# CONSTITUTIVE EQUATIONS FOR SPECIAL LINEAR VISCOELASTIC COMPOSITES WITH GROWING DAMAGE

R.A. SCHAPERY

Department of Aerospace Engineering and Engineering Mechanics, The University of Texas, Austin, Texas 78712

#### ABSTRACT

Constitutive equations for a class of linear viscoelastic composites with damage growth are reviewed and then applied to modeling the behavior of a particle-reinforced rubber with microcracking and a fiber-reinforced plastic with transverse cracking. Conditions which lead to relatively simple constitutive equations for composites are first reviewed. These equations and the evolution equations for damage growth are like those for elastic materials, except history-modified strains, called pseudo strains, replace physical strains. Use of these equations is then illustrated for a highly-filled rubber and a fiber-composite laminate, both with significant amounts of damage.

#### KEYWORDS

Viscoelasticity, fiber composites, particulate composites, damage growth.

## INTRODUCTION

For many important viscoelastic polymeric composites, the reinforcement is very stiff relative to that of the polymer matrix. This condition, together with the common case in which the matrix can be characterized by a single creep or relaxation function (e.g., the matrix is linearly viscoelastic with an essentially constant Poisson's ratio) lead to a relatively simple mathematical model of the composite, with or without distributed microcracking. Basically, through a modification of the strains, the homogenized constitutive equations are like those for an elastic composite. The evolution equations for damage growth simplify in the same way. Theory and application of these ideas for linear and nonlinear media were published many years ago (Schapery, 1981, 1982, 1984). Here we shall briefly review the basis for these simplified models in the linear viscoelastic range and then describe two recent applications. The first one is concerned with the behavior of a highly-filled particle-reinforced rubber under multiaxial loading, while the second deals with transverse cracking and its effect on the homogenized constitutive equation for a fiber composite.

# CONSTITUTIVE EQUATIONS WITHOUT DAMAGE

We consider first the case in which there is no damage and the monolithic or composite material is linearly viscoelastic and anisotropic. Then, with  $\epsilon$  and  $\sigma$  representing the strain and stress tensors, respectively, the constitutive equation is

# $\epsilon = \{ Sd\sigma \} + \epsilon_T \tag{1}$

where S is the fourth order creep compliance tensor and  $\epsilon_T$  is the strain tensor due to temperature and moisture (and other absorbed substances which affect the strains). The braces are abbreviated notation for a linear hereditary integral. Although the most general form could be used, allowing for general aging effects, for notational symplicity we shall use the familiar form for thermoheologically simple materials,

$$\{fdg\} = \int_{-\infty}^{t} f(\xi - \xi') \frac{\partial g}{\partial t'} dt' = \int_{-\infty}^{\xi} f(\xi - \xi') \frac{\partial g}{\partial \xi'} d\xi'$$
 (2)

where

$$\xi \equiv \int_0^t dt'' / a_T [T(t'')], \quad \xi' = \xi(t')$$
 (3)

and  $a_T$  is the temperature shift factor. If the temperature is constant in time then  $\xi - \xi' = (t - t')/a_T$ . Physical aging (Struik, 1978) may be taken into account by introducing explicit time-dependence in  $a_T$ ; i.e. use  $a_T = a_T(T,t'')$  in Eq.(3). The effect of plasticizers, such as moisture, may also be included in  $a_T$ . When Eq.(2) is used with Eq.(1), f and g are components of the creep compliance and stress tensors, respectively.

In certain important cases, the creep compliance components are proportional to one function of time,

$$S = kD \tag{4}$$

where k is a constant tensor and  $D=D\left(\xi\right)$  is a creep compliance. Isotropic materials with a constant Poisson's ratio satisfy Eq.(4). If such a material has mechanically rigid reinforcements and/or holes (of any shape), it is easily shown by dimensional analysis that its homogenized constitutive equation satisfies Eq.(4); in this case the stress and strain tensors in Eq.(1) should be interpreted as volume-averaged quantities (Hashin, 1983). The Poisson's ratio for polymers at temperatures which are not close to their glass-transition temperature,  $T_g$ , is nearly constant; above  $T_g$  Poisson's ratio is essentially one-half, while below  $T_g$  it is commonly in the range 0.35-0.40 (Schapery, 1974).

Equations (1) and (4) give

$$\epsilon = \mathbf{k} \left\{ Dd\sigma \right\} + \epsilon_T \tag{5}$$

The inverse is

$$\sigma = k_I \{ E d \epsilon \} - k_I \{ E d \epsilon_T \} \tag{6}$$

where  $k_I = k^{-1}$  and  $E = E(\xi)$  is the relaxation modulus.

In relating solutions of elastic and viscoelastic boundary value problems, it is helpful to introduce the quantities

$$\epsilon^R \equiv \frac{1}{E_R} \{ E d \epsilon \}, \quad \epsilon_T^R \equiv \frac{1}{E_R} \{ E d \epsilon_T \}, \quad u^R \equiv \frac{1}{E_R} \{ E d u \}$$
(7)

where  $E_R$  is a arbitrary constant with dimensions of modulus, called the reference modulus; also  $\epsilon^R$  and  $\epsilon^R_I$  are so-called pseudo strains and  $u^R$  is the pseudo displacement. Equation (6) may now be written as

$$\sigma = C\epsilon^R - C\epsilon_T^R \tag{8}$$

where  $C \equiv E_R k_I$  is like an elastic modulus tensor. Equation (8) reduces to that for an elastic material by taking  $E = E_R$ ; it reduces to the constitutive equation for a viscous material by taking E as a Dirac delta function.

# A CORRESPONDENCE PRINCIPLE WITH DAMAGE GROWTH

When Eq.(8) is combined with boundary conditions and the remaining field equations, and  $a_T$  is spacewise constant, a simple method for deriving viscoelastic solutions results, which may be stated using a so-called correspondence principle.

First, we introduce the so-called reference elastic boundary value problem and solution. This reference problem uses Eq.(8) together with pseudo strain-pseudo displacement equations and the equilibrum equations in terms of  $\sigma$ . The boundary conditions are  $T_i = \sigma_{ij} n_j$  (where  $\mathbf{n}$  is the outer unit normal) and  $U_i^R = u_i^R$  on the surfaces  $S_T$  and  $S_U$ , respectively. The solution to this problem is  $\sigma_{ij}$ ,  $\epsilon_{ij}^R$  and  $u_i^R$ , called the reference elastic solution.

Let us now state the correspondence principle, designated as CP-II by Schapery (1981), and adapted here to the present linear formulation: For the viscoelastic body, let surface tractions  $T_i$ , and displacements  $U_i$  be specified functions of time and position on surfaces  $S_T$  and  $S_U$ , respectively. Further, we suppose  $dS_T/dt \geq 0$ , where the total, instantaneous surface is  $S = S_T + S_U$ . Then, the viscoelastic solution is

$$\sigma_{ij}, \quad \epsilon_{ij} = E_R \{ Dd\epsilon_{ij}^R \}, \quad u_i = E_R \{ Ddu_i^R \}$$

$$(9)$$

Thus, one first solves the reference elastic boundary value problem. Then the viscoelastic stresses are equated to the elastic stresses, while the viscoelastic strains and displacements are found by evaluating the hereditary integrals in Eq.(9). It should be noted that the specified boundary tractions in the elasticity and viscoelasticity problems are the same. However, in order that the viscoelastic displacements in Eq.(9) equal  $U_i$  on  $S_U$ , it is necessary to specify  $U_i^R$  such that

$$U_i^R = \frac{1}{E_R} \{ EdU_i \} \tag{10}$$

where E is the relaxation modulus associated with the creep compliance D. It should also be added that the instantaneous geometry of the elastic and viscoelastic bodies is to be identical, including the crack geometry. The condition  $dS_T/dt \geq 0$  allows for crack growth, but not crack face contact and healing (unless the tractions on rejoining crack faces are treated artifically as specified quantities).

If the viscoelastic body is a composite, these cracks may be of any size relative to the size of the constituent phases (assumed to be perfectly rigid or soft). In using this correspondence principle to develop constitutive equations for homogenized media with distributed cracking, the boundary conditions are specified in the usual way; the external surfaces of a representative volume element are subjected to tractions which are spacewise uniform or to displacements which vary linearly with respect to spacial location (Hashin, 1983).

For the special type of composites considered here, Eq.(8) for the viscoelastic matrix and the correspondence principle lead to the same form of a constitutive equation for the composite. For the composite, C is to be interpreted as the effective modulus tensor for the reference elastic problem and, in general, depends on the reinforcement and hole geometry as well as the instaneous geometry of all cracks.

Equations for predicting crack growth (or, more generally, damage growth) in the special type of viscoelastic materials studied here have been developed and applied in previous work (e.g. Schapery, 1981, 1982, 1984). We shall discuss specific growth (or evolution) equations in the two examples that follow.

## APPLICATION TO A PARTICLE-REINFORCED RUBBER

Rubber is frequently filled with relatively high modulus particles. Two importnt structural applications are tires and solid propellant. Here we shall briefly illustrate the composite version of Eq.(8) for an inert solid propellant. Theoretical and experimental details are given elsewhere (Park and Schapery, 1996). The matrix is a lightly crosslinked rubber, and it is filled with 70 volume percent of a bimodal distribution of particles; their modulus is at least three orders of magnitude higher than that of the rubber. Crack-like defects develop and grow in the rubber and at particle-rubber interfaces even under applied stress that is well below the global strength of solid propellant (Cornwell and Schapery, 1975). Initially the material is isotropic. However, the damage (microcracking) produces global anisotropy. For the particular case studied here, in which specimens (3 x .4 x .5 inches) are subjected to specified, time-dependent axial elongation and constant confining pressure, the damage leads to transverse isotropy, with the axis of isotropy obviously being in the specimen's axial direction. The axial stress  $\sigma$  (force/initial area) and dilatation were measured experimentally over a wide range of constant axial strain rates (400:1), pressures (0 to 800 psi) and temperatures (-40C to 25C). Figure 1 shows the effect of different constant pressures on the stress-strain and dilatation-strain response curves.

The composite constitutive model is assumed to be that of a linear elastic material, but with pseudo strains in place of physical strains, as predicted by the correspondence principle. Thermal expansion strains  $\epsilon_T$  are neglected. For the particular problem at hand it is convenient to write the constitutive equation in a form for which the axial pseudo strain  $\epsilon^R$  and pressure p appear on the right side and the axial tensile stress  $\sigma$  (above that due to the pressure) and pseudo dilatation are on the left side. Then, the stress-pseudo strain equations are

$$\sigma = \hat{C}_{11}\epsilon^R + \hat{C}_{12}p \qquad v^R = \hat{C}_{12}\epsilon^R + \hat{C}_{22}p \tag{11}$$

The symmetry of  $\hat{C}_{ij}$  follows from the fact that  $\sigma$  and  $\epsilon^R$  as well as p and  $v^R$  are work conjugates for an elastic material. As a result of this symmetry, we may construct a so-called pseudo dual strain energy function,

$$W_D = \frac{1}{2}\hat{C}_{11} \left(\epsilon^R\right)^2 + \hat{C}_{12}\epsilon^R p + \frac{1}{2}\hat{C}_{22}p^2 \tag{12}$$

Obviously,

$$\sigma = \partial W_D / \partial \epsilon^R, \quad v^R = \partial W_D / \partial p \tag{13}$$

The material is thermoheologically simple, so that the hereditary integrals for pseudo strains are based on Eqs.(2) and (3).

The coefficients  $\hat{C}_{ij}$  depend on the damage, which we have found requires the use of two damage parameters or internal state variables,  $S_1$  and  $S_2$ , say. Evolution laws of the following form are used

$$\dot{S}_1 \equiv dS_1/d\xi = f_1(S_1) \left(-\frac{\partial W_D}{\partial S_1}\right)^{\alpha_1} \text{ when } \dot{S}_1 \neq 0$$
 (14a)

$$\dot{S}_2 \equiv dS_2/d\xi = f_2(S_2) \left(-\frac{\partial W_D}{\partial S_2}\right)^{\alpha_2} \text{ when } \dot{S}_2 \neq 0.$$
 (14b)

where, for the particular material studied,  $\alpha_1 = 6$  and  $\alpha_2 = 4.5$ .

Observe that the same reduced time is used in Eq.(14) as in Eq.(3). Namely,  $d\xi = dt/a_T$ , where  $a_T$  is the same as for the linear viscoelastic functions  $E(\xi)$  and  $D(\xi)$ . That this

is possible appears to imply the intrinsic viscoelastic behavior and microcrack growth are associated with the same physical rate mechanisms, as reported previously (Schapery, 1978). The form of Eq.(14) is motivated by the power-law growth behavior of macrocracks (Schapery,1978). The quantity  $-\partial W_D/\partial S_i$  is a so-called thermodynamic force, which is pseudo energy release rate when  $S_i$  is crack area; the use of pseudo energy is motivated by viscoelastic crack growth theory (Schapery, 1984). By redefinition of  $S_i$ , the positive factors  $f_i$  may be eliminated, as done in the earlier work. We should also add that it was found unnecessary to use threshold values of  $-\partial W_D/\partial S_i$ , below which  $\dot{S}_i$  vanishes. It is believed the very high stress concentrations due to the high filler volume fraction initiate microcracking at very low applied axial load.

The way in which the  $\hat{C}_{ij}$  depend on  $S_i$  is given by Park and Schapery (1996). Here, we observe only that

$$\hat{C}_{11} = C_{11}(S_1) C(S_2), \quad \hat{C}_{12} = C_{12}(S_1), \quad \hat{C}_{22} = C_{22}(S_1)$$
 (15)

Under very high pressures ( $\geq$  800 psi),  $v \simeq 0$  and  $C_{12} \simeq C_{22} \simeq 0$ . For this case  $S_1 \simeq 0$  and the Poisson's ratio is essentialy 0.5. The damage is associated with shearing in this case, as accounted for by  $S_2$ . The stress-strain curve in Fig. 1 at 800 psi reflects changes in only  $C(S_2)$ . The behavior of  $C_{ij}(S_1)$  is similar to that predicted from a micromechanical model for opening-mode microcrack growth in a highly-filled rubber (Schapery, 1991); in this case  $S_1$  is a function of the number and size of the microcracks (which are responsible for the dilatation), such as that observed in Fig. 1.

Equations (11)-(14) are in good agreement with all of the experimental results for constant strain rate loading. Dual strain rate tests were used to further evaluate the model. Figure 2, from Park and Schapery (1996), shows that there is good agreement between theory and experiment. The present model was extended to three-dimensional behavior by Ha and Schapery (1996) using only the material functions and parameters in Eqs.(11)-(14) by assuming the material is locally transversely isotropic, with the current maximum principal stress defining the axis of isotropy. Good agreement between theory and experiment for load-displacement data and strain distributions in rectangular strips with holes and cracks was found. In these tests, specimens were clamped along their long edges and loaded by different constant displacement rates.

#### APPLICATION TO TRANSVERSE CRACKING IN LAMINATES

A common form of damage in laminates of continuous fiber-reinforced plastics is transverse cracking. Figure 3 illustrates these cracks, which are normal to the  $(x_1 - x_2)$  ply-plane and extend in the  $x_1$ -direction parallel to the fibers (normal to the page). These cracks often form suddenly in multi-directional fiber laminates at loads that are well-below ultimate values for the laminate. In standard tensile specimens, after initiation they grow quickly to the full width of a specimen. These cracks are important because they often precipitate other types of damage, such as fiber breakage in adjacent layers and delamination. There are many publications on this subject (e.g. Friedrich, 1989) with most dealing with timeindependent behavior. Time-dependent effects are described by Moore and Dillard (1990) and Schapery and Sicking (1995). Here we shall discuss only the problem of determining the effective transverse modulus  $E_2$  of a homogenized layer with transverse cracks and the evolution equation for these cracks. The average spacing of these cracks diminishes as the loading is increased, and thus 2a in Fig. 3 (representing the average spacing) is a decreasing function of time. When the effective modulus  $E_2$  is determined from theory or experiment, and is expressed as a function of crack density, 1/2a, or normalized crack density,  $\beta \equiv h/a$ , it is found to be somewhat sensitive to the adjacient layers (such as their fiber orientation and the number of plies). However, Schapery and Sicking (1995) found that this is not the case when  $E_2$  is expressed as a function of the work/volume,  $S_c$ , for creating

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 $\frac{\partial W}{\partial S_c} = -1 \tag{20}$ 

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these cracks. For a linear composite, this work is the shaded area in Fig. 4; the bottom dashed line is not necessarily the unloading line. It should be emphasized that this work of damage is not simply equal to the critical energy release rate times the total crack surface area. Typically, most cracks initiate at defects that are small compared to layer thickness and then grow dynamically until they reach the free edges of the laminate and span the full layer thickness. Thus, the shaded area is greater than what one may calculate from quasi-static crack growth.

Here, we extend the earlier study (Schapery and Sicking, 1995) by giving a simple analytical demonstration of why  $E_2(S_c)$  is insensitive to the adjacent layers for an elastic composite, and then show that this insensitivity can be expected to apply in certain cases to viscoelastic composites.

By definition,  $S_c$  is given by (cf. Fig. 4),

$$S_{c} = \int_{o}^{\epsilon} E_{2}(\beta') \, \epsilon' d\epsilon' - \frac{1}{2} E_{2}(\beta) \, \epsilon^{2} \tag{16}$$

assuming that the layer is subjected to simple tensile stress normal to the fibers. Here,  $E_2\left(\beta\right)$  is the transverse modulus for the homogenized unidirectional layer. It is shown only as a function of normalized crack density, but it also depends on parameters of the adjacent layers. In order to predict  $S_c$  from Eq.(16), one needs a relationship between  $\beta$  and the strain  $\epsilon$ . The average transverse strength  $\sigma_f$  of unidirectional specimens has been found in many cases to apply to the behavior of undirectional layers which are within laminates, when the shear stress along the fibers is not large. Namely, a transverse crack initiates when the internal normal stress  $\sigma_2$  reaches  $\sigma_f$ . New transverse cracks within a laminate relax the normal stress  $\sigma_2$  between them, and so additional external loading is needed to initiate further cracks. Although the crack density actually increases discontinously in time, we assume here that the process is continuous.

We have used Hashin's (1985) model for crossply laminates to predict  $E_2(\beta)$ . For small values of  $\beta$ ,

$$E_2 = E_{20} - c_1 \beta \tag{17}$$

where  $E_{20} \equiv E_2(0)$  is the undamaged modulus and  $c_1$  is a function of the thickness of adjacent layers with fibers perpendicular to the layer with transverse cracks. Substitution of Eq.(17) into Eq.(16) and then evaluating the latter equation we find for  $0 \le \epsilon - \epsilon_f << 1$ ,

$$E_2/E_{20} = 1 - S_c/W_o (18)$$

where

$$W_o \equiv \frac{1}{2} E_{20} \epsilon_f^2 \tag{19}$$

and  $\epsilon_f \equiv \sigma_f/E_{20}$  are the strain energy density and transverse strain for a unidirectional layer when  $\sigma_2$  is equal to the strength  $\sigma_f$ . It is seen that Eq.(18) does not depend on the adjacent layers. Direct calculation shows that  $E_2$  is only slightly dependent on the adjacent layers for even high crack densities.

Figure 5 from Schapery and Sicking (1995) illustrates the behavior of  $e_c \equiv E_2/E_{20}$ , as found from experiment and theory, when plotted against  $S_c$ . Observe that  $e_c$  is practically linear in  $S_c$ , as in Eq.(18), for  $e_c > 0.4$  (corresponding to  $\beta < 0.4$ ). At high crack densities the experimental values fall below Hashin's model, apparently due to local delaminations at the tip of the transverse cracks, which his model does not account for. In any event,  $S_c$  serves as an excellent correlating parameter for three significantly different laminate constructions, all the way out to specimen failure (by edge delamination). In applications requiring the prediction of  $S_c$ , one uses the equation (Schapery and Sicking, 1995),

where W is the strain energy density of the homogenized layer.

For these fiber reinforced plastics, the elastic-viscoelastic correspondence principle as described above does not apply directly, as the fiber deformations cannot be neglected. However, the relatively high axial fiber modulus leads to an approximation which enables an equation like Eq.(8) to be applied for cross-sections normal to the fibers. As described by Schapery and Sicking (1995) this leads to a correspondence principle that can be used in predicting transverse cracks and their effect on the homogenized viscoelastic behavior of unidirectional layers. For example, in Fig. 4 and Eq.(16), the strain is to be replaced by pseudo strain, using the linear viscoelastic relaxation modulus  $E_{20}(\xi)$  for the undamaged state to calculate pseudo strain. This transverse pseudo strain is,

$$\epsilon^R \equiv \frac{1}{E_o^R} \{ E_{20} d\epsilon \} \tag{21}$$

where  $E_o^R$  is the reference modulus.

As a final topic, we show that in certain cases a simple modification of the elastic analysis enables the elastic softening function  $e_c(S_c)$  and Eq.(20) to be used for viscoelastic behavior. Using the correspondence principle, we may write the transverse stress midway between a pair of transverse cracks in the form

$$\sigma_m = f_m(\beta) E_o^R \epsilon^R \tag{22}$$

where  $f_m(\beta)$  may be found from Hashin's (1985) analysis for elastic crossply composites. The function  $f_m$  decreases monotonically in  $\beta$ , with  $f_m(0) \simeq 1$ . That the transverse cracking occurs locally when  $\sigma_m = \sigma_f$ , as discussed above for the elastic composite, leads us to assume that the relevant critical initial defects are microcracks that are small compared to layer thickness and are distributed uniformly over distances as small as the spacing between the closest transverse cracks realized. For the case of rate-dependent crack growth we assume for simplicity that these very small defects are penny-shaped, with a radial growth rate of  $\dot{a} \sim K_I^q$ , where  $K_I$  is the stress intensity factor for an isolated crack; this representation is equivalent to using an energy release rate or J integral based on a pseudo strain energy. Inasmuch as  $K_I \sim \sigma_m$  and assuming q >> 1, we find for a monotone increasing  $\epsilon^R$  that a given microcrack becomes unstable (and grows quickly into a full transverse crack) at the (nondimensionalized) time  $\hat{t}$ , where

$$\sigma_m = C_f \hat{t}^{-p} \tag{23}$$

Also, p = 1/q and  $C_f$  is a constant; when  $q \to \infty$ , this result reduces to the strength criterion used earlier. In view of Eq.(22),

$$C_f \hat{t}^{-p} = f_m(\beta) E_o^R \epsilon^R \tag{24}$$

where  $\epsilon^R$  is the transverse strain for which a transverse crack pops-in at the time  $\hat{t}$ .

The relationship between  $S_c$ ,  $\beta$  and  $\hat{t}$  may be found by solving Eq.(24) for  $\epsilon^R$  and substituting it into Eq.(16), where  $E_0^R = E_2(0)$ ,

$$S_{c}\hat{t}^{2p} = -\left(\frac{C_{f}}{E_{0}^{R}}\right)^{2} \left[ \int_{0}^{\beta} E_{2}\left(\beta'\right) f_{0}^{-3}\left(\beta'\right) \frac{df_{0}}{d\beta'} d\beta' + \frac{1}{2} E_{2}\left(\beta\right) f_{0}^{-2}\left(\beta\right) - \frac{E_{0}^{R}}{2} \right]$$
(25)

This result is formally the same as for an elastic material, with  $E_2(\beta)$  from elasticity theory, except  $\hat{S}_c \equiv S_c \hat{t}^{2p}$  appears in place of  $S_c$  and  $C_f$  replaces  $\sigma_f$ . Thus, the results in Fig. 5

$$\frac{\partial W^R}{\partial \hat{S}_c} = -\hat{t}^{-2p} \tag{26}$$

where  $W^R$  is the strain energy density in terms of pseudo strains. This equation implies that for a given state of pseudo strain, the extent of transverse cracking increases with increasing time, as expected.

## CONCLUDING REMARKS

Viscoelastic behavior of polymer composites often exhibits a considerable amount of nonlinearity. Here, we have considered only that nonlinearity which is due to damage growth. The experimental results on particle-filled rubber provide considerable support for the approach used. Furthermore, the predictions of transverse cracking in laminates are encouraging in that they indicate possible simplifications can be achieved in material characterization by appropriate choice of variables, viz. pseudo strains and  $\hat{S}_c$  for the measure of transverse cracking. What is needed at this time are experimental studies of viscoelastic laminates that can be used to assess existing models and guide further theoretical development.

## ACKNOWLEDGMENT

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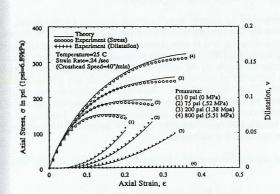


Fig. 1. Stress-strain-dilatation behavior of filled rubber.

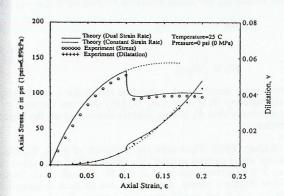


Fig. 2. Response of filled rubber in dual strain rate test (0.12/s, 0.003/s).

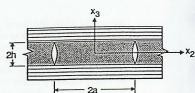


Fig. 3. Layer with transverse cracks.

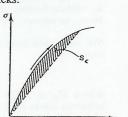


Fig. 4. Stress-strain curve for uniaxial loading.

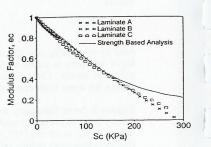


Fig. 5. Effect of transverse cracking on transverse modulus from theory and experiment.