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AN EVALUATION OF STRAIN AMPLIFICATION CONCEPTS VIA MONTE-CARLO SIMULATIONS OF AN IDEAL COMPOSITE

BY

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An Evaluation of Strain Amplification Concepts via Monte-Carlo Simulations of an Ideal Composite

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§ Abstract

In the modeling of carbon-black filled elastomers it is important to have a good estimate of the state of the elastomer itself, since many nonlinear effects originate in the matrix material. A common notion in such estimates is the idea of a "strain amplification" factor that relates a macroscopically imposed strain state to the average strain state in the elastomer matrix material. In this paper the Guth-Gold strain amplification factor, Smallwood's strain amplification factor, and a more recent proposal by Govindjee and Simo will be examined. All three theories are compared to the results of a series of Monte-Carlo simulations on an ideal composite with a Neo-Hookean matrix and semi-rigid inclusions. It is shown that for the idealized material, one can not interpret the the Guth-Gold and Smallwood strain amplification factors as an estimate of the state of the matrix material. The theory of Govindjee and Simo, on the other hand, is shown to accurately predict the state of the matrix.

§1. Introduction

An important trend in the development of (mechanical) constitutive models in general is the incorporation of micro- and meso-scale phenomena into macroscale models. When dealing with carbon-black filled elastomers this entails a knowledge of the processes occurring in the various constituents and their mutual interfaces. Examples of such processes are elastomer-filler debonding, elastomer saltation over filler particles, filler aggregate breakdown, and matrix, ie. elastomer, degradation. In order to properly model these various phenomena, the individual mechanisms that give rise to them and their driving forces must be understood. For degradation phenomena that occur in the elastomeric or matrix phase of the material, one must have an accurate measure of the stress and strain fields in the elastomer phase as opposed to the corresponding fields averaged over the entire composite.

The stress and strain fields are well known to be very inhomogeneous in such materials even under nominally homogeneous macroscopic boundary conditions. For degradation processes that occur in the bulk portion of the elastomeric phase, the average stress and average strain fields in the elastomeric phase itself can be considered as driving forces depending on the specific process being studied. In the carbon-black filled elastomer literature, the most commonly used theory to determine the state of the matrix is that of GUTH & GOLD [1, 2]. This theory as interpreted by MULLINS & TOBIN [3] gives an expression for the matrix average strain in the composite material. This theory has been widely adopted and successfully applied by many authors; see eg. HARWOOD, MULLINS, & PAYNE [4], HARWOOD & PAYNE [5], or MEINECKE & TAFTAF [6].

In this paper, an evaluation of this theory and an alternative proposal by GOVINDJEE & SIMO [7] is made via a direct Monte-Carlo simulation of a random ideal composite. The outline of the paper is as follows: in Sec. 2 a brief review of some aspects of non-linear composite theory is given; in Sec. 3 the strain amplification theories of GUTH & GOLD [1,2] and GOVINDJEE & SIMO [7] are reviewed; in Sec. 4 a series of Monte-Carlo simulations are presented that examine the stress and strain fields in an ideal random composite under the states of plane-stress uniaxial extension and plane-stress simple shear; this is followed with some concluding remarks (Sec. 5).

§2. Composite Theory

Carbon-black filled elastomers can be viewed as non-linear elastic composite materials where the elastomer phase is considered the matrix material and the carbon-black filler phase is considered the inclusion material. For common applications, the distribution of the carbon-black is not intentionally ordered and thus can be approximated as random. The basic theoretical notions for modeling such materials were presented by HILL [8] and OGDEN [9]. In what follows we briefly review the basic aspects of this theory; for technical mathematical restrictions the reader is referred to CASTAÑEDA [10].

2.1. Hill-Ogden Theory.

Consider a statistically representative volume B_X centered at a point $\boldsymbol{X}$ in the undeformed configuration $\Omega \subset \mathbb{R}^3$ of the composite body of interest. Points in B_X will be labeled by their position vector $\boldsymbol{\xi}$. Also consider the set of points in $B_X^m \subset B_X$ corresponding to the matrix phase and $B_X^p = B_X \setminus B_X^m$ the set of points corresponding to the carbon-black filler. In terms of experimentally measurable quantities one is interested in the volume average values over statistically representative volumes; these are defined as

$$\overline{(\cdot)} := \frac{1}{\text{vol}(B_X)} \int_{B_X} (\cdot) d\boldsymbol{\xi} \,. \tag{2.1}$$

Also of interest will be volume averages over the matrix phase in the statistically representative volumes; these are defined as

$$\overline{(\cdot)}^m := \frac{1}{\text{vol}(B_X^m)} \int_{B_X^m} (\cdot) d\boldsymbol{\xi} \,. \tag{2.2}$$

The essence of the theory is the assumption of the existence of an effective strain energy density for the composite that is a function of the volume average deformation gradient. The form of this function is given as

$$W^*(\overline{\boldsymbol{F}}(\boldsymbol{X})) = \frac{1}{\text{vol}(B_X)} \int_{B_X} W(\boldsymbol{F}(\boldsymbol{X}), \boldsymbol{X}) \, d\boldsymbol{\xi} \,, \tag{2.3}$$

where W^* is the effective strain energy density for the composite, W is the pointwise strain energy density of the material, $\boldsymbol{F} = \frac{\partial \boldsymbol{x}}{\partial \boldsymbol{X}}$ is the deformation gradient, and $\boldsymbol{x}$ is the position of a material point in the deformed configuration that was originally located at $\boldsymbol{X}$. The effective stress-strain relation for the composite is then given as

$$\overline{\boldsymbol{P}} = \frac{\partial W^*}{\partial \overline{\boldsymbol{F}}} \,, \tag{2.4}$$

where $\overline{\boldsymbol{P}}$ is the volume average 1st Piola-Kirchhoff stress tensor. Note that in typical experimental situations it is $\overline{\boldsymbol{P}}$ and $\overline{\boldsymbol{F}}$ that are measured.

2.2. Rigid Fillers.

Because the carbon-black filler modulus is typically at least 2 orders of magnitude greater than that of the matrix phase, one can reasonably treat the filler as being rigid. In this case, we can ignore the contributions from B_X^p in (2.3) since when the derivative is taken in (2.4) there will be no contribution to the stress. Therefore, one can write

$$W^*(\overline{\boldsymbol{F}}) = (1 - v) \frac{1}{\text{vol}(B_X^m)} \int_{B_X^m} W^m(\boldsymbol{F}(\boldsymbol{X}), \boldsymbol{X}) \, d\boldsymbol{\xi} \,, \tag{2.5}$$

where W^m is the strain energy density for the elastomer phase alone and v is the volume fraction of the filler phase. If the integrand in (2.5) is expanded in a Taylor series about the average value of the deformation gradient in the matrix phase then one has that

$$W^*(\overline{\boldsymbol{F}}) \approx (1-v) W^m(\overline{\boldsymbol{F}}^m) + \frac{1}{\text{vol}(B_X^m)} \int_{B_X^m} (\overline{\boldsymbol{F}}^m - \boldsymbol{F}(\boldsymbol{X})) : \frac{\partial^2 W^m}{\partial \boldsymbol{F}^2}(\overline{\boldsymbol{F}}^m) : (\overline{\boldsymbol{F}}^m - \boldsymbol{F}(\boldsymbol{X})) \, d\boldsymbol{\xi} \,. \tag{2.6}$$

When the stress is computed from (2.6), the first term on the right gives a lower bound estimate to the composite response and the second term accounts for the mean square fluctuations in the deformation gradient field that occur because of the presence of the filler phase. In order to use (2.6) in (2.4), one needs a relationship between the average deformation gradient in the composite, $\overline{\boldsymbol{F}}$, and the average deformation gradient in the matrix material, $\overline{\boldsymbol{F}}^m$; ie. a strain amplification relation.

§3. Strain Amplification

The literature contains many different theories for the calculation of strain amplification relations. Within the theory of large deformation composites there are few analytical calculations not restricted to second order expansions other than the result of GOVINDJEE & SIMO [7]. Most expressions are derived from the theory of liquid suspensions in the fashion pioneered by EINSTEIN [11]. These expressions are then taken to hold for small deformation elasticity due to the formal mathematical analogy between Stokes flow and incompressible small deformation elasticity. The idea of how one applies such relations to large-deformation problems was successfully introduced by MULLINS & TOBIN [3].

3.1. Guth and Gold.

The strain amplification result knows as the GUTH-GOLD relation is of the form

$$E^* = E^m(1 + 2.5v + \beta v^2)\,, \tag{3.1}$$

where E^* is the effective (small strain) Young's modulus of the composite, E^m is the Young's modulus of the matrix material, and β is scalar constant. The exact value of β depends on the assumed interactions between the filler particles. BATCHELOR & GREEN [12] give $\beta = 5.2 \pm 0.3$; CHEN & ACRIVOS [13,14] give $\beta \approx 5.01$; GUTH & SIMHA [15] give $\beta = 7.79$; GUTH & GOLD [2] give $\beta = 20.35$; and GUTH [1] gives $\beta = 14.1$. This last value is the most commonly adopted number in the rubber literature. Note that most of these calculations were originally performed for the determination of the effective viscosity of fluids with particle suspensions. Within the theory of small deformation elasticity, the linear term was apparently first computed by SMALLWOOD [16].

Within the context of nonlinear elasticity MULLINS & TOBIN [3] proposed using (3.1) through the following argument: In (1-D) small deformation theory the average stress $\overline{\sigma}$ is related to the average strain $\overline{\epsilon}$ by the relation $\overline{\sigma} = E^*\overline{\epsilon} = E^m\overline{\epsilon}(1 + 2.5v + \beta v^2)$. They further considered writing this as $\overline{\sigma} = E^m\epsilon^*$, where $\epsilon^* = \overline{\epsilon}(1 + 2.5v + \beta v^2)$ is a modified strain measure. To apply this idea to finite deformations they applied the notion of a modified strain measure to the average "principal strains" $\overline{\epsilon_A} = \overline{\lambda_A} - 1$, where $\overline{\lambda_A}$ are the average (macroscopic) principal stretches of the deformation. The resultant expression for the average principal 1st Piola-Kirchhoff stress was thus given as

$$\overline{P_A} = \left.\frac{\partial W^m}{\partial \lambda_A}\right|_{\overline{\lambda_B}=\lambda_B^*}\,, \tag{3.2}$$

where $\lambda_B^* = (\overline{\lambda_B} - 1)(1 + 2.5v + \beta v^2) + 1$ was interpreted as the average stretch in the matrix material; ie. they identified λ_B^* as $\overline{\lambda_B}^m$. Note, that as presented this theory is only applicable to isotropic materials.

3.2. Govindjee and Simo.

In the paper of GOVINDJEE & SIMO [7], an alternative to the GUTH-GOLD strain amplification result is derived by assuming that the particles are rigid and have affine *rotation*. Note that this assumption does *not* imply affine displacements. In general, the displacements in this theory are non-affine; only the rotation of the particles is taken to be affine (with respect to the volume average deformation gradient). The resultant expression for the average matrix deformation gradient is given as

$$\overline{\boldsymbol{F}}^m = \frac{\overline{\boldsymbol{F}} - v\overline{\boldsymbol{R}}}{1-v}, \tag{3.3}$$

where $\overline{\boldsymbol{R}}$ is the rotation in the (unique) polar decomposition of $\overline{\boldsymbol{F}}$; see e.g. GURTIN [17]. In principal stretches this reduces to

$$\overline{\lambda_A}^m = \frac{\lambda_A - v}{1-v}. \tag{3.4}$$

Remark 3.1.

This result is the 3-D counterpart to BUECHE'S [18] expression:

$$\overline{\lambda}^m = \frac{\lambda - v^{1/3}}{1 - v^{1/3}}. \tag{3.5}$$

Note that BUECHE'S result is exact in 1-D if one interprets the terms $v^{1/3}$ as the line fraction of rigid segments in a 1-D composite rod. □

§4. Monte-Carlo Simulations

The determination of the validity of these theories in terms of predicting the state of the matrix material is very difficult from an experimental point of view. A suitable alternative for considering their validity is the use of numerical instead of physical experiments. Not only does this circumvent the need for difficult experiments, it permits one to experiment on ideal materials where the constitutive relations for the individual components are exactly known. In what follows we will directly simulate the physical behavior of small statistically representative composite specimens under plane-stress loading conditions for uniaxial extension and simple shear. The simulations will be carried out using the finite element method. Note that since the composite materials we are interested in have a random microstructure, one can not simply perform the numerical experiments on a single distribution of particle in an elastomeric matrix. This would not be representative of the macroscopic behavior of real materials. In what follows we will properly compute the needed phase space averages of the quantities of interest by Monte-Carlo integration.

The geometry of the problem will be idealized as a 50 × 50 cell square array as shown in Fig 4.1. The individual cells are either elastomer or carbon-black and are 500Å × 500Å;

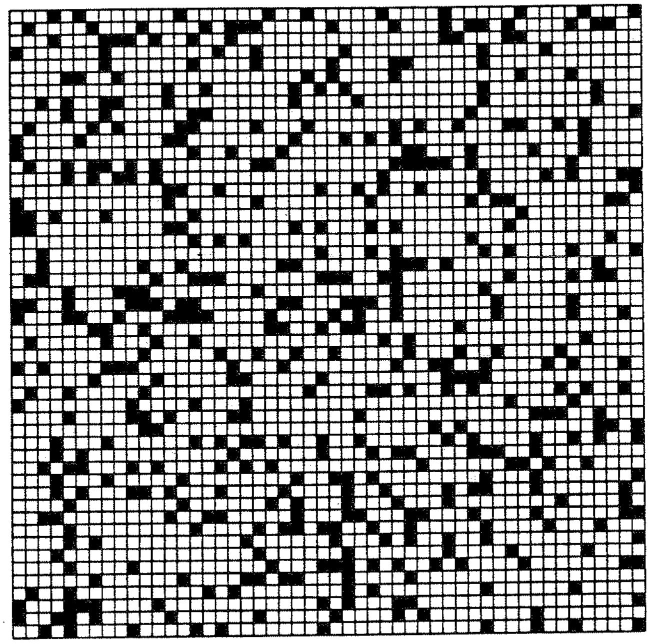

FIGURE 4.1. Sample element of the composite phase space. Dark regions indicate carbon-black filler at 20% volume fraction and light regions denote elastomer.

thus the entire specimen is $2.5\mu\text{m} \times 2.5\mu\text{m}$. The elastomer is modeled as an incompressible Neo-Hookean material

$$W^m = \frac{\mu}{2}(\text{tr}\,[\boldsymbol{C}] - 3)\,, \tag{4.1}$$

where $\text{tr}\,[\cdot]$ is the trace operator and $\boldsymbol{C} = \boldsymbol{F}^T\boldsymbol{F}$ is the right Cauchy-Green strain tensor. For all simulations, the value of $\mu = 0.6$ GPa was used. The carbon particles were also modeled for convenience as (incompressible) Neo-Hookean with a shear modulus of $\mu = 150$ GPa. Note that the carbon always responded in the linear range; thus this was an acceptable model choice.

The elements of phase space over which the averaging is to be done are different realizations of the composite specimen where the number of particles is fixed (ie. given volume fraction) and the locations are variable. If this phase space is denoted at $\mathcal{P}$, then the phase space averages of interest are given by

$$\langle(\cdot)\rangle = \frac{1}{\text{vol}(\mathcal{P})}\int_{\mathcal{P}}(\cdot)\,. \tag{4.2}$$

These are the quantities that represent the expected value of an actual physical experiment on specimens made at random. Note that the quantities of interest in evaluating the strain-amplification theories are themselves averages – volume averages. In particular, we will examine the phase space averages of the following spatial averages: $\overline{F_{22}}^m$, $\overline{\lambda_2}^m$, $\overline{P_{22}}$, $\overline{P_{22}}^m$, $\overline{\sigma_{12}}$, and $\overline{\sigma_{12}}^m$. The choice of quantities that will be examined is not intended to be exhaustive, but rather representative. It is remarked that all the components of the deformation gradient and the principal stretches have been examined and the results are qualitatively similar to those presented below.

In order to compute the phase space averages would in theory require a mechanical stress analysis of over 2500 choose $v2500$ different realizations of the composite system. For a volume fraction of 10% this represents roughly e^{800} different configurations. Clearly this is infeasible. Thus integrals of the form (4.2), are integrated using the Monte-Carlo approximation; ie.

$$\langle (\cdot) \rangle \approx \{(\cdot)\}_N = \frac{1}{N} \sum_{i=1}^{N} (\cdot)_i$$

where the number N represents the number of random samples (realizations) utilized in computing the phase space average. The error in such a scheme is well known to be approximately of the form

$$E_{int} \approx \sqrt{\frac{\{(\cdot)^2\}_N - \{(\cdot)\}_N}{N}}. \tag{4.3}$$

Thus the convergence of the scheme is $O(N^{-1/2})$. The interested reader may consult any text on statistics; see eg., MARTIN [19]. Amazingly, very few samples are required to accurately converge the integration scheme for the problems at hand. For the simulations shown below, error bars of $\pm E_{int}$ have been plotted on all points determined via Monte-Carlo integration. Note that in most cases the integration errors are too small to be discernible at the scales shown. For all simulations a value of $N = 100$ was used; in most cases, however, a value of $N = 10$ would have been quite adequate.

To efficiently collect the Monte-Carlo statistics the following scheme was utilized. First a sample was drawn from the phase space. This was done by considering each cell in the composite and drawing a random number between 0 and 1 using a maximal linear congruential random number generator on a double precision word. For cells where the random number was below the desired volume fraction of particles, its material properties were set to those of carbon; the others were set to elastomer. Note that this results in a composite with a small variation in volume fraction about the desired value. This error is, however, too small to effect the results presented here. Next, for each realization, a series of nonlinear finite element analyses are performed at different mechanical load levels. At a given load level, the nonlinear finite element problem is solved, the desired spatial averages are computed, and the phase space averages updated. The mechanical load level is then increased and the averages recomputed. Only once the largest mechanical load level desired has been reached is another sample drawn from the phase space. This process is repeated until the error in the phase space averages reaches the desired level.

Remark 4.1.

The calculations were performed using the finite element code FEAP developed at UC Berkeley and partially documented in the text of ZIENKIEWICZ & TAYLOR [20,21]. The Monte-Carlo collection routines are easily implemented in this code utilizing the user definable solution macro procedures, `umacr`. This allows for very efficient calculations since data does not need to be passed to an external program to compute the Monte-Carlo averages. Note that the element used was a user defined exact incompressible plane-stress element with a pure displacement formulation. The reader is reminded that, in plane-stress, element locking due to incompressible material behavior does not occur. □

4.1. Uniaxial Extension: Strains.

The first test consists of a simple uniaxial extension of the sample in the 2-direction. The boundary conditions on the sample conformed to macroscopic incompressibility. If the lower left-hand corner of the sample is taken as the origin of a Cartesian coordinate system, (X_1, X_2), then the imposed displacement boundary conditions were given by:

$$u_1 = (\frac{1}{\sqrt{\overline{\lambda}}} - 1)X_1 \quad \text{and} \quad u_2 = (\overline{\lambda} - 1)X_2 \tag{4.4}$$

at $X_1 = 0, 2.5\mu$m and $X_2 = 0, 2.5\mu$m. $\overline{\lambda}$ is the imposed macroscopic (average) axial stretch on the sample.

Shown in Figs. 4.2 and 4.3 is the matrix average axial stretch, $\langle\overline{F_{22}}\rangle$, versus filler volume fraction, v; Fig. 4.2 is for $\overline{\lambda} = 1.05$ and Fig. 4.3 is for $\overline{\lambda} = 1.5$. The curves correspond to the theories of Guth & Gold (GG) with $\beta = 14.1$, Govindjee & Simo (GS), and Smallwood (SW). The data points are the results of the Monte-Carlo simulations with error bars. Note the excellent agreement between the GS-theory and the strain in the matrix material and the large over-prediction according to the GG-theory. In uniaxial extension one does not expect large rotations of the carbon-black particles at the strain levels shown. Thus, the affine rotation assumption in the GS-theory should be expected to work well. A more demanding situation is that of simple shear where there is appreciable rotation.

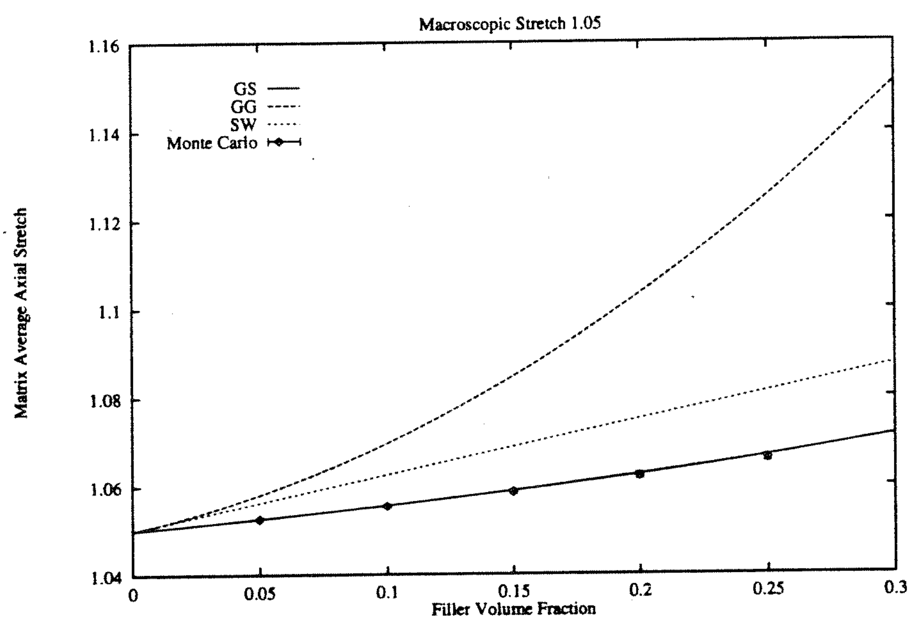

FIGURE 4.2. Variation of average axial stretch in the matrix material as a function of filler volume fraction at a macroscopic stretch of 1.05.

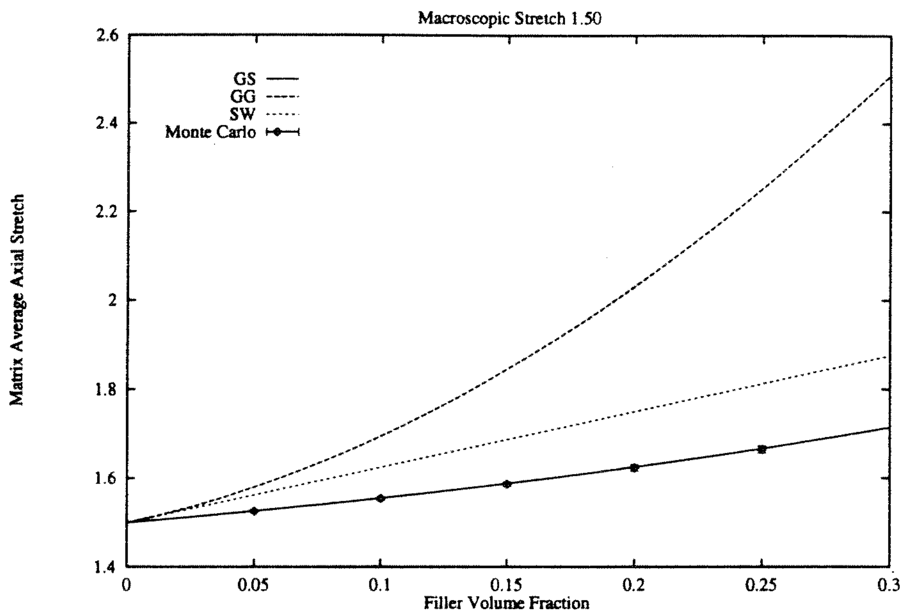

FIGURE 4.3. Variation of average axial stretch in the matrix material as a function of filler volume fraction at a macroscopic stretch of 1.5.

4.2. Simple Shear: Strains.

In this example, the sample is deformed in simple shear. The imposed boundary conditions were given by:

$$u_1 = kX_2 \quad \text{and} \quad u_2 = 0 \tag{4.5}$$

at $X_1 = 0, 2.5\mu\text{m}$ and $X_2 = 0, 2.5\mu\text{m}$. k is the imposed macroscopic "shear slope" of the sample; ie. the engineering shear strain.

Shown in Figs. 4.4 and 4.5 is the matrix average second principal stretch (minimum in the plane of the sample) versus the shear slope k; Fig. 4.4 is for a volume fraction of $v = 0.05$ and Fig. 4.5 is for a volume fraction of $v = 0.20$. The curves and data points are as in the previous example. Note once again the good agreement between the GS-theory for the average behavior of the matrix material and the Monte-Carlo data. The small variation between the GS-theory and the data points indicates the approximate character of the affine rotation assumption at the filler and mechanical loading levels shown. Note that the second principal stretch is representative of the behavior of the other kinematic quantities in this problem.

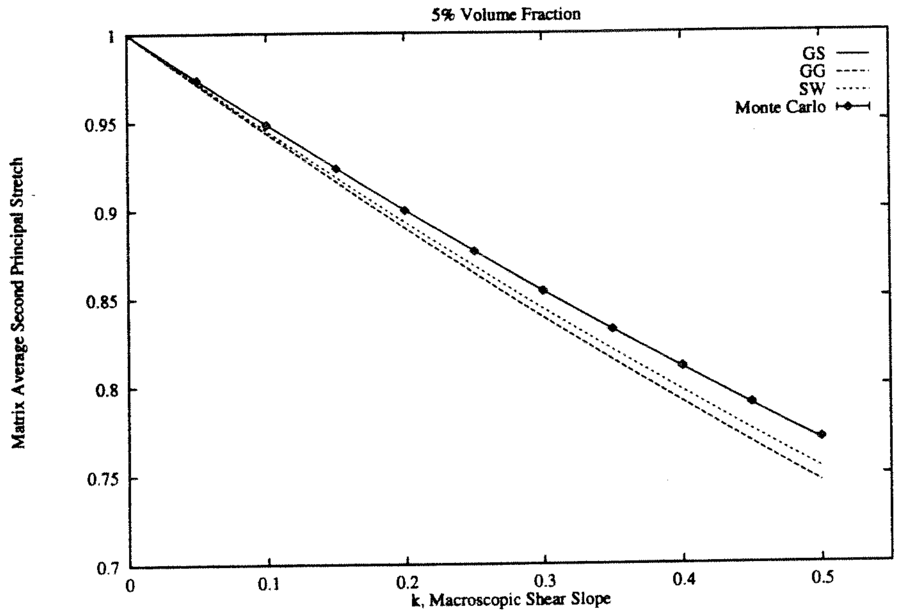

FIGURE 4.4. Variation of average second principal stretch in the matrix material as a function of macroscopic shear slope for a volume fraction of 5%.

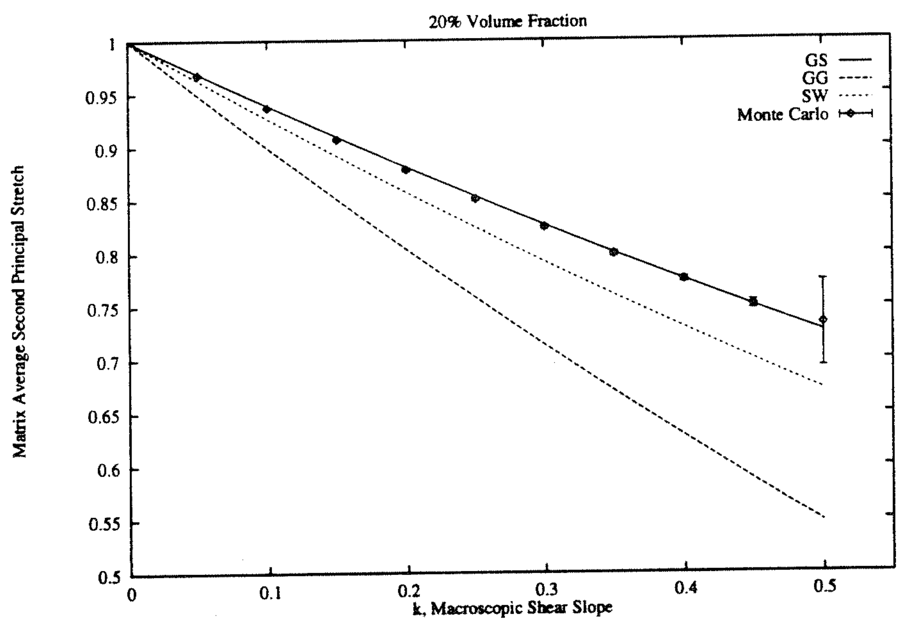

FIGURE 4.5. Variation of average second principal stretch in the matrix material as a function of macroscopic shear slope for a volume fraction of 20%.

4.3. Uniaxial Extension: Stresses.

The two previous examples were concerned with the ability of the theories to predict the average strain state of the matrix material under given macroscopic loading conditions. We now turn our attention to the behavior of the stresses in the sample. For the case of uniaxial extension, Figs 4.6 and 4.7 show the average axial 1st Piola-Kirchhoff stress in the composite and in the matrix, $\langle\overline{P_{22}}\rangle$ and $\langle\overline{P_{22}}^m\rangle$, versus the imposed macroscopic axial stretch, $\overline{\lambda}$, for volume fractions of 5% and 15%. The curves denoted GG and SW are plotted using the MULLINS & TOBIN relation (3.2) with $\beta = 14.1$ and $\beta = 0$, respectively. For uniaxial extension this gives

$$\overline{P_{22}} = \mu\left(\lambda^* - \frac{1}{(\lambda^*)^2}\right), \tag{4.6}$$

where $\lambda^* = (\overline{\lambda} - 1)(1 + 2.5v + \beta v^2) + 1$. The curve denoted GS is relation (2.4) where W^* is taken as the first term of (2.6) and the strain amplification is given by (3.3). Because of the assumed incompressibility, this gives

$$\overline{P_{22}} = \mu\left(\frac{\overline{\lambda} - v}{1 - v} - (\overline{\lambda})^{-3/2}\frac{(\overline{\lambda})^{-1/2} - v}{1 - v}\right), \tag{4.7}$$

The Monte-Carlo data points for $\langle\overline{P_{22}}\rangle$ are given by the diamond markers and the data points for $\langle\overline{P_{22}}^m\rangle$ are given by the horizontal-dash markers. Note that in computing the spatial averages, one must exclude a roughly 1000 Å boundary layer. This boundary layer contains a strong Saint-Venant effect.

As seen from the figures, the theory of MULLINS & TOBIN provides roughly the correct overall stress-strain response of the sample. Though is it clear that the value of β would have to be concentration dependent for good agreement; note that for the case of 30% volume fraction (not shown) the Monte-Carlo data points move to the other side of the GG-theory curve. In terms of predicting the average stress state of the matrix material, the GS-theory using only the first term in (2.6) appears to provide good agreement. The difference between the GS-theory and the overall composite stress values is then also seen to be a measure of the strength of the higher order terms in (2.6) (ie. the mean square fluctuations of the strain field).

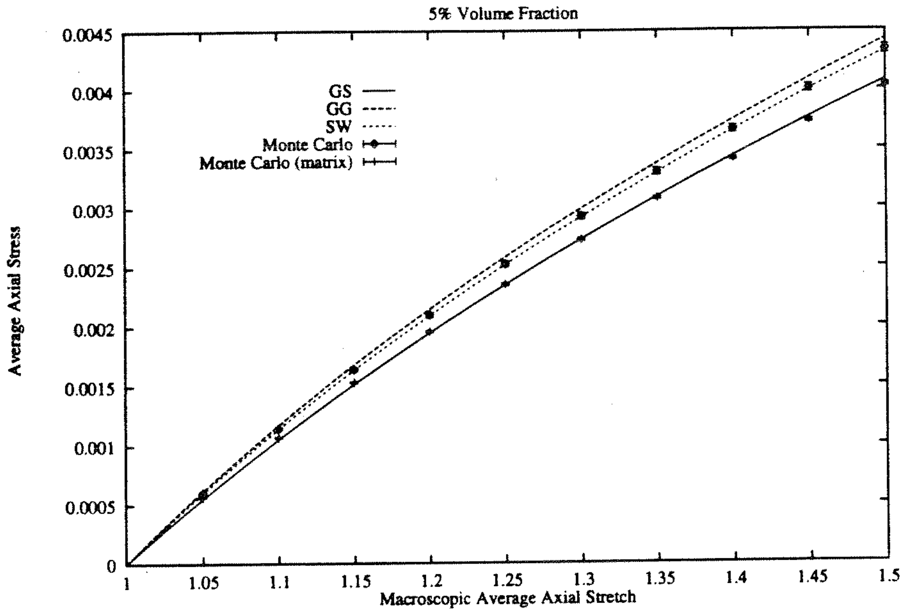

FIGURE 4.6. Variation of average axial stresses versus average macroscopic stretch for a volume fraction of 5%; stress in units of $N/(2.5\mu m)^2$.

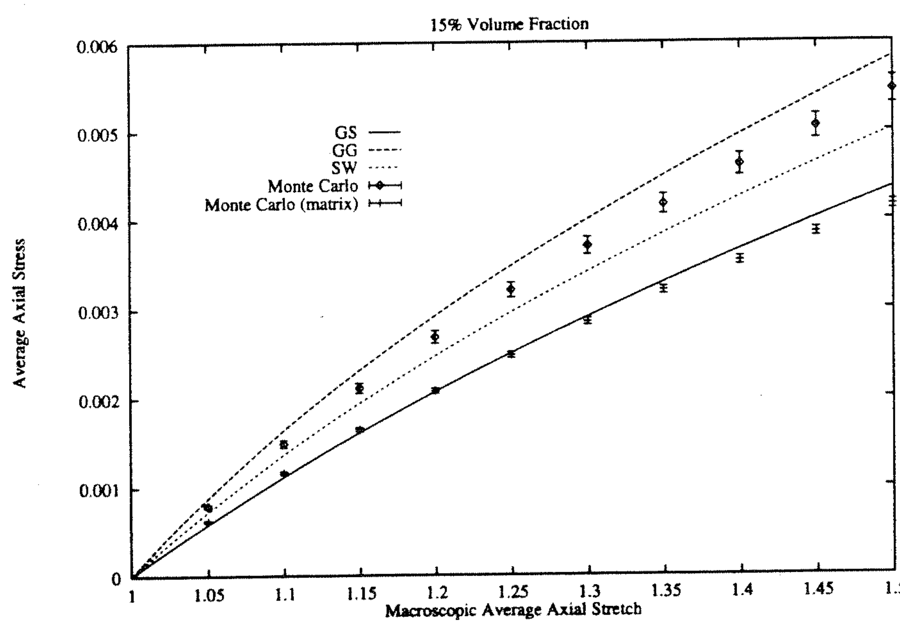

FIGURE 4.7. Variation of average axial stress versus average macroscopic stretch for a volume fraction of 15%; stress in units of $N/(2.5\mu m)^2$.

4.4. Simple Shear: Stresses.

For simple shear, we consider the Cauchy shear stresses in the material. To compute the shear stress value from the GS-theory, proceed as follows: Note first that when only the leading order term in (2.6) is used, then

$$\overline{\boldsymbol{P}} = \mu \sum_{B=1}^{3} \overline{\lambda_B}^m \boldsymbol{n}_B \otimes \boldsymbol{N}_B + p\overline{\boldsymbol{F}}^{-T} , \tag{4.8}$$

where $\boldsymbol{n}_B$ and $\boldsymbol{N}_B$ are the Eulerian and Lagrangian principal directions and are the Eigenvectors of $\overline{\boldsymbol{V}} = \sqrt{\overline{\boldsymbol{F}}\,\overline{\boldsymbol{F}}^T}$ and $\overline{\boldsymbol{U}} = \sqrt{\overline{\boldsymbol{F}}^T\overline{\boldsymbol{F}}}$, respectively; p is the pressure from the incompressibility constraint. If one converts the 1st Piola-Kirchhoff stress to Cauchy stress, then one has

$$\overline{\boldsymbol{\sigma}} = \frac{1}{\overline{J}}\overline{\boldsymbol{P}}\,\overline{\boldsymbol{F}}^T = \mu \sum_{B=1}^{3} \overline{\lambda_B}^m \overline{\lambda_B} \boldsymbol{n}_B \otimes \boldsymbol{n}_B + p\boldsymbol{1} , \tag{4.9}$$

where $\overline{J} = \det[\overline{\boldsymbol{F}}] = 1$ and $\boldsymbol{1}$ is the identity tensor. The plane-stress condition $\overline{\sigma_{33}} = 0$ gives the pressure $p = \mu$. Further, the Eulerian Eigenvectors are given (in the original Cartesian coordinate system) by $\boldsymbol{n}_1 = (\cos\theta_E \quad \sin\theta_E \quad 0)^T$, $\boldsymbol{n}_2 = (-\sin\theta_E \quad \cos\theta_E \quad 0)^T$, and $\boldsymbol{n}_3 = (0 \quad 0 \quad 1)^T$, where $\theta_E = \frac{1}{2}\tan^{-1}(2/k)$; see eg. OGDEN [22]. Thus the shear stress in our coordinate system is

$$\overline{\sigma_{12}} = \mu \frac{\overline{\lambda_1}^m \overline{\lambda_1} - \overline{\lambda_2}^m \overline{\lambda_2}}{\sqrt{4+k^2}} , \tag{4.10}$$

where $\overline{\lambda_1}^2, \overline{\lambda_2}^2 = 1 + (k^2/2) \pm k\sqrt{1+(k^2/4)}$.

The computation of the shear stress from the MULLINS & TOBIN theory is not uniquely defined. In their prescription they indicate that one should replace the stretches in the stress-stretch relationship by the amplified stretches. The question, however, arises as to which stress-stretch relationship – the one for the 1st Piola-Kirchhoff stress, the Cauchy stress, the Kirchhoff stress, etc? Depending on which stress-stretch relation one chooses, different results are obtained. Keeping with the cases shown in MULLINS & TOBIN [3] and used above for the uniaxial case, we apply the amplified strains in the relation for the 1st Piola-Kirchhoff stress and then convert the result to Cauchy stress; this is also the same technique that the GS-theory uniquely prescribes. In this case, we are lead to

$$\overline{\sigma_{12}} = \mu \frac{\lambda_1^* \overline{\lambda_1} - \lambda_2^* \overline{\lambda_2}}{\sqrt{4+k^2}} . \tag{4.11}$$

Figs. 4.8 and 4.9 show, in the Eulerian frame defined by $\boldsymbol{n}_B$, the average shear stress in the composite, $\overline{\sigma_{12}}$, and the average shear stress in the matrix material, $\overline{\sigma_{12}}^m$, versus shear slope for volume fractions of 10% and 30%. The curves denoted GG and SW are plotted using (4.11) with $\beta = 14.1$ and $\beta = 0$, respectively. The curve denoted GS is (4.10). As seen from the figures, the agreement between the theories and the Monte-Carlo data is not good. Note also that the good agreement between the GS-theory and the stress state of the matrix itself disappears in the simple shear mode of deformation; though its utility as a weak lower bound remains.

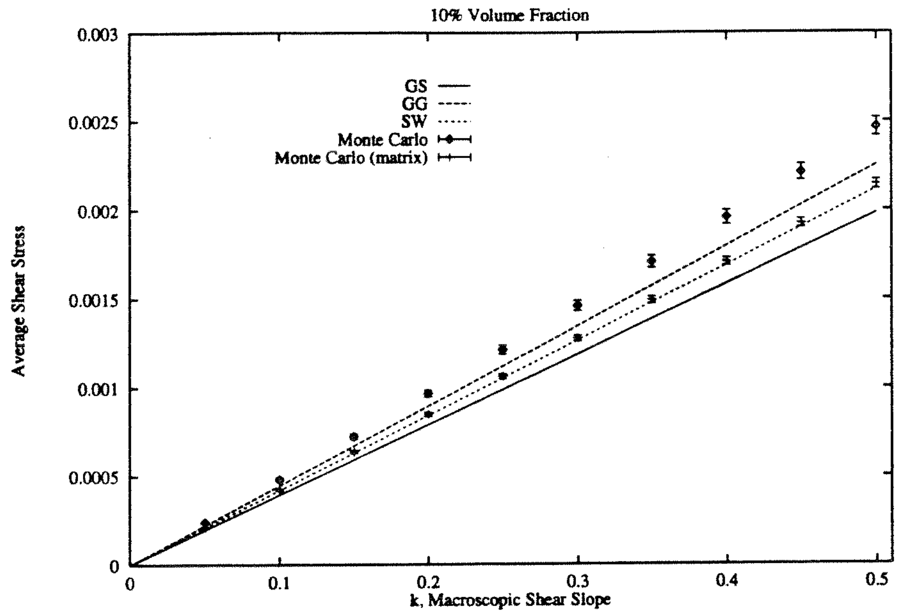

FIGURE 4.8. Variation of average shear stress versus average macroscopic shear angle for a volume fraction of 10%; stress in units of $N/(2.5\mu m)^2$.

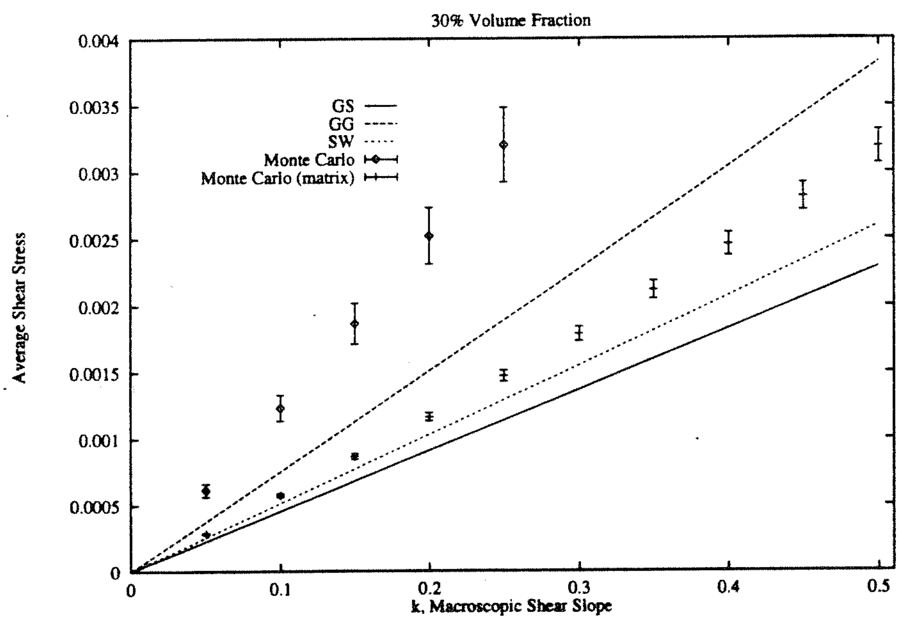

FIGURE 4.9. Variation of average shear stress versus average macroscopic shear angle for a volume fraction of 30%; stress in units of $N/(2.5\mu m)^2$.

§5. Conclusions

This study has shown that for the ideal Neo-Hookean† (lattice) composite material the GS-strain amplification theory gives a very accurate measure of the average strain state of the matrix material in a two-phase composite when the filler-phase is approximately rigid. This agreement is predictive at least up to 50% extension and shear for volume fractions below 30%. Beyond this range more sophisticated methods of numerical or physical experimentation will be needed to determine the theory's applicability. In terms of predicting stress-strain response of the composite it is seen that the prescriptions of GOVINDJEE & SIMO [7] and MULLINS & TOBIN [3] are lacking; though the GS-theory can be used as a loose lower bound. The GG-theory does not provide any bounding properties. Thus for the modeling of strain activated processes in carbon-black filled elastomers the GS-theory of strain amplification appears to be the correct choice. For the modeling of processes that depend on stress activation or on a combination of stress and strain activation, the correct theory appears to still be an open question.

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† Similar results have also been obtained using the recent model of GENT [23] which possesses an upturn in the stress-strain relation.

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