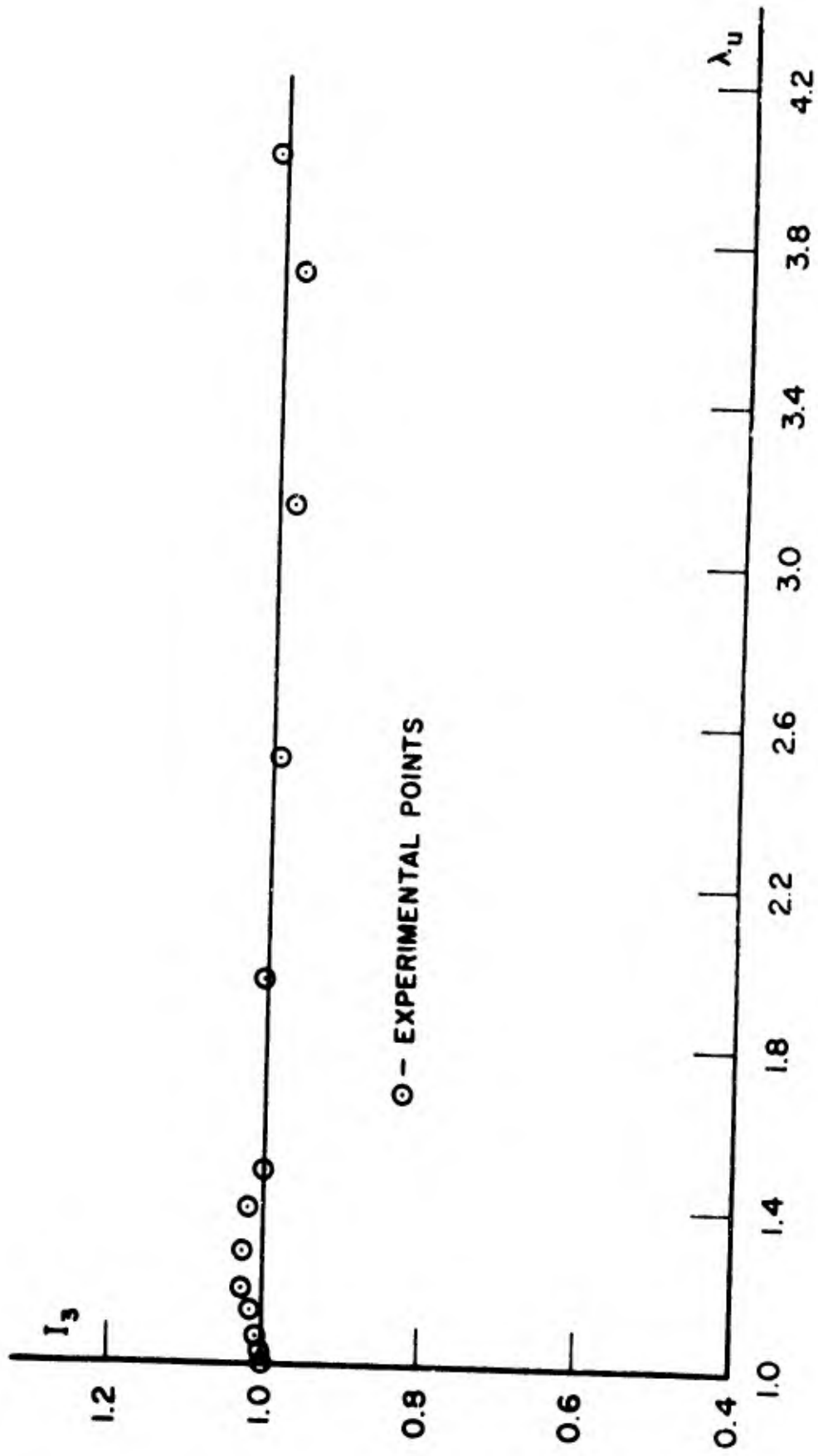


FIG.12

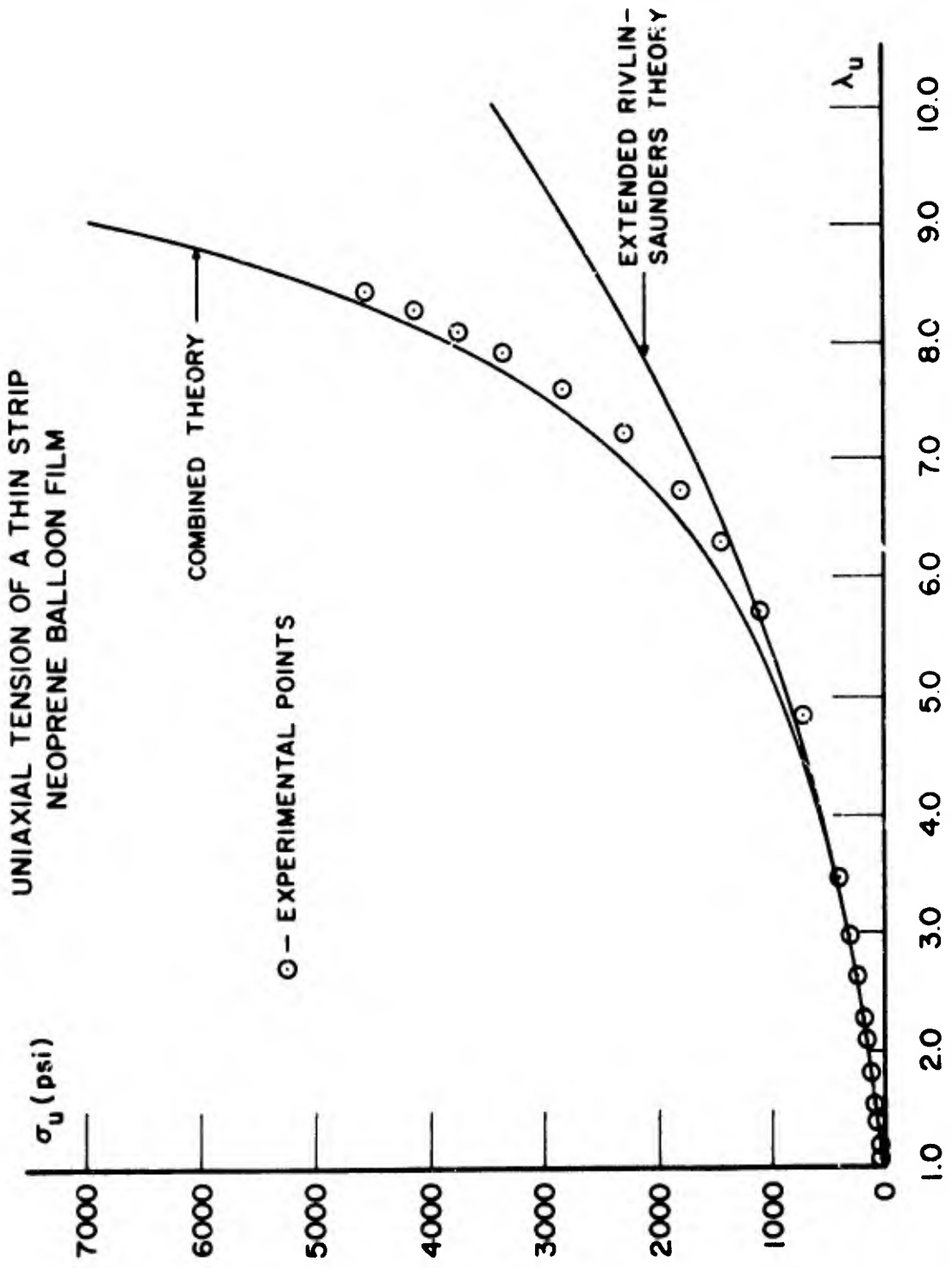


FIG. 13

EQUI-BIAxIAL TENSION OF A THIN SHEET
NEOPRENE BALLOON FILM

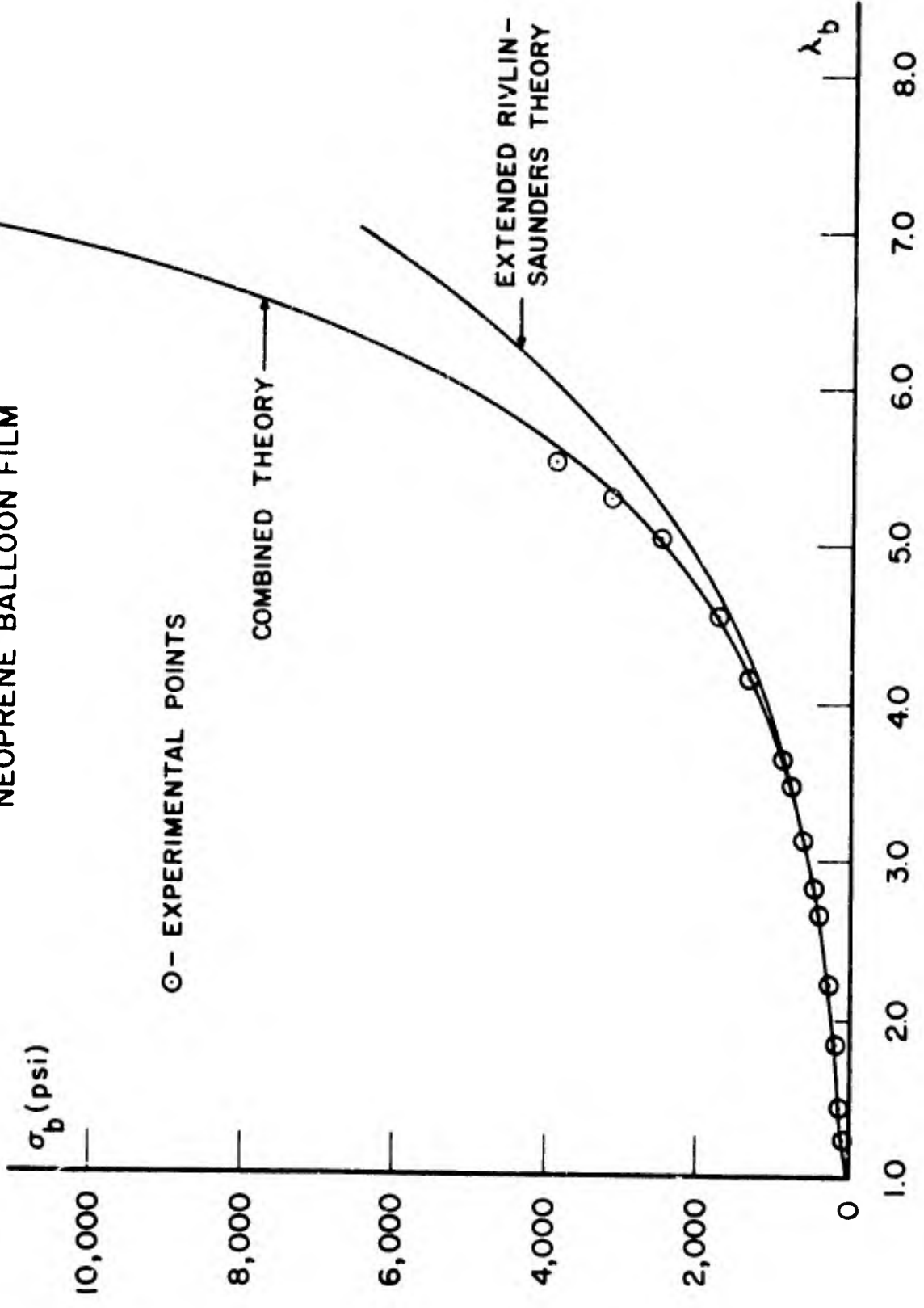


FIG. 14

NEOPRENE BALLOON FILM

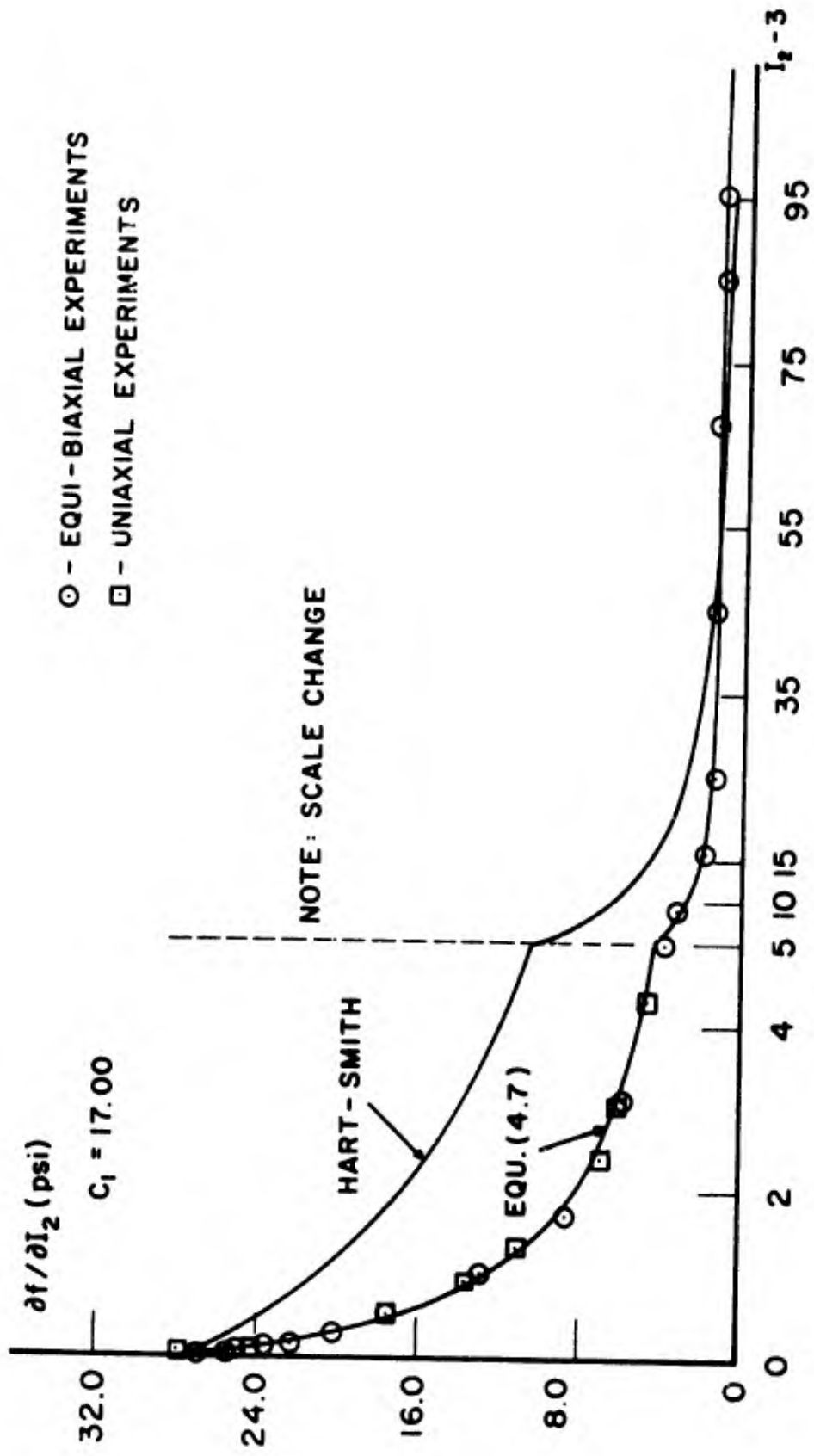


FIG. 15

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13. ABSTRACT

The physical and mechanical characteristics of neoprene film, a commonly used expandable-type balloon skin material, are presented for the purpose of developing constitutive equations that can be used in balloon analyses.

"Day" balloon film is found to be an almost perfectly elastic material through an extremely large range of deformations. A survey of previously proposed elastic constitutive equations for rubber-like materials is presented and all are found to be inadequate for neoprene. Consequently, a new constitutive relation based on the attempts of previous investigators and experiments performed by the author is proposed. It is shown to be a generalization of most earlier theories.

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14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
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