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Second-order estimates for the large-deformation response of particle-reinforced rubbers

Estimations homogénéisées pour le comportement mécanique des caoutchoucs renforcés en grandes déformations

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Abstract

This paper presents the application of a recently proposed 'second-order' homogenization method (J. Mech. Phys. Solids 50 (2002) 737–757) to the estimation of the effective behavior of hyperelastic composites subjected to *finite* deformations. The main feature of the method is the use of 'generalized' secant moduli that depend not only on the phases averages of the fields, but also on the phase covariance tensors. The use of the method is illustrated in the context of particle-, or fiber-reinforced elastomers and estimates analogous to the well-known Hashin–Shtrikman estimates for linear-elastic composites are generated. The new estimates improve on earlier estimates (J. Mech. Phys. Solids 48 (2000) 1389–1411) neglecting the use of fluctuations. In particular, the new estimates, unlike the earlier ones, are capable of recovering the exact incompressibility constraint when the matrix is also taken to be incompressible. ***To cite this article: O. Lopez-Pamies, P. Ponte Castañeda, C. R. Mecanique 331 (2003).***
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Résumé

Cet article présente l'application d'une méthode d'homogénéisation récente, dite de deuxième ordre (J. Mech. Phys. Solids 50 (2002) 737–757), afin d'estimer le comportement effectif de matériaux composites hyperélastiques sujets à des grandes déformations. La principale caractéristique de cette méthode est l'utilisation d'un module sécant généralisé par phase qui dépend non seulement des moyennes des champs par phase mais aussi des tenseurs de covariance des fluctuations des phases. L'utilisation de cette procédure est illustrée dans le cas particulier d'élastomères renforcés à particules ou à fibres. Ces nouvelles estimations généralisent celles bien connues de Hashin–Shtrikman obtenues pour des composites élastiques linéaires et améliorent certaines estimations décrites antérieurement (J. Mech. Phys. Solids 48 (2000) 1389–1411) qui, elles, négligeaient l'existence de fluctuations des champs. En particulier, ces nouvelles estimations, contrairement a celles étudiées précédemment, sont capables d'établir la contrainte exacte d'incompressibilité lorsque la matrice est elle aussi supposée incompressible. ***Pour citer cet article : O. Lopez-Pamies, P. Ponte Castañeda, C. R. Mecanique 331 (2003).***
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1. Introduction

The aim of this paper is to develop estimates for the effective behavior of *hyperelastic* composite materials subjected to *finite deformations*. The materials are made up of N different (homogeneous) phases, which are assumed to be distributed *randomly* in a specimen occupying a volume Ω_0 in the reference configuration. Furthermore, the size of the typical inhomogeneity (e.g., particle, or void) is much smaller than the size of the specimen and the scale of variation of the loading conditions. The constitutive behavior of the phases is characterized by stored energies $W^{(r)}$ $(r = 1, \ldots, N)$ that are *nonconvex* functions of the deformation gradient $\mathbf{F}$, which is required, in turn, to satisfy the *impenetrability* condition: $\det \mathbf{F}(\mathbf{X}) > 0$ for $\mathbf{X}$ in Ω_0. The local or microscopic constitutive relation for the composite is given by

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X}, \mathbf{F}), \quad W(\mathbf{X}, \mathbf{F}) = \sum_{r=1}^{N} \theta^{(r)}(\mathbf{X})\ W^{(r)}(\mathbf{F}) \tag{1}$$

where $\mathbf{S}$ denotes the first Piola–Kirchhoff stress tensor, and the functions $\theta^{(r)}$ are such that $\theta^{(r)} = 1$ if $\mathbf{X} \in \Omega_0^{(r)}$ and 0 otherwise.

Following Hill [1], the effective or macroscopic constitutive relation for the hyperelastic composite is defined by

$$\overline{\mathbf{S}} = \frac{\partial \widetilde{W}}{\partial \overline{\mathbf{F}}}, \quad \widetilde{W}(\overline{\mathbf{F}}) = \min_{\mathbf{F} \in \mathcal{K}(\overline{\mathbf{F}})} \big\langle W(\mathbf{X}, \mathbf{F}) \big\rangle = \min_{\mathbf{F} \in \mathcal{K}(\overline{\mathbf{F}})} \sum_{r=1}^{N} c^{(r)} \big\langle W^{(r)}(\mathbf{F}) \big\rangle^{(r)} \tag{2}$$

where $\overline{\mathbf{S}} = \langle \mathbf{S} \rangle$ is the *average stress*, $\overline{\mathbf{F}} = \langle \mathbf{F} \rangle$ is the *average deformation gradient* and $\widetilde{W}$ is the *effective stored-energy function* of the composite. In these expressions, the symbols $\langle \cdot \rangle$ and $\langle \cdot \rangle^{(r)}$, denote volume averages over the composite (Ω_0) and over phase r $(\Omega_0^{(r)})$, respectively, so that the scalars $c^{(r)} = \langle \theta^{(r)} \rangle$ serve to denote the volume fractions of the given phases, and $\mathcal{K}$ denotes the set of admissible deformation gradients $\mathbf{F}$, such that there exists a deformation field $\mathbf{x} = \boldsymbol{\chi}(\mathbf{X})$ with $\mathbf{F} = \operatorname{Grad} \boldsymbol{\chi}$, $\det \mathbf{F} > 0$ in Ω_0, $\mathbf{x} = \overline{\mathbf{F}}\mathbf{X}$ on $\partial\Omega_0$. More precise definitions of the effective energy $\widetilde{W}$ are available for *periodic* microstructures [2], which generalize the classical definition of the effective energy for periodic media with convex energies, by allowing for possible interactions between unit cells, essentially by taking an infimum over all possible sets of units cells. Physically, this corresponds to accounting for the possible development of instabilities in the composite at sufficiently high deformation [3].

The focus here will be in the characterization of the effective behavior of composites made up of *rubber elastic* phases. Their stored-energy functions are objective and isotropic, so that the stored-energy functions of the phases are symmetric functions of the principal stretches $\lambda_1,\ \lambda_2,\ \lambda_3$, such that $W^{(r)}(\mathbf{F}) = \Phi^{(r)}(\lambda_1, \lambda_2, \lambda_3)$. Numerous models have been proposed in the literature for the constitutive behavior of rubbers. A simple, special class of materials, which will be considered in some detail below, is given by the *compressible* neo-Hookean material with:

$$W(\mathbf{F}) = \frac{\mu}{2}\big(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\big) + \frac{\mu'}{2}(J-1)^2 - \mu \ln J \tag{3}$$

where the parameters $\mu > 0$ and $\mu' > 0$ denote the standard Lamé moduli, and $J = \det \mathbf{F} = \lambda_1\lambda_2\lambda_3$. Note that $W(\mathbf{F}) \sim (1/2)\mu'(\operatorname{tr}\boldsymbol{\varepsilon})^2 + \mu \operatorname{tr}\boldsymbol{\varepsilon}^2$ as $\mathbf{F} \to \mathbf{I}$, so that the stored-energy function (3) linearizes properly. In addition, the limit as $\mu' \to \infty$ corresponds to incompressible behavior $J \to 1$.

The objective of this work then becomes to obtain estimates for the effective stored-energy function $\widetilde{W}$ of hyperelastic composites subjected to finite deformations. This is an extremely difficult problem, because it amounts

to solving a set of highly nonlinear partial differential equations with random coefficients. As a consequence, there are precious few *analytical* estimates for $\widetilde{W}$. An upper bound analogous to the Voigt bound in linear elasticity has been proposed by Ogden [4]. The existence of a lower bound corresponding to the Reuss bound in linear elasticity is hampered by the difficulties associated with the lack of convexity of the stored-energy functions in finite elasticity, which makes troublesome the position of a principle of minimum complementary energy. However, some non-trivial lower bounds have been proposed by Ponte Castañeda [5], exploiting the polyconvexity hypothesis. There are also numerous empirically based and *ad hoc* estimates for various special cases, including the case of rigidly reinforced rubbers [6–9]. Our aim here is to develop a general class of *analytical estimates* that are based on homogenization theory and that are applicable to large classes of composite systems, including rigidly reinforced rubbers. Such estimates should allow for the incorporation of statistical information beyond the phase volume fractions, thus allowing for a more precise characterization of the influence of microstructure on effective behavior. Some progress along these lines has been made recently [10,11] with the extension to finite elasticity of an earlier version of the 'second-order' nonlinear homogenization technique, originally developed [12] in the context of nonlinearly viscous composites with convex, nonlinear potentials.

2. The second-order variational homogenization method

In this section, a recently developed new version (Ponte Castañeda [13]) of the 'second-order' homogenization procedure is adapted to finite elasticity. This new method is a generalization of the 'linear comparison' variational method [14] in a way that incorporates many of the desirable features of an earlier version of the second-order method [12], including the fact that the estimates generated should be exact to second-order in the contrast [16]. A brief description of the proposed method is provided next.

The key idea is to introduce a 'comparison' linear composite with *thermoelastic* phases and the same microstructure (i.e., same $\theta^{(r)}$) as the nonlinear composite defined by (1). Its phase potentials are quadratic and defined by:

$$W_0^{(r)}(\mathbf{F}) = W^{(r)}\big(\mathbf{F}^{(r)}\big) + \big(\mathbf{F} - \mathbf{F}^{(r)}\big) \cdot \frac{\partial W^{(r)}}{\partial \mathbf{F}}\big(\mathbf{F}^{(r)}\big) + \frac{1}{2}\big(\mathbf{F} - \mathbf{F}^{(r)}\big) \cdot \mathbf{L}_0^{(r)}\big(\mathbf{F} - \mathbf{F}^{(r)}\big) \tag{4}$$

where $\mathbf{F}^{(r)}$ and $\mathbf{L}_0^{(r)}$ are second- and fourth-order constant tensors to be specified later. The stored-energy functions $W^{(r)}$ of the hyperelastic composite can then be *approximated* as:

$$W^{(r)}(\mathbf{F}) = W_0^{(r)}(\mathbf{F}) + V^{(r)}\big(\mathbf{F}^{(r)}, \mathbf{L}_0^{(r)}\big) \tag{5}$$

where the $V^{(r)}$ are 'error' functions defined by

$$V^{(r)}\big(\mathbf{F}^{(r)}, \mathbf{L}_0^{(r)}\big) = \operatorname*{stat}_{\widehat{\mathbf{F}}^{(r)}} \big[W^{(r)}\big(\widehat{\mathbf{F}}^{(r)}\big) - W_0^{(r)}\big(\widehat{\mathbf{F}}^{(r)}\big) \big] \tag{6}$$

It is noted that the *stat*ionarity operation with respect to a variable means taking a derivative with respect to the variable and setting the result equal to zero to generate an expression for the optimal value of the relevant variable. Since expression (5) gives an approximation for the nonlinear energy functions $W^{(r)}$ in terms of quadratic energy functions $W_0^{(r)}$ corresponding to comparison materials with linear behavior, it is expected that the error functions $V^{(r)}$ will contain information on the nonlinearity of the original energies $W^{(r)}$. Indeed, it is easy to verify that stationarity with respect to $\widehat{\mathbf{F}}^{(r)}$ in (6) leads to the relations:

$$\frac{\partial W^{(r)}}{\partial \mathbf{F}}\big(\widehat{\mathbf{F}}^{(r)}\big) - \frac{\partial W^{(r)}}{\partial \mathbf{F}}\big(\mathbf{F}^{(r)}\big) = \mathbf{L}_0^{(r)}\big(\widehat{\mathbf{F}}^{(r)} - \mathbf{F}^{(r)}\big) \tag{7}$$

These conditions can be visualized as *linearizations* of the nonlinear constitutive relations of the hyperlastic materials in each of the phases, interpolating between the deformations $\mathbf{F}^{(r)}$ and $\widehat{\mathbf{F}}^{(r)}$. They correspond to a new

type of approximation – different from the more standard 'secant' and 'tangent' approximations that have been used in the past – which has been referred to as a 'generalized secant' approximation [13].

Making use of expressions (5) in relation (2), it is possible to generate an expression for the effective energy function of the hyperelastic composite involving the effective energy function of the above-defined linear comparison composite, which is given by:

$$\widetilde{W}_0\big(\overline{\mathbf{F}};\mathbf{F}^{(s)},\mathbf{L}_0^{(s)}\big)=\min_{\mathbf{F}\in\mathcal{K}}\big\langle W_0(\mathbf{X},\mathbf{F})\big\rangle=\min_{\mathbf{F}\in\mathcal{K}}\sum_{r=1}^{N}c^{(r)}\big\langle W_0^{(r)}(\mathbf{F})\big\rangle^{(r)} \tag{8}$$

Note that this fictitious problem for a linear thermoelastic composite is one involving, in general, non-symmetric 'stress' and 'strain' measures, so that suitable generalizations [10] of the classical [15] thermoelastic analyses are required.

It follows from the form of expressions (8) with (4) that optimization with respect to the variables $\mathbf{F}^{(r)}$ and $\mathbf{L}_0^{(r)}$ in the resulting expression for $\widetilde{W}$ will involve the first moments of the deformation fields in the phases $\overline{\mathbf{F}}^{(r)}\doteq\langle\mathbf{F}\rangle^{(r)}$, as well as on the *covariance tensor* of the *deformation fluctuations* $\mathbf{C}_{\mathbf{F}}^{(r)}\doteq\langle(\mathbf{F}-\overline{\mathbf{F}}^{(r)})\otimes(\mathbf{F}-\overline{\mathbf{F}}^{(r)})\rangle^{(r)}$. In fact, it can be shown [13] that optimization with respect to the variables $\mathbf{F}^{(r)}$ and $\mathbf{L}_0^{(r)}$ in the resulting expression for $\widetilde{W}$ leads to the prescriptions:

$$\mathbf{F}^{(r)}=\overline{\mathbf{F}}^{(r)}\quad\text{and}\quad\big(\widehat{\mathbf{F}}^{(r)}-\overline{\mathbf{F}}^{(r)}\big)\otimes\big(\widehat{\mathbf{F}}^{(r)}-\overline{\mathbf{F}}^{(r)}\big)=\mathbf{C}_{\mathbf{F}}^{(r)} \tag{9}$$

In connection with these prescriptions, it is necessary to make the following clarifications. Concerning the first prescription, it should be noted that relation $(9)_1$ only makes stationary with respect to $\mathbf{F}^{(r)}$ the terms arising from the linear comparison energy $\widetilde{W}_0$. In other words, there are additional terms arising from the functions $V^{(r)}$, which have been neglected, for simplicity. Concerning the second, it needs to be emphasized that it is not possible to satisfy conditions $(9)_2$ in full generality. This is due to the fact that the left-hand side of relation $(9)_2$ is a fourth-order tensor of rank 1, whereas the right-hand side is generally of full rank. This means that only certain components (or traces) of these expressions can be enforced. This point will be discussed in more detail in the context of the specific examples considered in the applications section.

The prescriptions (9), together with condition (7), serve to completely specify the properties $\mathbf{F}^{(r)}$ and $\mathbf{L}_0^{(r)}$ of the linear comparison composite introduced above. Making use of all these conditions, the following estimate can be generated for the effective energy function of the hyperelastic composite:

$$\widetilde{W}(\overline{\mathbf{F}})=\sum_{r=1}^{N}c^{(r)}\left[W^{(r)}\big(\widehat{\mathbf{F}}^{(r)}\big)-\frac{\partial W^{(r)}}{\partial\mathbf{F}}\big(\overline{\mathbf{F}}^{(r)}\big)\cdot\big(\widehat{\mathbf{F}}^{(r)}-\overline{\mathbf{F}}^{(r)}\big)\right] \tag{10}$$

This estimate can be seen to depend not only on the phase averages $\overline{\mathbf{F}}^{(r)}$ of the deformation field in the linear 'thermoelastic' comparison composite, but also – through the variables $\widehat{\mathbf{F}}^{(r)}$, as well as the 'generalized secant' moduli $\mathbf{L}_0^{(r)}$ of the phases of the linear comparison composite – on (appropriate traces of) the deformation field *fluctuations* $\mathbf{C}_{\mathbf{F}}^{(r)}$. Furthermore, like the earlier 'second-order' estimates, they are known to be exact to second-order in the heterogeneity contrast [16].

It is remarked finally that the linear comparison problem (8) that needs to be considered for the determination of the phase averages $\overline{\mathbf{F}}^{(r)}$ and fluctuations $\mathbf{C}_{\mathbf{F}}^{(r)}$ needed in expression (10) for $\widetilde{W}$ is precisely the same that was considered by Ponte Castañeda and Tiberio [10] in the earlier version of the second-order method. These authors provided expressions of the Hashin–Shtrikman type [17] for the average deformations $\overline{\mathbf{F}}^{(r)}$ in these generalized N-phase 'thermoelastic' composites, from which estimates may be generated for the corresponding effective stored-energy functions $\widetilde{W}_0$, and, in turn, for the fluctuations using the relation $\mathbf{C}_{\mathbf{F}}^{(r)}=(2/c^{(r)})\,\partial\widetilde{W}_0/\partial\mathbf{L}_0^{(r)}$. The relevant general expressions will not be repeated here for brevity, and only the appropriate specialized versions of the results will be quoted in the applications section for the special case of rigidly reinforced systems.

3. Application to particle- and fiber-reinforced elastomers

3.1. Results for general matrix behavior and loading conditions

The second-order estimates (10) for the effective stored-energy function of hyperelastic composites apply for general N-phase systems. In this section, the special case of isotropic rigidly reinforced rubbers is considered. This case has already been considered using the earlier version of the second-order method [10] and it is considered here again using the earlier results as a reference. Thus, the focus will be on two-phase composites consisting of rigid, spherical inclusions distributed isotropically with volume fraction $c^{(2)} = c$ in a hyperelastic matrix with energy function $W^{(1)} = W$, such that the composite is statistically isotropic in the undeformed configuration.

Because of the objectivity of $\widetilde{W}$, it suffices to consider macroscopic stretch loading histories, such that $\overline{\mathbf{F}} = \overline{\mathbf{U}}$, $\overline{\mathbf{R}} = \mathbf{I}$, where $\overline{\mathbf{U}}$ and $\overline{\mathbf{R}}$ are the stretch and rotation tensors in the polar decomposition $\overline{\mathbf{F}} = \overline{\mathbf{R}}\,\overline{\mathbf{U}}$. Because of the spherical (isotropic) symmetry of the reinforcement and its distribution, it is expected [10] that the average rotation tensor of the rigid phase is the identity, so that the average deformation gradient in the inclusion phase is also equal to the identity (i.e., $\overline{\mathbf{F}}^{(2)} = \mathbf{I}$). It then follows trivially that the average deformation gradient in the hyperelastic phase is given by

$$\overline{\mathbf{F}}^{(1)} = \frac{1}{1-c}(\overline{\mathbf{U}} - c\,\mathbf{I}) \tag{11}$$

Note that $\overline{\mathbf{F}}^{(1)} = \overline{\mathbf{U}}^{(1)}$, so it is convenient to define the principal stretches associated with $\overline{\mathbf{F}}^{(1)}$ via $\bar{\lambda}_i^{(1)} = (\bar{\lambda}_i - c)/(1-c)$ $(i = 1, 2, 3)$, where $\bar{\lambda}_i$ $(i = 1, 2, 3)$ are the principal stretches associated with $\overline{\mathbf{F}}$.

For the above-defined class of rigidly reinforced elastomers, the second-order estimate (10) reduces to

$$\widetilde{W}(\overline{\mathbf{U}}) = (1-c)\left[W\big(\widehat{\mathbf{F}}^{(1)}\big) - \frac{\partial W}{\partial \mathbf{F}}\big(\overline{\mathbf{F}}^{(1)}\big)\cdot\big(\widehat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}\big)\right] \tag{12}$$

where $\overline{\mathbf{F}}^{(1)}$ has already been specified in (11). It remains to determine the variables $\widehat{\mathbf{F}}^{(1)}$, as well as the modulus tensor $\mathbf{L}_0$ of the matrix phase in the linear comparison composite, which can be achieved from the relation

$$\frac{\partial W}{\partial \mathbf{F}}\big(\widehat{\mathbf{F}}^{(1)}\big) - \frac{\partial W}{\partial \mathbf{F}}\big(\overline{\mathbf{F}}^{(1)}\big) = \mathbf{L}_0\big(\widehat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}\big) \tag{13}$$

together with suitably chosen traces of the relation

$$\big(\widehat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}\big)\otimes\big(\widehat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}\big) = \mathbf{C}_{\mathbf{F}}^{(1)} = \frac{2}{1-c}\,\frac{\partial \widetilde{W}_0}{\partial \mathbf{L}_0} \tag{14}$$

In this last relation, $\mathbf{C}_{\mathbf{F}}^{(1)}$ is the covariance of the fluctuations in the matrix phase, and

$$\widetilde{W}_0(\overline{\mathbf{U}}) = (1-c)W\big(\overline{\mathbf{F}}^{(1)}\big) + \frac{1}{2}(\overline{\mathbf{U}} - \mathbf{I})\cdot\left(\tilde{\mathbf{L}}_0 - \frac{1}{1-c}\mathbf{L}_0\right)(\overline{\mathbf{U}} - \mathbf{I}) \tag{15}$$

is the effective energy of the rigidly reinforced linear comparison composite, which has been generated by making use of a suitable generalization [10] of Levin's relation [18] for two-phase *thermoelastic* composites, letting phase 2 take the energy function $W^{(2)}(\mathbf{F}) = (\mu_0^{(2)}/2)(\mathbf{F}-\mathbf{I})\cdot(\mathbf{F}-\mathbf{I})$, and taking the limit of the resulting energy expression as $\mu_0^{(2)} \to \infty$.

In expression (15), $\tilde{\mathbf{L}}_0$ denotes the effective modulus tensor of a two-phase, *linear-elastic* comparison composite consisting of a distribution of rigid inclusions with volume fraction c in a matrix with elastic modulus $\mathbf{L}_0$ and the *same* microstructure as the nonlinear elastic composite (in its undeformed configuration). This means that any

estimate for $\tilde{\mathbf{L}}_0$ can be used to generate a corresponding estimate for $\widetilde{W}$. For example, use can be made of the following Hashin–Shtrikman estimates [17] for the $\tilde{\mathbf{L}}_0$ of linear composites with *isotropic* microstructures:

$$\tilde{\mathbf{L}}_0 = \mathbf{L}_0 + \frac{c}{1-c}\mathbf{P}^{-1}, \quad \text{where } \mathbf{P} = \frac{1}{4\pi}\int\limits_{|\boldsymbol{\xi}|=1} \mathbf{H}(\boldsymbol{\xi})\,\mathrm{d}S \tag{16}$$

with $K_{ik} = L_{0ijkh}\xi_j\xi_h$, $\mathbf{N} = \mathbf{K}^{-1}$, $H_{ijkh}(\boldsymbol{\xi}) = N_{ik}\xi_j\xi_h$.

While fairly explicit, the above results require, in general, the computation of the tensor $\mathbf{P}$, which depends on the anisotropy of $\mathbf{L}_0$. In turn, the anisotropy of these tensors depends on the functional form of W and on the loading configuration, as determined by $\overline{\mathbf{F}} = \overline{\mathbf{U}}$. In addition, the derivatives of the tensor $\mathbf{P}$ with respect to $\mathbf{L}_0$ are needed in the characterization of the fluctuations $\mathbf{C}^{(1)}_{\mathbf{F}}$, which requires further computations. In this work, which presents the first application of the (improved version of the) second-order method to finite elasticity, a simple, yet illustrative example, where the computation of the $\mathbf{P}$ tensor and its derivatives is simple, will be worked out in detail. Thus, estimates of the Hashin–Shtrikman type will be derived for *plane strain* loading of a *two-dimensional* fiber-reinforced composite.

3.2. Results for in-plane loading of fiber-reinforced composites with a neo-Hookean matrix

In this section, plane strain deformations of a fiber-reinforced composite are considered where the rigid fibers, which are perpendicular to the plane of the deformation, are aligned in the x_3 direction. The distribution of the reinforcement in the plane is isotropic, so that the hypotheses that were made in the derivation of relation (12) for $\widetilde{W}$ carry over to this special case, with an appropriate (two-dimensional) modification of the relevant $\mathbf{P}$ tensor in expressions (16) for the Hashin–Shtrikman estimates for $\tilde{\mathbf{L}}_0$. The applied deformation $\overline{\mathbf{F}} = \overline{\mathbf{U}}$ in this case is entirely characterized by the two in-plane principal stretches $\bar{\lambda}_1$ and $\bar{\lambda}_2$, the out-of-plane principal stretch $\bar{\lambda}_3$ being identically 1.

Because of the transverse isotropy of the microstructure and the orthogonal symmetry of the loading condition, it is reasonable to assume that the linear comparison problem of relevance here will also exhibit orthotropic symmetry, with the symmetry axes aligned with the applied loading $\overline{\mathbf{F}} = \overline{\mathbf{U}}$. For plane strain conditions, it suffices to consider the in-plane components of a general deformation tensor $\mathbf{F}$ relative to the symmetry axes, which are not expected to be symmetric in general. The same is true of the modulus tensor $\mathbf{L}_0$, which is expected to also exhibit orthotropic symmetry, as well as major symmetry (i.e., $L_{ijkl} = L_{klij}$), but not minor symmetry.

Now, given the above assumptions, the tensor $\widehat{\mathbf{F}}^{(1)}$ is seen to have at most 4 independent components ($\widehat{F}^{(1)}_{11}$, $\widehat{F}^{(1)}_{22}$, $\widehat{F}^{(1)}_{12}$, $\widehat{F}^{(1)}_{21}$), which must be extracted from relation (14). This suggests that the tensor $\mathbf{L}_0$ should have at most 4 independent components, with respect to which $\widetilde{W}_0$ should be differentiated to generate 4 relations for the 4 components of $\widehat{\mathbf{F}}^{(1)}$ using relation (14). At the present time, it is not clear what the best choice of the components of $\mathbf{L}_0$ should be. Here, use of the prescriptions

$$L_{1212} = L_{2121} \quad \text{and} \quad L_{1221} + L_{1122} = \sqrt{(L_{1111} - L_{1212})(L_{2222} - L_{1212})} \tag{17}$$

will be made to reduce the components of the $\mathbf{L}_0$ to only 4 independent ones ($L_{1111}, L_{2222}, L_{1122}, L_{1212}$). These choices, which specify the relevant traces of relation (14) are motivated for consistency with the tangent modulus of a neo-Hookean material, and for simplicity of the resulting expressions for the tensor $\mathbf{P}$.

With these additional hypotheses, relations (14) to (15), together with Eqs. $(16)_1$ for the Hashin–Shtrikman estimate for $\tilde{\mathbf{L}}_0$, can be used to generate 4 equations for the 4 components of $\widehat{\mathbf{F}}^{(1)}$, which depend on the components of $\mathbf{L}_0$, as well as on the deformation $\overline{\mathbf{F}}$. These equations can be shown to have only two distinct solutions for $\widehat{F}^{(1)}_{11}$ and $\widehat{F}^{(1)}_{22}$, in terms of which $\widehat{F}^{(1)}_{12}$ and $\widehat{F}^{(1)}_{21}$ may be computed. Then, for each of the two essentially distinct roots for the components of $\widehat{\mathbf{F}}^{(1)}$ in terms of the 4 independent components of $\mathbf{L}_0$, two sets of 4 additional equations are generated for L_{1111}, L_{2222}, L_{1122}, and L_{1212} from the generalized secant conditions (13), which must be

solved numerically. Having computed the values of the components of $\mathbf{L}_0$ for a given particle volume fraction c, given material parameters (μ and μ'), and given loading ($\bar{\lambda}_1$ and $\bar{\lambda}_2$), the values of the components of $\widehat{\mathbf{F}}^{(1)}$ can be computed. These results are used together with the expression (11) for $\overline{\mathbf{F}}^{(1)}$ to compute the effective stored-energy function $\widetilde{W}$ for the rigidly reinforced composite using relation (12).

In this Note, results will be given only in the *incompressible* limit ($\mu' \to \infty$), where the equations simplify considerably, leading to explicit results. In this context, it is important to note that the above two distinct roots have very different asymptotic behaviors in the limit as μ' increases. The main distinguishing feature of the solutions associated with the two roots of the equations is that for one root, which is labeled the 'positive' (+) root, $\det\widehat{\mathbf{F}}^{(1)} \geqslant \det\overline{\mathbf{F}}^{(1)}$, while for the other, labeled the 'negative' (−) root, the opposite is true. For the negative-root solution, it can be shown that consideration of the incompressible limit of the energy for $\widetilde{W}$ leads to the same 'approximate' incompressibility constraint ($\det\overline{\mathbf{F}}^{(1)} = 1$) arising in the context of the earlier version [10] of the 'second-order' theory (not incorporating field fluctuations). Because of this negative feature, this solution will not be detailed further here.

On the other hand, for the positive-root solution, it can be shown that the incompressible limit of $\widetilde{W}$ is consistent with the *exact* overall incompressibility constraint ($\det\overline{\mathbf{F}} = 1$). The mathematical limit is a bit unusual in that some of the components of $\mathbf{L}$ (i.e., $L_{1111},\ L_{1122},\ L_{2222}$) become unbounded at *finite* values of μ', depending on the loading level and the particle concentration. Further details will be given elsewhere, but the final result for the effective stored-energy function of a rigidly reinforced composite with a neo-Hookean matrix may be written as follows: $\widetilde{W}^I_{HS}(\overline{\mathbf{U}}) = \widetilde{\Phi}^I_{HS}(\bar{\lambda}_1, \bar{\lambda}_2)$, where

$$\begin{aligned}\widetilde{\Phi}^I_{HS}(\bar{\lambda}_1,\bar{\lambda}_2) \;=\; & (1-c)\frac{\mu}{2}\left[\left(\bar{\lambda}_1^{(1)}\right)^2 + \left(\bar{\lambda}_2^{(1)}\right)^2 - 2\right] \\ & + \frac{\mu}{2}\frac{c}{(1-c)}\left[\frac{\bar{\lambda}_2^{(1)}}{\bar{\lambda}_1^{(1)}}(\bar{\lambda}_1-1)^2 + \frac{\bar{\lambda}_1^{(1)}}{\bar{\lambda}_2^{(1)}}(\bar{\lambda}_2-1)^2 + (\bar{\lambda}_1-\bar{\lambda}_2)^2\right]\end{aligned} \tag{18}$$

where again it is emphasized that $\bar{\lambda}_1\bar{\lambda}_2 = 1$ ($\bar{\lambda}_3 = 1$). Results for the corresponding stress-strain relation (fixing the pressure appropriately) are given in Fig. 1, for several values of the particle concentration c. Note that the behavior of the composite is quite different from that of the neo-Hookean matrix phase in that it becomes much stiffer as the applied stretch $\bar{\lambda}$ tends to $1/c$, where the composite is found to lock up. This is an interesting feature that was

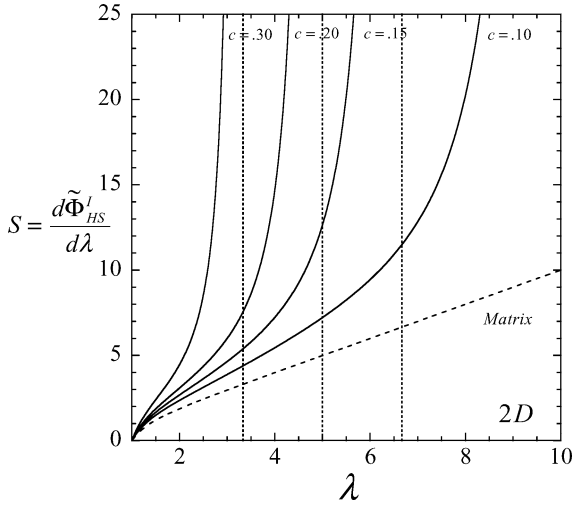

Fig. 1. Plot of the average stress $S = \mathrm{d}\widetilde{\Phi}^I_{HS}/\mathrm{d}\lambda$ versus the stretch λ for an incompressible neo-Hookean rubber reinforced with various concentrations c of aligned rigid fibers, and loaded in pure shear $\bar{\lambda}_1 = \lambda$ and $\bar{\lambda}_2 = 1/\lambda$.

already predicted by the earlier version of the theory [10] and is confirmed by the more accurate results arising from the improved theory incorporating fluctuations. However, from the theoretical point of view, the main virtue of the new second-order estimate is that – unlike the earlier version of the result [10] – it leads to a prediction that is consistent with the overall incompressibility constraint, when the matrix phase is made incompressible. Simple as this requirement may be from the physical point of view, it is a non-trivial mathematical result due to the significant nonlinearities associated with the incompressibility of the matrix phase ($\det \mathbf{F} = 1$).

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