An Exact Result for the Macroscopic Response of Particle-Reinforced Neo-Hookean Solids

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Making use of an iterated homogenization procedure in finite elasticity, an exact and explicit result is derived for the macroscopic response of Neo-Hookean solids reinforced by a random and isotropic distribution of rigid particles. The key theoretical and practical features of the result are discussed in light of comparisons with recent approximations and full-field simulations. [DOI: 10.1115/1.3197444]

1 Introduction

[1]We are concerned with the problem of predicting the macroscopic mechanical properties of hyperelastic solids reinforced by a random, statistically homogeneous distribution of rigid particles. It is assumed that the characteristic size of the particles is much smaller than the macroscopic size and the scale of variation in the applied loading. Under the above assumptions, it follows [1] that the macroscopic response of the reinforced solid can be formally characterized by the effective stored-energy function

$$\bar{W}(\bar{\mathbf{F}}, c) = (1 - c) \min_{\mathbf{F} \in \mathcal{K}(\bar{\mathbf{F}})} \frac{1}{|\Omega_m|} \int_{\Omega_m} W(\mathbf{F}) d\Omega_m \qquad (1)$$

Here, W is the stored-energy function describing the behavior of the matrix material, c is the volume faction of rigid particles in the undeformed configuration, Ω_m stands for the spatial domain occupied by the matrix material in the undeformed configuration, and $\mathcal{K}$ denotes the set of admissible deformation gradient tensors $\mathbf{F}$ with prescribed volume average $\bar{\mathbf{F}}$. The physically required *nonconvexity* of W in $\mathbf{F}$ and the assumed *randomness* of the microstructure render solving the minimization in Eq. (1) a formidable problem. Consequently—with the exception of material systems with infinite-rank laminate microstructures [2–4], which will be described in more detail further below—there are no exact results known for $\bar{W}$. Moreover, it is only relatively recent that *approximations* with a reasonable theoretical basis were put forward; see, e.g., Refs. [5,6], and references therein.

In this work, our goal is to "construct" a particle-reinforced hyperelastic solid, with a suitably designed distribution of particles, in such a manner that it is possible to compute exactly its effective stored-energy function $\bar{W}$. To this end, we will make use of an iterated dilute homogenization procedure, or differential scheme. This approach has repeatedly been proved helpful in deriving the macroscopic properties (including, for instance, electrostatic [7], viscous [8], and elastic [9] properties) of *linear* composites with a wide variety of random microstructures (see, e.g., Chapter 10.7 in the monograph by Milton [10] and references therein). By contrast, its use for *nonlinear* composites has not been pursued to nearly the same extent, presumably because of the inherent technical difficulties. Nevertheless, the central idea of

iterated dilute homogenization is geometrical in nature, and can therefore be applied to any constitutively nonlinear problem of choice (see, e.g., Ref. [11] for an application to power-law materials). In this paper, as will be described in Sec. 2, we propose an iterated dilute homogenization procedure in finite elasticity to determine the macroscopic mechanical response of hyperelastic solids reinforced by a *random* and *isotropic* distribution of rigid particles.

2 Iterated Dilute Homogenization in Finite Elasticity

We begin by considering a volume Ω_0 (in the undeformed configuration) of matrix material 0, a homogeneous, hyperelastic solid with stored-energy function W. We then embed a *dilute* distribution of rigid particles,[2] with infinitesimal volume fraction f_1, in material 0 in such a way that the total volume of the composite remains unaltered at Ω_0; that is, we remove the total volume $f_1\Omega_0$ of material 0 and replace it with rigid particles. Assuming a regular asymptotic behavior in f_1, the resulting reinforced material has an effective stored-energy function of the form

$$\bar{W}_1 = W + \mathcal{G}[W, \bar{\mathbf{F}}]f_1 + O(f_1^2) \qquad (2)$$

where $\mathcal{G}$ is a functional[3] with respect to its first argument, and a function with respect to its second argument.

Next, considering $\bar{W}_1$ as the stored-energy function of a homogeneous matrix material 1, we repeat the same process of removal and replacing while keeping the volume fixed at Ω_0. Note that this second iteration requires utilizing rigid particles that are much larger in size than those used in the first iteration, since the matrix material 1 with stored-energy function (2) is being considered as homogeneous. Specifically, we remove the total volume $f_2\Omega_0$, where f_2 is infinitesimal, of matrix material 1 and replace it with rigid particles. The composite has now an effective stored-energy function[4]

$$\bar{W}_2 = \bar{W}_1 + \mathcal{G}[\bar{W}_1, \bar{\mathbf{F}}]f_2 \qquad (3)$$

In this last expression, it is important to remark that the functional $\mathcal{G}$ is the same as in Eq. (2) because we are considering *exactly the same* dilute distribution as in Eq. (2). The more general case of different dilute distributions (corresponding, for instance, to using different particle shapes and orientations) at each iteration will be

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[1]These include the restriction that the rigid particles can only undergo rigid body rotations.

[2]The shape and orientation of the particles is arbitrary at this stage.

[3]That is, $\mathcal{G}$ is an operator (e.g., a differential operator) with respect to the stored-energy function W, so that it can depend, for instance, not just on W but also on any derivative $\partial^n W / \partial \mathbf{F}^n$, $n \in \mathbb{N}$.

[4]Here and subsequently, the order of the asymptotic correction terms will be omitted for notational simplicity.

studied in a future work. It is also worth remarking that the *total volume fraction* of rigid particles at this stage is given by $c_2 = f_2 + f_1(1-f_2) = 1 - \Pi_{j=1}^2 (1-f_j)$, and hence, that the *increment* in total volume fraction of rigid particles in this second iteration is given by $c_2 - f_1 = f_2(1-f_1)$.

It is apparent now that repeating the same above process $i+1$ times—where i is an arbitrarily large integer—generates a particle-reinforced hyperelastic solid with effective stored-energy function

$$\bar{W}_{i+1} = \bar{W}_i + \mathcal{G}[\bar{W}_i, \bar{\mathbf{F}}] f_{i+1} \tag{4}$$

which contains a total volume fraction of rigid particles given by

$$c_{i+1} = 1 - \prod_{j=1}^{i+1} (1 - f_j) \tag{5}$$

Note that for unbounded i, the right-hand side of expression (5) is, roughly speaking, the sum of infinitely many volume fractions of infinitesimal value, which can amount to the total volume fraction c_{i+1} of *finite* value. Note further that the increment in total volume fraction of rigid particles in this iteration (i.e., in passing from i to $i+1$) reads as

$$c_{i+1} - c_i = \prod_{j=1}^{i} (1 - f_j) - \prod_{j=1}^{i+1} (1 - f_j) = f_{i+1}(1 - c_i) \tag{6}$$

from which it is a trivial matter to establish the following identity:

$$f_{i+1} = \frac{c_{i+1} - c_i}{1 - c_i} \tag{7}$$

Next, substituting Eq. (7) in expression (4) renders

$$(1 - c_i) \frac{\bar{W}_{i+1} - \bar{W}_i}{c_{i+1} - c_i} - \mathcal{G}[\bar{W}_i, \bar{\mathbf{F}}] = 0 \tag{8}$$

This *difference equation* can be finally recast—upon using the facts that the increment $c_{i+1} - c_i$ is infinitesimally small, and that i is arbitrarily large—as the following *initial value problem*

$$(1 - c) \frac{\partial \bar{W}}{\partial c}(\bar{\mathbf{F}}, c) - \mathcal{G}[\bar{W}, \bar{\mathbf{F}}] = 0, \quad \bar{W}(\bar{\mathbf{F}}, 0) = W(\bar{\mathbf{F}}) \tag{9}$$

for the effective stored-energy function $\bar{W} = \bar{W}(\bar{\mathbf{F}}, c)$ of a hyperelastic solid with stored-energy function W that is reinforced by a random distribution of rigid particles with infinitely diverse sizes and finite volume fraction c.

2.1 The Auxiliary Dilute Problem. The usefulness of the exact formulation (9) hinges upon being able to compute the functional $\mathcal{G}$, describing the relevant dilute response of the composite of interest. In general, it is *not* possible to solve dilute problems in *finite elasticity* by analytical means—this is in contrast to dilute problems in linear elasticity for which there is, for instance, the explicit solution of Eshelby [12] and its generalizations. There is, however, a novel class of particulate microstructures for which the dilute response functional $\mathcal{G}$ can be computed analytically: infinite-rank laminates [2–4]. Alternatively, an analytical solution for $\mathcal{G}$ may also be approximately obtained by means of linear-comparison methods [5]; this latter approach has already proved fruitful in the related context of dilute distributions of vacuous imperfections and cavitation instabilities [13].

In this work, we will consider the dilute response functional $\mathcal{G}$ of materials with infinite-rank laminate microstructures (results based on linear-comparison methods will be reported elsewhere). In particular, as a first effort to make use of the general formulation (9), we will restrict attention to the case of two-dimensional (2D) rigidly reinforced Neo-Hookean solids with an isotropic infinite-rank laminate microstructure, for which an explicit solution was recently worked out [2] (see also Refs. [4,14] for more

general material systems). As it will become apparent in Sec. 3, this application is general enough to illustrate the essential features of our approach while permitting, at the same time, explicit mathematical treatment.

3 Application to 2D Particle-Reinforced Neo-Hookean Solids

In the sequel, we examine the case of a 2D incompressible Neo-Hookean solid with stored-energy function

$$W(\mathbf{F}) = \Phi(\lambda) = \frac{\mu}{2}(\lambda^2 + \lambda^{-2} - 2) \tag{10}$$

reinforced by an isotropic distribution of rigid particles with volume fraction c. In this last expression, the positive material parameter μ denotes the shear modulus in the ground state, and the notation $\lambda = \lambda_1$ and $\lambda^{-1} = \lambda_2$, where λ_α ($\alpha = 1, 2$) stands for the principal stretches associated with $\mathbf{F}$, has been employed to account for the incompressibility constraint $\det \mathbf{F} = \lambda_1 \lambda_2 = 1$ (i.e., $W(\mathbf{F}) = +\infty$ if $\det \mathbf{F} \neq 1$).

In view of the incompressibility of the matrix phase (10) and the rigidity of the particles, together with the assumed overall isotropy, it suffices to restrict attention to macroscopic deformation gradient tensors of the form $\bar{\mathbf{F}} = \text{diag}(\bar{\lambda}, \bar{\lambda}^{-1})$, such that $\det \bar{\mathbf{F}} = 1$. Moreover, the effective stored-energy function $\bar{W}$ of the reinforced material can be expediently expressed as

$$\bar{W}(\bar{\mathbf{F}}, c) = \bar{\Phi}(\bar{\lambda}, c) \tag{11}$$

For rigidly reinforced solids with Neo-Hookean matrix behavior of the form (10) and isotropic infinite-rank laminate microstructure, deBotton [2] obtained the following exact effective stored-energy function:

$$\bar{\Phi}_{\text{LAM}}(\bar{\lambda}, c) = \frac{1+c}{2(1-c)} \mu(\bar{\lambda}^2 + \bar{\lambda}^{-2} - 2) \tag{12}$$

which in the dilute limit of particles ($c \to 0$) reduces to

$$\bar{\Phi}_{\text{LAM}}(\bar{\lambda}, c) = \frac{\mu}{2}(\bar{\lambda}^2 + \bar{\lambda}^{-2} - 2) + c\mu(\bar{\lambda}^2 + \bar{\lambda}^{-2} - 2) + O(c^2)$$

$$= \Phi(\bar{\lambda}) + c 2\Phi(\bar{\lambda}) + O(c^2) \tag{13}$$

Here, it is important to emphasize that in the limit of small deformations ($\bar{\lambda} \to 1$), expression (13) agrees exactly with the Eshelby solution for the problem of a circular rigid particle embedded in an infinite linearly elastic (incompressible, isotropic) matrix under uniform displacement boundary conditions. In the finite-deformation regime, however, expression (13) does not correspond to the exact solution for the more general problem of a circular rigid particle embedded in an infinite Neo-Hookean matrix under uniform displacement boundary conditions (this nonlinear problem is of course much harder, and there is no available analytical solution for it). Nevertheless, for arbitrary finite deformations, expression (13) *does* constitute a very good approximation to the solution of such a problem [4]. This entails that the result (13)—which, again, is exact for certain infinite-rank laminate microstructure—can alternatively be interpreted as a very good approximation for the effective response of a Neo-Hookean solid reinforced by a dilute distribution of circular rigid particles.

From expression (13), the dilute response functional $\mathcal{G}$, as required in Eq. (9), is seen to be simply given (with a mild abuse of notation) by

$$\mathcal{G}(\bar{\Phi}, \bar{\lambda}) = 2\bar{\Phi} \tag{14}$$

Having determined Eq. (14), it is now a simple matter to deduce that the initial value problem (9) specializes in this case to

$$(1-c)\frac{\partial\bar{\Phi}}{\partial c} - 2\bar{\Phi} = 0, \quad \bar{\Phi}(\bar{\lambda},0) = \frac{\mu}{2}(\bar{\lambda}^2 + \bar{\lambda}^{-2} - 2) \quad (15)$$

This linear first-order partial differential equation admits the following closed-form solution:

$$\bar{\Phi}(\bar{\lambda},c) = \frac{\mu}{2(1-c)^2}(\bar{\lambda}^2 + \bar{\lambda}^{-2} - 2) \quad (16)$$

Expression (16) *constitutes an exact stored-energy function for 2D Neo-Hookean solids reinforced by a random and isotropic distribution of rigid particles with infinitely diverse sizes.*

The following are a few theoretical and practical remarks regarding the above-derived result.

1. In the limit of small isochoric deformations ($\bar{\mathbf{F}}\rightarrow\mathbf{I}$ with $\det\bar{\mathbf{F}}=1$), the stored-energy function (16) reduces to

$$\bar{\Phi} = \bar{\mu}(\bar{\varepsilon}_1^2 + \bar{\varepsilon}_2^2) + O(\|\bar{\mathbf{F}} - \mathbf{I}\|^3) \quad (17)$$

   where $\bar{\varepsilon}_1 = \bar{\lambda} - 1$, $\bar{\varepsilon}_2 = -\bar{\varepsilon}_1$, and

$$\bar{\mu} = \frac{1}{(1-c)^2}\mu \quad (18)$$

   is the effective shear modulus in the ground state. Expression (18) agrees exactly with the effective shear modulus of a linearly elastic (incompressible, isotropic) solid reinforced by a random and isotropic distribution of circular rigid particles of infinitely diverse sizes (see, e.g., Ref. [9]). Furthermore, in the limit of vanishingly small volume fraction of particles ($c\rightarrow0$), Eq. (18) recovers the Eshelby solution for a solid containing a dilute distribution of circular rigid particles.

2. For any finite stretch $\bar{\lambda}$, expression (16) is bounded from below by Eq. (12)—which was conjectured to be a rigorous lower bound [4]—since

$$\bar{\mu}_{\text{LAM}} = \frac{1+c}{1-c}\mu \leq \frac{1}{(1-c)^2}\mu = \bar{\mu} \quad \forall\, c \in [0,1] \quad (19)$$

   Here, it is relevant to remark that $\bar{\mu}_{\text{LAM}}$ is nothing more than the Hashin–Shtrikman lower bound for the shear modulus of 2D rigidly reinforced, incompressible, isotropic linearly elastic materials.

3. The effective stored-energy function (16) is strictly polyconvex, and therefore, strictly rank-one convex (or strongly elliptic). Thus, this exact result provides further evidence supporting ongoing studies [6,15,16], which have suggested that rigidly reinforced hyperelastic materials with random isotropic microstructures do not develop long-wavelength instabilities.

4. The functional form in $\bar{\lambda}$ of the effective stored-energy function (16) is identical to that of the underlying matrix phase (10), namely, Neo-Hookean. In this regard, however, it is important to emphasize that the functional form of the homogenized response, defined by Eq. (9), is not equal in general to that of the corresponding matrix phase.

5. By construction, the result (16) is expected to be accurate for the macroscopic response of Neo-Hookean solids reinforced by an isotropic distribution of circular particles with a very wide distribution of sizes for the *entire range* of volume fractions $c \in [0,1]$. However—as illustrated in Sec. 4—the result (16) also appears to constitute a remarkably accurate approximation for the macroscopic response of Neo-Hookean solids reinforced by an isotropic distribution of circular rigid particles with monodisperse diameters, from low to up to relatively high particle volume fractions (below the percolation limit, of course).
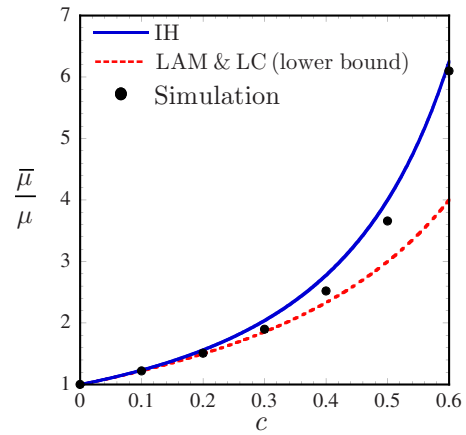
Fig. 1 The effective shear modulus $\bar{\mu}/\mu$ at zero strain of isotropic particle-reinforced Neo-Hookean solids, as a function of the volume fraction of particles *c*. Plots are shown for the IH result (18) (solid line), the LAM, and LC results as given by the left-hand side of inequality (19) (dashed line), and the boundary element simulations of Eischen and Torquato [18] (filled circles).

4 Comparisons With Estimates and Full-Field Simulations

In order to provide further insight into the new stored-energy function (16), henceforth referred to as IH, we next compare it with the infinite-rank laminate (LAM) result (13) of deBotton [2], the "linear-comparison" (LC) estimate of Lopez-Pamies and Ponte Castañeda [6]; recorded here for convenience

$$\bar{\Phi}_{\text{LC}}(\bar{\lambda},c) = \frac{\mu}{2(1-c)}\frac{(\bar{\lambda}-1)^2((\bar{\lambda}+1)^2 + 2c(\bar{\lambda}^2+1))}{\bar{\lambda}^2} \quad (20)$$

and, when available, with the finite element (FE) simulations of Moraleda et al. [17]. In this regard, it is fitting to recall, again, that the LAM result of deBotton [2] is thought to correspond to a *rigorous lower bound* for the effective stored-energy function of 2D isotropic rigidly reinforced Neo-Hookean materials [4]. The LC *estimate* of Lopez-Pamies and Ponte Castañeda, on the other hand, is appropriate for an isotropic distribution of polydisperse circular particles with small to moderate values of volume fraction *c*. Finally, the FE simulations of Moraleda et al. correspond to a (quasi)isotropic distribution of monodisperse circular particles.

We begin by examining the small-deformation regime. Figure 1 depicts plots for the normalized effective shear modulus at zero strain $\bar{\mu}/\mu$, as a function of the volume fraction of particles *c*. Results are shown for the IH, LAM, and LC responses. To aid the discussion, we have also included in the figure the boundary element simulations of Eischen and Torquato [18] for a hexagonal distribution of monodisperse circular particles—which, like the random microstructures of the IH, LAM, and LC formulations, also leads to an overall isotropic constitutive behavior for the composite in the small-deformation regime. A clear observation from Fig. 1 is that the IH result (18) is bounded from below by the LAM and LC estimates, which agree exactly with the Hashin–Shtrikman rigorous lower bound, as given by the left-hand side of inequality (19). More specifically, it is observed that the IH result and the Hashin–Shtrikman bound are essentially identical up to a concentration of particles of about $c=0.2$, after which, the IH result becomes increasingly stiffer. Although exact for a distribution of infinitely polydisperse particles, the IH response is also seen to be in good agreement with the simulations of Eischen and Torquato for a distribution of monodisperse circular particles, for the *wide range* of particle concentrations considered $0 \leq c \leq 0.6$. This favorable comparison is consistent with the notion that poly-
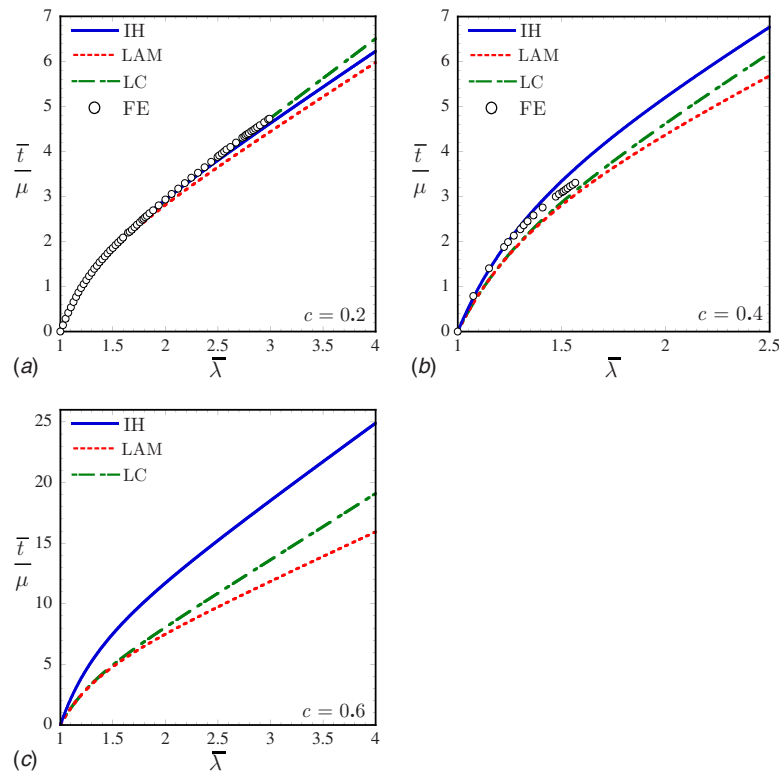
Fig. 2 Macroscopic response of isotropic particle-reinforced Neo-Hookean solids with random microstructures and various values of volume fraction of particles *c*. The macroscopic stress $\bar{t}/\mu = (1/\mu)\partial\bar{\Phi}/\partial\bar{\lambda}$ for *c*=0.2, 0.4, 0.6 ((*a*), (*b*), (*c*)), as a function of applied stretch $\bar{\lambda}$. Results are shown for the IH result (16) (solid line), the LAM result (12) of deBotton [2] (dashed line), the LC estimate (20) of Lopez-Pamies and Ponte Castañeda [6] (dashed-dotted line), and the FE simulations of Moraleda et al. [17] (circles).

dispersity might not play a major role in the response of particle-reinforced elastic materials, at least for particle volume fractions sufficiently below the percolation limit (see, e.g., Chapter 2.7 in Ref. [19]).

Figures 2(*a*)–2(*c*) show plots for the normalized macroscopic stress $\bar{t}/\mu = (1/\mu)\partial\bar{\Phi}/\partial\bar{\lambda}$ as a function of the applied stretch $\bar{\lambda}$. The results correspond, respectively, to volume fractions of particles of *c*=0.2, 0.4, and 0.6. In complete accordance with the small-deformation response illustrated in Fig. 1, Fig. 2(*a*) shows that for the case of moderate particle concentration *c*=0.2, all three results :IH, LAM, and LC, are in good agreement for the entire range of finite deformations considered. For the cases of higher particle concentrations *c*=0.4 and *c*=0.6, Figs. 2(*b*) and 2(*c*) show that the IH stress-stretch response is stiffer than the LC response, which in turn is stiffer than the LAM response. These trends are to be expected since the LAM stored-energy function (12) is a conjectured lower bound, and the LC estimate (20) was derived by solving the underlying linear-comparison problem with a Hashin–Shtrikman lower bound (see Ref. [6] for details).

The IH finite-deformation response in Figs. 2(*a*) and 2(*b*) is also seen to agree with the FE simulations for Neo-Hookean solids rigidly reinforced by monodisperse circular particles. This remarkable agreement may be attributed to the fact that—much like in the small-deformation regime—particle polydispersity has a small effect on the macroscopic response of these material systems. Indeed, in their work, Moraleda et al. [17] also carried out a series of finite element simulations for rigidly reinforced Neo-Hookean materials with a variety of distributions of polydisperse circular particles. All of their results (carried out in the range

$0 \leq c \leq 0.4$) for the effective stress-stretch response exhibited a very small dependence on the dispersion of particle sizes. At a practical level, this insensitivity to size polydispersity suggests the use of the exact result (16) to model the response of Neo-Hookean solids reinforced not just by polydisperse circular particles with any particle concentration $c \in [0,1]$—as intended by construction—but also by monodisperse circular particles with any possibly high particle concentration *c* sufficiently below the relevant percolation limit.

5 Concluding Remarks

By means of an example, we have illustrated the capabilities of iterated dilute homogenization in finite elasticity to predict the macroscopic response of particle-reinforced hyperelastic solids with random microstructures. The proposed approach leads to results that are particularly appropriate for material systems with a very wide distribution of sizes of particles. This feature happens to be well suited for most filler-reinforced elastomers of practical interest, in which the reinforcing phase (e.g., carbon black, silica) forms aggregates of very different sizes (see, e.g., the classical work of Mullins and Tobin [20]). Perhaps more significantly, the proposed approach—unlike earlier methods—leads to results that are applicable over the entire range of volume fraction of particles before percolation ensues. These general features, together with the encouraging 2D results obtained in this work, provide ample motivation to carry out further analyses for more general 3D material systems. Such analyses are in progress.

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