The nonlinear elastic response of suspensions of rigid inclusions in rubber: I—An exact result for dilute suspensions

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A solution is constructed for the problem of the overall elastic response of ideal (Gaussian or, equivalently, Neo-Hookean) rubber reinforced by a dilute isotropic distribution of rigid particles under arbitrarily large deformations. The derivation makes use of a novel iterative homogenization technique in finite elasticity that allows to construct exact solutions for the homogenization problem of two-phase nonlinear elastic composites with particulate microstructures. The solution is fully explicit for axisymmetric loading, but is otherwise given in terms of an Eikonal partial differential equation in two variables for general loading conditions. In the limit of small deformations, it reduces to the classical Einstein–Smallwood result for dilute suspensions of rigid spherical particles. The solution is further confronted to 3D finite-element simulations for the large-deformation response of a rubber block containing a single rigid spherical inclusion of infinitesimal size. The two results are found to be in good agreement for all loading conditions. We conclude this work by devising a closed-form approximation to the constructed solution which is remarkably accurate and— as elaborated in Part II— proves particularly amenable as a fundamental building block to generate approximate solutions for suspensions with finite concentration of particles.

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1. Introduction

It is well known that adding filler particles — such as carbon black and silica — to elastomers greatly improves the stiffness of this increasingly pervasive class of materials. The precise nature of such a strong stiffening remains unresolved, but a number of dominant microscopic mechanisms have been identified including the so-called “hydrodynamic” effect and the presence of bound and occluded rubber (see, e.g., Ramier, 2004; Leblanc, 2010; Qu et al., 2011). In this work we shall focus on investigating the hydrodynamic reinforcing effect, sometimes also referred to as “strain-amplification” effect, within the context of nonlinear elastic deformations. That is, we view filled elastomers as two-phase particulate composites — comprising a continuous elastomeric matrix reinforced by a statistically uniform distribution of firmly bonded inclusions — and study their macroscopic (or overall) elastic response, which, roughly speaking, is expected to be some weighted average of the elastic response of the elastomer and the comparatively rigid response of the fillers.
Following the approach of Einstein (1906) and exploiting the mathematical analogy between Stokes flow and small-strain linear elastostatics, Smallwood (1944) generated a first rigorous result for the overall linear elastic response of isotropic incompressible rubber reinforced by a dilute distribution of rigid spherical particles. Yet within the restricted setting of small-strain linear theory, significant efforts were thereafter devoted to account for non-spherical particles and non-dilute distributions (see Guth, 1945 and references therein; see also Eshelby, 1957; Batchelor and Green, 1972; Chen and Acivos, 1978; Willis, 1977).

It was not until the early 1970s that a formal framework for describing the overall nonlinear elastic response of filled elastomers undergoing finite deformations was first made available by Hill (1972). Before then, however, Mullins and Tobin (1965) had proposed an empirical approach based on the notion of “strain-amplification” factor also within the context of finite elasticity. Their idea was to describe the behavior of filled elastomers as the behavior of the underlying matrix material evaluated at an amplified measure of strain. As pursued by various authors (see, e.g., Govindjee and Simo, 1991; Govindjee, 1997; Bergström and Boyce, 1999), different results can be generated depending on the choice of strain measure that is amplified.

In spite of the fact that the framework of Hill (1972) has been available for several decades, relatively little progress has been made in its application to generate rigorous results. This is because the constitutive non-convexity and nonlinear incompressibility constraint inherent of elastomers render the relevant equations formidable complex to solve (see, e.g., Braides, 1985; Müller, 1987). Such a degree of complexity is perhaps best highlighted by the fact that (upper or lower) bounds for the response of filled elastomers are still nonexistent. In terms of analytical estimates, progress has recently been made via linear comparison methods (see Lopez-Pamies and Ponte Castañeda, 2006a and references therein). Yet while these methods have desirable features — such as the ability to incorporate information on particle concentration, shape, and spatial distribution (Lopez-Pamies and Ponte Castañeda, 2006b) — and in addition have proved fairly accurate when compared with full-field simulations (Moraleta et al., 2009; Michel et al., 2010), they are unable to rigorously recover the overall incompressibility constraint typical of filled elastomers beyond 2D problems (Lahelec et al., 2004; Lopez-Pamies, 2008). In terms of computational estimates, a variety of techniques and results have been successfully worked out in the context of small-strain linear elasticity (see, e.g., Michel et al., 1999; Segurado and Llorca, 2002; Lusti et al., 2003). However, with the exception of a few preliminary and admittedly coarse finite-element (FE) simulations (Bergström and Boyce, 1999), no 3D full-field simulations of filled elastomers undergoing finite deformations have been reported to date in the literature.

The objective of this paper is to generate a rigorous analytical result for the fundamental problem of the overall elastic response of rubber reinforced by a dilute distribution of rigid particles under arbitrarily large 3D deformations. The focus is on the basic case of ideal (Gaussian or, equivalently, Neo-Hookean) rubber and isotropic distributions of particles. This is accomplished here by making use of a novel iterated homogenization technique that allows to construct exact solutions for the homogenization problem of two-phase nonlinear elastic composites with particulate microstructures. This technique has been recently developed and utilized to generate solutions for the related fundamental problem of elastomers containing a dilute distribution of cavities — as opposed to rigid inclusions — within the analysis of cavitation instabilities (Lopez-Pamies et al., 2011a,b).

In addition to the analytical result, in this paper we also generate full 3D FE results for the large-deformation response of a block of Neo-Hookean rubber that contains a single rigid spherical inclusion of infinitesimal size located at its center. While this numerical approach for such a fundamental Eshelby-type inclusion problem seems simple enough, its presentation in the literature is not known to the authors.

We begin in Section 2 by formulating the elastostatics problem of filled rubber, with a random and isotropic distribution of rigid particles at dilute concentration, subjected to finite deformations. In Section 3, the iterated homogenization technique is presented and utilized to derive the main result of this paper: the overall nonlinear elastic response of Neo-Hookean rubber reinforced by a dilute isotropic distribution of rigid particles is characterized by the effective stored-energy function

$$W = \frac{\mu}{2} [I_1 - 3] + 2\mu H(I_1, I_2)c,$$  

(1)

to first order in the concentration of particles $c$. Here, $\mu$ is the initial shear modulus of the Neo-Hookean matrix, $I_1 = F : F$ and $I_2 = F^T F$ are the first and second principal invariants associated with the macroscopic deformation gradient $F$, and the function $H = H(I_1, I_2)$ is solution of an Eikonal partial differential equation (described in Section 3.1 and Appendix B). Section 4 presents the FE calculations. The analytical and numerical solutions constructed in Sections 3 and 4 are plotted and discussed in Section 5. We conclude in Section 6 by showing that the closed-form result

$$W = \frac{\mu}{2} [I_1 - 3] + \frac{5\mu}{4} [I_1 - 3]c$$  

(2)

is a remarkably accurate approximation of the exact result (1).

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2 The only two rigorous bounds currently available in finite elasticity, the Voigt-type upper bound of Ogden (1978) and the Reuss-type lower bound of Ponte Castañeda (1989), become unbounded (plus and minus infinity, respectively) and hence trivial when the fillers are taken to be rigid.
2. The problem

Consider a filled elastomer, made up of a continuous matrix containing a random distribution of firmly bonded rigid particles, that occupies a domain $\Omega$ with boundary $\partial \Omega$ in its undeformed stress-free configuration. The regions occupied individually by the matrix and particles are collectively denoted by $\Omega_m$ and $\Omega_p$ so that $\Omega = \Omega_m \cup \Omega_p$. It is assumed that the random distribution is statistically uniform and that the characteristic length scale of the particles (e.g., their average diameter) is much smaller than the size of $\Omega$.

Material points are identified by their initial position vector $X$ in the undeformed configuration $\Omega$, while the current position vector of the same point in the deformed configuration $\Omega^\prime$ is given by $\bf x = \bf \chi (X)$. Motivated by physical arguments, the mapping $\bf \chi$ is required to be one-to-one on $\Omega$ and twice continuously differentiable, except possibly on the particles/matrix boundaries where is only required to be continuous. The deformation gradient $\bf F$ at $\bf X$ is defined by

$$\bf F = \text{Grad} \: \bf \chi \quad \text{in} \: \Omega \tag{3}$$

and satisfies the local material impenetrability constraint $f \equiv \det \bf F > 0$.

The matrix is taken to be a nonlinear elastic solid characterized by a quasiconvex stored-energy function $W$ of $\bf F$. For convenience, the rigid particles are also described as nonlinear elastic solids with stored-energy function

$$W_p(\bf F) = \begin{cases} 0 & \text{if } \bf F \in \text{Orth}^+ \\ +\infty & \text{otherwise} \end{cases}. \tag{4}$$

Here, Orth$^+$ stands for the set of all proper orthogonal second-order tensors. At each material point $\bf X$ in the undeformed configuration, the first Piola–Kirchhoff stress $\bf S$ is formally given in terms of the deformation gradient $\bf F$ by

$$\bf S = \frac{\partial W}{\partial \bf F} (\bf X, \bf F), \quad W(\bf X, \bf F) = (1 - \theta(\bf X)) W(\bf F) + \theta(\bf X) W_p(\bf F), \tag{5}$$

where the indicator function $\theta$ takes the value 1 if the position vector $\bf X$ is in a particle, and 0 otherwise, and serves therefore to describe the microstructure (here, the size, shape, and spatial location of the particles) in the undeformed configuration $\Omega$.

Granted the hypotheses of separation of length scales and statistical uniformity of the microstructure together with the constituent quasiconvexity of $W$, the overall or macroscopic response of the filled elastomer can be defined as the relation between the volume averages of the first Piola–Kirchhoff stress $\bf S$ and the deformation gradient $\bf F$ over $\Omega$ when the material is subjected to the affine boundary condition

$$\bf x = \bf F \bf X \quad \text{on} \: \partial \Omega, \tag{6}$$

where the second-order tensor $\bf F$ is a prescribed quantity (Hill, 1972). In this case, it directly follows from the divergence theorem that the average deformation gradient over $\Omega$ is given by $|\Omega|^{-1} \int_\Omega \bf F \bf x \, d\bf x = \bf F$, and hence the derivation of the macroscopic response reduces to finding the average stress $\bf S = |\Omega|^{-1} \int_\Omega \bf S \bf x \, d\bf x$ for a given $\bf F$. The result reads formally as

$$\bf S = \frac{\partial W}{\partial \bf F} (\bf F, c), \tag{7}$$

with

$$W(\bf F, c) = (1 - c) \min_{\bf F \in \kappa} \frac{1}{|\Omega_m|} \int_{\Omega_m} W(\bf F) \, d\bf x. \tag{8}$$

In this last expression, $c = |\Omega|^{-1} \int_\Omega \theta(\bf X) \, d\bf x$ is the initial volume fraction or concentration of particles, $W$ is the so-called effective stored-energy function, which physically corresponds to the total elastic energy (per unit undeformed volume) stored in the material, and $\kappa$ denotes the set of kinematically admissible deformation gradient fields

$$\kappa = \{ \bf F : \exists \bf x = \bf \chi (\bf X) \quad \text{with} \quad \bf F = \text{Grad} \: \bf \chi, J > 0 \ \text{in} \: \Omega, \bf F = \bf Q \in \text{Orth}^+ \ \text{in} \: \Omega_p, \bf x = \bf F \bf X \ \text{on} \: \partial \Omega \}. \tag{9}$$

The foregoing formulation for the overall finite-deformation response of filled elastomers is valid for any physically admissible value of concentration of particles $c$. The interest here is in the asymptotic limit as $c \to 0^+$, when the above-defined material reduces to a nonlinear elastic solid with stored-energy function $W$ that contains a random distribution of rigid particles, with shapes and spatial locations characterized by $\theta$, at dilute concentration. Assuming a polynomial asymptotic behavior, the effective stored-energy function (8) in this limiting case takes the form

$$W(\bf F, c) = W(\bf F) + \mathcal{G}(W, F) c + O(c^2), \tag{10}$$

where $\mathcal{G}$ is a functional with respect to its first argument $W$ and a function with respect to its second argument $\bf F$. 

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3 For the problem of filled Neo-Hookean rubber considered in this work the asymptotic behavior is indeed of the polynomial form (10).
4 That is, $\mathcal{G}$ is an operator with respect to the stored-energy function $W$ of the elastomeric matrix.
2.1. The case of dilute isotropic suspensions in Neo-Hookean rubber

The main objective of this work is to determine the functional $\mathcal{G}$ in (10) for the basic case when the distribution of particles is isotropic and the elastomeric matrix is Neo-Hookean rubber with stored-energy function

$$W(F) = \begin{cases} \frac{\mu}{2} \left[ I_1 - 3 \right] - \frac{\mu}{2} \left[ \lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3 \right] & \text{if } J = \lambda_1 \lambda_2 \lambda_3 = 1 \\ +\infty & \text{otherwise} \end{cases} \tag{11}$$

Here, it is recalled that the parameter $\mu$ denotes the initial shear modulus of the Neo-Hookean matrix, $I_1 = F \cdot F$, and $\lambda_1, \lambda_2, \lambda_3$ have been introduced to denote the singular values of the deformation gradient $F$.

Owing to the assumed isotropy of the microstructure and the constitutive isotropy and incompressibility of the matrix material $(11)$ and rigid particles $(4)$, the resulting overall elastic response is isotropic and incompressible. This implies that the effective stored-energy function $\overline{W}$ in this case depends on the macroscopic deformation gradient $F$ only through its singular values $\overline{\lambda}_1, \overline{\lambda}_2, \overline{\lambda}_3$ and becomes unbounded for non-isochoric deformations when $J \equiv \det F = \overline{\lambda}_1 \overline{\lambda}_2 \overline{\lambda}_3 \neq 1$. More explicitly, the result $(10)$ specializes to

$$\overline{W} = \begin{cases} \frac{\mu}{2} \left[ \overline{\lambda}_1^2 + \overline{\lambda}_2^2 + \overline{\lambda}_3^2 - 3 \right] + \mu G(\overline{\lambda}_1, \overline{\lambda}_2, \overline{\lambda}_3) c + O(c^2) & \text{if } J = \overline{\lambda}_1 \overline{\lambda}_2 \overline{\lambda}_3 = 1 \\ +\infty & \text{otherwise} \end{cases} \tag{12}$$

where $G(\overline{\lambda}_1, \overline{\lambda}_2, \overline{\lambda}_3)$ is a symmetric function.

In order to assist the presentation of the results, the unbounded branch of the energy (12) is omitted in most of the sequel. For this purpose and without loss of generality we restrict attention to isochoric pure stretch loadings of the form

$$F = \text{diag}(\overline{\lambda}_1, \overline{\lambda}_2, \overline{\lambda}_3) \quad \text{with} \quad \overline{\lambda}_3 = \frac{1}{\overline{\lambda}_1 \overline{\lambda}_2}, \tag{13}$$

and, with a slight abuse of notation, rewrite the effective stored-energy function (12) of the filled Neo-Hookean rubber as

$$\overline{W} = \frac{\mu}{2} \left[ \overline{\lambda}_1^2 + \overline{\lambda}_2^2 + \frac{1}{\overline{\lambda}_1 \overline{\lambda}_2} - 3 \right] + \mu G(\overline{\lambda}_1, \overline{\lambda}_2) c, \tag{14}$$

to first order in the concentration of particles $c$.

3. An exact solution via a novel iterated homogenization method

In this section, we derive a precise form of the effective stored-energy function (14) for isotropic dilute suspensions of rigid particles in Neo-Hookean rubber. This amounts to solving asymptotically the relevant minimization problem $(8)$ with $(11)$ in the limit as $c \to 0^+$. Our strategy involves two main steps. In the first step (Section 3.1), we make use of the new iterated homogenization technique of Lopez-Pamies et al. (2011a) to work out an exact result for the overall response of a Neo-Hookean solid containing a particular class of isotropic distributions of rigid particles with finite concentration $c$. The second step (Section 3.2) deals with the asymptotic analysis of this result in the limit as the concentration of the particles is taken to zero.

3.1. Iterated homogenization solution for finite concentration of particles

By means of a combination of iterative processes (Idiart, 2008; Lopez-Pamies, 2010), Lopez-Pamies et al. (2011a) have recently generated an exact solution for the effective stored-energy function of a two-phase composite made up of a nonlinear elastic matrix containing a specific — but fairly general — class of distributions (i.e., a specific class of indicator functions $\ell$) of nonlinear elastic particles. For the special case of isotropic distributions of interest in this work, their result for $\overline{W}(F, c)$ in the present notation is implicitly given by the following first-order nonlinear partial differential equation (pde)

$$c \frac{\partial \overline{W}}{\partial c} - \frac{1}{4\pi} \int_{\xi = 1} \max_{\omega} \left[ \omega \cdot \frac{\partial \overline{W}}{\partial F} \xi - \overline{W}(F + \omega \otimes \xi) \right] d\xi = 0 \tag{15}$$

subject to the initial condition

$$\overline{W}(F, 1) = W_0(F). \tag{16}$$

The interested reader is referred to Section 3 of Lopez-Pamies et al. (2011a) for the derivation and thorough discussion of the above result, but at this stage it is appropriate to record a few of its properties:

- **Constitutive behavior and concentration of the matrix and particles.** The result $(15)$–$(16)$ is valid for any choice of (including compressible and anisotropic) stored-energy functions $W$ and $W_0$ for the elastomeric matrix and particles, provided that these satisfy usual physically based mathematical requirements. The result also holds applicable for any value of concentration of particles in the physical range $c \in [0,1]$. 
• **Interaction among particles.** By construction, the underlying microstructure associated with the stored-energy function (15)–(16) corresponds to an isotropic distribution of disconnected particles of polydisperse sizes that interact in such a manner that they deform *uniformly*, irrespective of the applied macroscopic deformation \( \mathbf{F} \) or the value of particle concentration \( c \). Such a special type of deformation is usually associated with the softest possible response of stiff materials. Thus, (15)–(16) is generally expected to bound from below the effective stored-energy functions of nonlinear elastic solids reinforced by any type of isotropic distribution of particles (whether (15)–(16) is a rigorous lower bound remains yet to be proved or disproved).

• **Connection with the classical result of Eshelby.** A direct implication of the fact that the particles deform uniformly is that in the limit of small deformations and small particle concentration as \( \mathbf{F} \to \mathbf{I} \) and \( c \to 0^+ \), expressions (15)–(16) recover identically the classical result for Eshelby for the overall response of a dilute distribution of linearly elastic spherical particles embedded in a linearly elastic matrix. The formulation (15)–(16) can thus be thought of as a direct extension of the classical result of Eshelby to deal with finite deformations. Further comments on this key aspect are deferred to the end of this section and to Section 5.

Now, for a Neo-Hookean matrix characterized by (11) it is not difficult to show that the maximizing vector \( \mathbf{\omega} \) in (15) specializes to

\[
\mathbf{\omega} = \frac{1}{\mu} \frac{\partial W}{\partial \mathbf{F}} \mathbf{\xi} - \mathbf{F}^{-T} \mathbf{\xi} + \frac{1}{J} \mathbf{F}^{-T} \mathbf{F} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi} - \frac{\partial W}{\partial \mathbf{F}} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi} - \frac{\partial W}{\partial \mathbf{F}} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi},
\]

and hence that the effective stored-energy function in this case can be conveniently written as

\[
W(\mathbf{F}, c) = 2\mu \mathcal{U}(\mathbf{F}, c) + \frac{\mu}{2} [\mathbf{F} \cdot \mathbf{F} - 3],
\]

where the function \( \mathcal{U} \) is solution of the initial-value problem

\[
\frac{\partial \mathcal{U}}{\partial t} \mathcal{U} - \int_{|\xi| = 1} \frac{1}{4\pi} \left[ \frac{\partial \mathcal{U}}{\partial \mathbf{F}} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi} + \left( \frac{\partial \mathcal{U}}{\partial \mathbf{F}} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi} \right)^2 \right] d\xi = 0,
\]

\[
\mathcal{U}(\mathbf{F}, 1) = \frac{1}{2\mu} W_p(\mathbf{F}) - \frac{1}{4} [\mathbf{F} \cdot \mathbf{F} - 3],
\]

and where it is reemphasized that (19) holds applicable for any choice of \( W_p \).

In order to account for the perfectly rigid behavior (4) within the context of the formulation (15)–(16), it proves expedient not to work with (4) directly but to consider instead the regularized and hence more general case of *elastic isotropic incompressible* particles with stored-energy function

\[
W_p(\mathbf{F}) = \begin{cases} \frac{4\mu_p - \mu_0}{5} H(\mathbf{F}) + \frac{\mu}{2} [\mathbf{F} \cdot \mathbf{F} - 3] & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases}
\]

In this last expression, the parameter \( \mu_p \) denotes the shear modulus of the particles in their undeformed state and \( H \) is an objective and isotropic function of \( \mathbf{F} \), satisfying the conditions \( H(\mathbf{Q}) = 0 \) for all \( \mathbf{Q} \in \text{Orth}^+ \) and \( H(\mathbf{F}) > 0 \) for \( \mathbf{F} \not\in \text{Orth}^+ \), to be specified subsequently. The perfectly rigid behavior (4) can then be readily recovered as a special case of (20) by taking the limit of \( \mu_p \to +\infty \).

Given the incompressible stored-energy function (20) for the particles, it follows that the effective stored-energy function (18) for the filled Neo-Hookean rubber reduces to

\[
W(\mathbf{F}, c) = \begin{cases} 2\mu \mathcal{U}(\mathbf{F}, c) + \frac{\mu}{2} [\mathbf{F} \cdot \mathbf{F} - 3] & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases},
\]

with \( \mathcal{U} \) now being defined by equations

\[
\frac{\partial \mathcal{U}}{\partial t} \mathcal{U} - \int_{|\xi| = 1} \frac{1}{4\pi} \left[ \frac{\partial \mathcal{U}}{\partial \mathbf{F}} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi} + \left( \frac{\partial \mathcal{U}}{\partial \mathbf{F}} \mathbf{\xi} \cdot \mathbf{F}^{-T} \mathbf{\xi} \right)^2 \right] d\xi = 0, \quad \mathcal{U}(\mathbf{F}, 1) = \frac{2(\mu_p - \mu)}{5\mu} H(\mathbf{F})
\]

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5 That is, the deformation gradient field \( \mathbf{F}(\mathbf{X}) \) — and hence the stress field \( \mathbf{S}(\mathbf{X}) \) — within each particle is uniform and the same for all particles.
subject to the constraint \( J = 1 \). To make further progress, it is helpful to exploit the overall isotropy and incompressibility of the problem. Thus, after restricting attention to isochoric pure stretch loadings of the form (13), carrying out the required integrals in (22), and with a little abuse of notation, \(^6\) the (finite branch of the) effective stored-energy function (21) for the filled Neo-Hookean rubber can be compactly rewritten as

$$
\mathcal{W}(\lambda_1, \lambda_2, c) = 2\mu \mathcal{U}(\lambda_1, \lambda_2, c) + \frac{\mu}{2} \left[ \lambda_1^2 + \lambda_2^2 + \frac{1}{\lambda_1^2 \lambda_2^2} - 3 \right],
$$

(23)

where the function \( \mathcal{U} \) is implicitly defined by the pde

$$
c \frac{\partial \mathcal{U}}{\partial c} - \mathcal{U} + \chi_1(\lambda_1, \lambda_2) \left( \frac{\partial \mathcal{U}}{\partial \lambda_1} \right)^2 + \chi_2(\lambda_1, \lambda_2) \left( \frac{\partial \mathcal{U}}{\partial \lambda_2} \right)^2 + \chi_3(\lambda_1, \lambda_2) \frac{\partial \mathcal{U}}{\partial \lambda_1} \frac{\partial \mathcal{U}}{\partial \lambda_2} = 0
$$

(24)

subject to the initial condition

$$
\mathcal{U}(\lambda_1, \lambda_2, 1) = \frac{2(\mu_p - \mu)}{5\mu} H(\lambda_1, \lambda_2).
$$

(25)

The coefficients \( \chi_1, \chi_2, \chi_3 \) above are functions of their arguments given in explicit form by expressions (56) in Appendix A.

In view of the definition (20), it is also appropriate to record here for later reference that the function \( H : (a, b) \in \mathbb{R}^2 : a, b > 0 \to \mathbb{R} \) in (25) must satisfy the following properties:

$$
H(1, 1) = 0,
H(\lambda_1, \lambda_2) > 0 \quad \forall \lambda_1, \lambda_2 \neq 1,
H(\lambda_1, \lambda_2) = H(H(\lambda_1, \lambda_2)^{-1}, \lambda_2) = H(H^{-1}(\lambda_1, \lambda_2), \lambda_2) = H(H^{-1}(\lambda_1, \lambda_2)^{-1}, \lambda_2) \quad \forall \lambda_1, \lambda_2.
$$

(26)

The final step in this first part of the derivation is to solve the initial-value problem (24)–(25) for the function \( \mathcal{U} \) and then take the limit of rigid particles \( \mu_p \to +\infty \). To this end, it is gainful to recognize two key aspects of Eqs. (24)–(25). First, the particular form of the function \( H \) in (25) is immaterial, provided that the choice satisfies conditions (26). Second, as a result of the overall isotropy of the problem, the function \( \mathcal{U} \) defined by (24)–(25) is symmetric with respect to its first two arguments, namely, \( \mathcal{U}(\lambda_1, \lambda_2, c) = \mathcal{U}(\lambda_2, \lambda_1, c) \). In the sequel, we exploit the flexibility in the choice of \( H \) and the symmetry of \( \mathcal{U} \) in order to construct a solution of (24)–(25). The idea is to utilize a particular choice of the function \( H \) that simplifies the calculations involved. And to make use of the symmetry of \( \mathcal{U} \) in order to rewrite the initial condition (25) in terms of the more convenient deformation variables \( \bar{\lambda}_1, \bar{\lambda}_2 \), instead of in terms of the concentration of particles \( c \). As elaborated next, the proposed strategy requires the successive analyses of axisymmetric \( (\bar{\lambda}_1 = \bar{\lambda}_2 = \bar{\lambda}) \) and general loading conditions.

(i) Axisymmetric loading conditions. We begin by analyzing the special case of axisymmetric loading with \( \bar{\lambda}_2 = \bar{\lambda}_1 = \bar{\lambda} \).

(27)

By introducing the notation

$$
\mathcal{U}_A(\bar{\lambda}, c) = \mathcal{U}(\bar{\lambda}, \bar{\lambda}, c),
$$

(28)

and recognizing the identities

$$
\frac{\partial \mathcal{U}}{\partial \lambda_1}(\bar{\lambda}, \bar{\lambda}, c) = \frac{\partial \mathcal{U}}{\partial \lambda_2}(\bar{\lambda}, \bar{\lambda}, c) = \frac{1}{2} \frac{\partial \mathcal{U}_A}{\partial \bar{\lambda}}(\bar{\lambda}, c),
$$

(29)

due to the symmetry of \( \mathcal{U} \), it is straightforward to show (see relations (60) and (61) in Appendix A) that the initial-value problem (24)–(25) reduces to the simpler initial-value problem

$$
c \frac{\partial \mathcal{U}_A}{\partial \bar{\lambda}} - \mathcal{U}_A = \frac{\bar{\lambda}^6}{12(\bar{\lambda} - 1)^2} \left[ 1 + 2\bar{\lambda}^6 - \frac{3\bar{\lambda}^6}{1 - \bar{\lambda}^6} \ln \left[ \frac{\sqrt{1 - \bar{\lambda}^6} + 1}{\bar{\lambda}^4} \right] \right] \left( \frac{\partial \mathcal{U}_A}{\partial \bar{\lambda}} \right)^2 = 0,
$$

(30)

with

$$
\mathcal{U}_A(\bar{\lambda}, 1) = \frac{2(\mu_p - \mu)}{5\mu} H(\bar{\lambda}, \bar{\lambda}),
$$

(31)

for the function \( \mathcal{U}_A \). In spite of the fact that the pde (30) is nonlinear, the suitable choice

$$
H(\bar{\lambda}, \bar{\lambda}) = 3 \int_1^{\bar{\lambda}} \frac{\sqrt{(z^6 - 1)^2}}{z^3 \sqrt{1 + 2z^6 - \frac{3z^6}{1 - z^6} \ln \left[ \frac{1 + \sqrt{1 - z^6}}{z^4} \right]}} \, dz
$$

(32)

\(^6\) The fact that the same symbols \( \mathcal{W} \), \( \mathcal{U} \), and \( H \) are utilized to denote the corresponding functions in terms of the stretches \( \lambda_1 \) and \( \lambda_2 \) should not lead to confusion.
makes it possible to solve (30)–(31) in closed form. The result reads as follows:

\[ U_H(z, c) = 3 \frac{2c(\mu_p - \mu)}{2(1-c)c_p + (3+2c)\mu} \left[ \int_1^z \frac{\sqrt{(z^6 - 1)^2}}{z^3 \sqrt{1 + 2z^6 - \frac{3z^6}{\sqrt{1 + \frac{1}{z^2}}}}} \, dz \right]^2. \]  

Here, we remark that the choice (32) for \( H \) is such that \( H(1,1) = 0 \) and \( H(z, z) > 0 \) if \( z \neq 1 \), as required by conditions (26).

Given the quadratic nonlinearity of the pde (30), it is also worth mentioning that Eqs. (30)–(31) with (32) have two solutions, but that both are identical and given by (33) in this case.

(II) General loading conditions. Having determined the axisymmetric solution (33) for any value of particle concentration \( c \in [0,1] \), the initial-value problem (24)–(25) can now be rewritten as

\[ \frac{\partial U}{\partial c} - U + x_1(\lambda_1, \lambda_2) \left( \frac{\partial U}{\partial \lambda_1} \right)^2 + x_2(\lambda_1, \lambda_2) \left( \frac{\partial U}{\partial \lambda_2} \right)^2 + x_3(\lambda_1, \lambda_2) \frac{\partial U}{\partial \lambda_1} \frac{\partial U}{\partial \lambda_2} = 0 \]  

subject to the alternative deformation-based initial condition

\[ U(z_1, z_2, c) = 3 \frac{2c(\mu_p - \mu)}{2(1-c)c_p + (3+2c)\mu} \left[ \int_1^z \frac{\sqrt{(z^6 - 1)^2}}{z^3 \sqrt{1 + 2z^6 - \frac{3z^6}{\sqrt{1 + \frac{1}{z^2}}}}} \, dz \right]^2 \]  

as opposed to the original concentration-based condition (25), where it is recalled that the coefficients \( x_1, x_2, x_3 \) in (34) are given in explicit form by expressions (56) in Appendix A.

In view of the separable structure of the alternative initial condition (35), it is not difficult to deduce from the governing pde (34) that the solution for \( U \) is given by

\[ U(\lambda_1, \lambda_2, c) = \frac{2c(\mu_p - \mu)}{2(1-c)c_p + (3+2c)\mu} H(\lambda_1, \lambda_2), \]  

where the function \( H \) is implicitly defined by the initial-value problem

\[ H + x_1(\lambda_1, \lambda_2) \left( \frac{\partial H}{\partial \lambda_1} \right)^2 + x_2(\lambda_1, \lambda_2) \left( \frac{\partial H}{\partial \lambda_2} \right)^2 + x_3(\lambda_1, \lambda_2) \frac{\partial H}{\partial \lambda_1} \frac{\partial H}{\partial \lambda_2} = 0, \]  

with

\[ H(\lambda_1, \lambda_2) = 3 \left[ \int_1^\lambda_1 \frac{\sqrt{(z^6 - 1)^2}}{z^3 \sqrt{1 + 2z^6 - \frac{3z^6}{\sqrt{1 + \frac{1}{z^2}}}}} \, dz \right]^2. \]  

Rather interestingly, the first-order nonlinear pde (37) subject to (38) is an Eikonal equation.\(^7\) This class of equations has appeared pervasively in a wide variety of problems concerning geometrical optics and other wave-propagation phenomena (see, e.g., Born et al., 2003). Unfortunately, the specific type of Eikonal equation (37)–(38) does not appear to be solvable in closed form, but it can be solved numerically by available techniques. Fig. 1 shows plots of such a solution over a large range of stretches \( \lambda_1 \) and \( \lambda_2 \).

In addition to the required properties (26), two further features of the function \( H \) defined by (37)–(38) worth recording for later use are that in the limit of small deformation as \( \varepsilon_1 = \lambda_1 - 1 \rightarrow 0 \) and \( \varepsilon_2 = \lambda_2 - 1 \rightarrow 0 \) it takes the polynomial asymptotic form

\[ H(\lambda_1, \lambda_2) = \frac{5}{4} \left[ \lambda_1^2 + \lambda_2^2 + (\varepsilon_1 + \varepsilon_2)^2 \right] - \frac{5}{2} \left( \varepsilon_1^2 + \varepsilon_2^2 \right) - \frac{55}{14} \left( \varepsilon_1^2 \varepsilon_2 + \varepsilon_1 \varepsilon_2^2 \right) + O(\varepsilon_1^4) + O(\varepsilon_2^4), \]  

while in the opposite limit of infinitely large deformations as \( \lambda_1 \rightarrow 0, +\infty \) and/or \( \lambda_2 \rightarrow 0, +\infty \) it reduces asymptotically to

\[ H(\lambda_1, \lambda_2) = \frac{3}{4} \left[ \lambda_1^2 + \lambda_2^2 + \frac{1}{\lambda_1 \lambda_2} \right]. \]  

to leading order, hence becoming unbounded. The proof of relations (26), (39), and (40) together with relevant comments on the numerical computation of \( H \) are given in Appendix B.

At this stage, it is a trivial matter to take the limit of rigid particles \( \mu_p \rightarrow +\infty \) in expression (36) to conclude that the effective stored-energy function for Neo-Hookean rubber reinforced by the class of isotropic distributions of rigid particles

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\(^7\) With the change of variables \( H = \tilde{H}^2 \) and the notation \( x_1 = -(M_{11}^2 + M_{12}^2)/4, x_2 = -(M_{12}^2 + M_{22}^2)/4, x_3 = -M_{12}(M_{11} + M_{22})/2 \), the Eikonal equation (37) takes the more standard invariant form \( |\mathbf{M}V\tilde{H}| = 1 \).
effective stored-energy function specified by the formulation (15)-(16) is given by
\[
W(\overline{\lambda}_1, \overline{\lambda}_2, c) = \frac{2\mu c}{1-c} H(\overline{\lambda}_1, \overline{\lambda}_2) + \frac{\mu}{2} \left[ \frac{\overline{\lambda}_1^2}{\overline{\lambda}_1^2 - 1} + \frac{1}{\overline{\lambda}_1^2} - 3 \right],
\]
with \( H \) being defined by Eqs. (37)-(38). Again, this result is valid\(^6\) for any value of concentration of particles in the range \( c \in [0,1] \). The analysis of its asymptotic behavior in the limit as \( c \to 0^+ \) is the final step of the derivation and the subject of the next subsection.

3.2. Asymptotic solution in the dilute limit of particles as \( c \to 0^+ \)

We are now in a position to readily take the limit \( c \to 0^+ \) in the result (41) to finally establish that the overall elastic response of Neo-Hookean rubber reinforced by a dilute isotropic distribution of rigid particles is characterized by the effective stored-energy function
\[
W(\overline{\lambda}_1, \overline{\lambda}_2, c) = \frac{\mu}{2} \left[ 2\frac{\overline{\lambda}_1^2}{\overline{\lambda}_1^2 - 1} + \frac{1}{\overline{\lambda}_1^2} - 3 \right] + 2\mu H(\overline{\lambda}_1, \overline{\lambda}_2) c,
\]
to first order in the concentration of particles \( c \). As derived in the foregoing development, the function \( H \) is implicitly defined by the Eikonal pde (37) subject to the initial condition (38). In general, again, these equations must be solved numerically. For the special case of axisymmetric loading, however, they admit a closed-form solution and expression (42) reduces to
\[
W(\overline{\lambda}, c) = \frac{\mu}{2} \left[ 2\frac{\overline{\lambda}^2}{\overline{\lambda}^2 - 1} + \frac{1}{\overline{\lambda}^2} - 3 \right] + 6\mu \left[ \int \frac{\overline{\lambda}}{z^3} \sqrt{1 + 2\epsilon^2 - \frac{3\epsilon^2}{\overline{\lambda}^2} \ln \left( 1 + \frac{\overline{\lambda}^2 - 1}{\overline{\lambda}^2} \right)} \, dz \right]^2 c.
\]

Comparing (14) with (42), it is seen that the asymptotic form of the solution is indeed polynomial and that \( G(\overline{\lambda}_1, \overline{\lambda}_2) = 2H(\overline{\lambda}_1, \overline{\lambda}_2) \). The following remarks are in order:

i. Owing to the properties (26) of the function \( H \), the effective stored-energy function (42) is such that
\[
W(1,1,c) = 0, \quad W(\overline{\lambda}_1, \overline{\lambda}_2, c) > 0 \quad \forall \overline{\lambda}_1, \overline{\lambda}_2 \neq 1, \quad W(\overline{\lambda}_1, \overline{\lambda}_2, c) = W(\overline{\lambda}_2, \overline{\lambda}_1, c) = W((\overline{\lambda}_1, \overline{\lambda}_2)^{-1}, c) = W((\overline{\lambda}_1, \overline{\lambda}_2)^{-1}, \overline{\lambda}_1, c) = W(\overline{\lambda}_2, (\overline{\lambda}_1, \overline{\lambda}_2)^{-1}, c) = W((\overline{\lambda}_1, \overline{\lambda}_2)^{-1}, \overline{\lambda}_2, c) \quad \forall \overline{\lambda}_1, \overline{\lambda}_2, \quad W(\overline{\lambda}_1, \overline{\lambda}_2, c) > W(\overline{\lambda}_1, \overline{\lambda}_2, 0) \quad \forall \overline{\lambda}_1, \overline{\lambda}_2 \neq 1, \quad c > 0.
\]
The first three of these conditions are direct consequences of the fact that the filled Neo-Hookean rubber is stress-free in the undeformed configuration, isotropic, and incompressible. The last condition entails physically that the addition of rigid particles consistently leads to a stiffer material response irrespectively of the applied loading, in agreement with experience.

ii. In the limit of small deformations as \( \tau_1 = \bar{\tau}_1 \to 0 \) and \( \tau_2 = \bar{\tau}_2 \to 0 \), based on the asymptotic behavior (39) of \( H \), the effective stored-energy function (42) takes the explicit asymptotic form

\[
\tilde{W}(\tilde{\tau}_1, \tilde{\tau}_2, c) = \mu \left( \tau_1^2 + \tau_2^2 - 2(\tau_1^2 + \tau_2^2) - 3(\tau_1^2 \tau_2 + \tau_1 \tau_2^2) \right) + \frac{5}{2} \mu \left[ \tau_1^2 + \tau_2^2 + (\tau_1 + \tau_2)^2 - 2(\tau_1^2 + \tau_2^2) - \frac{110}{35} (\tau_1^2 \tau_2 + \tau_1 \tau_2^2) \right] c,
\]

(45)
to order three in the deformation measures \( \tau_1 \) and \( \tau_2 \). As anticipated in the description of the formulation (15)–(16), the leading order of expression (45) agrees identically with the Einstein–Smallwood (or, more generally, Eshelby) result for the overall elastic response of a dilute distribution of rigid spherical particles embedded in an isotropic incompressible linearly elastic matrix (cf. Eq. (12) in Smallwood, 1944).

iii. In terms of the principal invariants \( \bar{T}_1 = \bar{F} : \bar{F} = \bar{\tau}_1^2 + \bar{\tau}_2^2 + \bar{\tau}_1^{-2} \bar{\tau}_2^{-2} \) and \( \bar{T}_2 = \bar{F}^{-1} : \bar{F}^{-1} = \bar{\tau}_1^{-2} + \bar{\tau}_2^{-2} + \bar{\tau}_1 \bar{\tau}_2 \), the asymptotic result (45) can be rewritten as

\[
\tilde{W}(\bar{\tau}_1, \bar{\tau}_2, c) = \frac{\mu}{2} (\bar{T}_1 - 3) + 2\mu \left[ \frac{145}{224} (\bar{T}_1 - 3) - \frac{5}{224} (\bar{T}_2 - 3) \right] c,
\]

(46)
to order one in the deformation measures \( (\bar{T}_1 - 3) \) and \( (\bar{T}_2 - 3) \). This expression illustrates explicitly that the overall response of the filled Neo-Hookean rubber depends not only on the first invariant \( T_1 \) but also on the second invariant \( T_2 \) (in spite of the fact that the underlying Neo-Hookean matrix depends only on the first invariant). Given that the associated coefficient \( 5/224 \) is significantly smaller than unity, however, the dependence on \( T_2 \) is weak. Rather remarkably, as discussed below and in Section 5, the dependence on \( T_2 \) remains weak for large deformations (of order \( (T_2 - 3)^2 \) and higher) and it completely disappears in the limit of deformations that are infinitely large.

iv. In the limit when the deformation becomes unbounded as \( \bar{\tau}_1 \to 0, + \infty \) and/or \( \bar{\tau}_2 \to 0, + \infty \), the function \( H \) reduces to (40) and hence it is straightforward to deduce that the effective stored-energy function (42) reduces in turn to the explicit form

\[
\tilde{W}(\bar{\tau}_1, \bar{\tau}_2, c) = \frac{\mu}{2} \left[ \bar{\tau}_1^2 + \bar{\tau}_2^2 + \frac{1}{\bar{\tau}_1 \bar{\tau}_2} \right] + \frac{6\mu}{4} \left[ \bar{\tau}_1^2 + \bar{\tau}_2^2 + \frac{1}{\bar{\tau}_1 \bar{\tau}_2} \right] c,
\]

(47)
or, equivalently,

\[
\tilde{W}(\bar{\tau}_1, \bar{\tau}_2, c) = \frac{\mu}{2} \bar{T}_1 + \frac{6\mu}{4} \bar{T}_1 c.
\]

(48)
to leading order. That is, for large enough deformations, the overall energy of the filled Neo-Hookean rubber grows linearly in the first invariant \( \bar{T}_1 \) and independently of \( \bar{T}_2 \).

v. Consistent with recent bifurcation analyses (Triantafyllidis et al., 2007; Michel et al., 2010), the effective stored-energy function (42) is strongly elliptic. In the present context of isotropic incompressible elasticity, it is possible to write down explicit necessary and sufficient conditions for strong ellipticity in the form of nine scalar inequalities involving first and second derivatives of (42) with respect to \( \bar{T}_1 \) and \( \bar{T}_2 \), as detailed in Appendix C. While difficult by analytical means, it is a simple matter to verify numerically that all such 9 scalar inequalities are satisfied by (42). Interestingly, despite being strongly elliptic, (42) is not polyconvex. To see this, as also elaborated in Appendix C, it suffices to recognize that (46) is not convex in \( \bar{T}_2 \) to leading order in the limit of small deformations, and hence that (42) is not convex in \( \bar{T}_2 \).

vi. By construction, the microstructure associated with the result (42) corresponds to a dilute isotropic distribution of rigid particles that interact in such a manner that the stress within each particle is uniform and the same in all particles, irrespectively of the value of the applied macroscopic stretches \( \bar{\tau}_1 \) and \( \bar{\tau}_2 \). For small enough deformations, such a special stress field is in precise agreement with that of a dilute distribution of rigid spheres (Eshelby, 1957). This is the key reason why the result (42) recovers the classical Einstein–Smallwood result in the limit of small deformations. For finite deformations, on the other hand, the intraparticle stress field in a dilute distribution of spherical particles does not remain uniform. Yet, the large-deformation response of a dilute distribution of rigid spherical particles is expected to be very similar to that characterized by (42). This expectation is based on the argument that the effect of particle interactions on the overall response of dilute suspensions should be small (even at large deformations), and hence that different interactions associated with different dilute isotropic distributions of particles should lead to similar overall responses. This argument is supported by comparisons with the FE simulations presented next.

4. FE solutions for a rigid spherical inclusion in a block of rubber under large deformations

In the sequel, we work out a 3D FE solution for the overall large-deformation response of a block of Neo-Hookean rubber that contains a single rigid spherical inclusion of infinitesimal size at its center. The comparison between this
solution for an isolated spherical particle with the above-derived analytical solution for an isotropic distribution of particles shall shed light on the importance of particle interactions in the overall elastic response of dilute suspensions of rigid particles in rubber.

4.1. The FE model

For convenience and without loss of generality, we consider the block of Neo-Hookean rubber to be a cube of side $L$ in its undeformed stress-free configuration. Given that the radius of the spherical inclusion, $a$, say, in the FE model must be necessarily finite, we need to identify how small its concentration $c = 4\pi a^3 / 3L^3$ ought to be in order to accurately approximate an actual infinitesimal particle with $c \rightarrow 0$. To this end, we carried out a parametric study with decreasing values of $c$ ranging from $10^{-6}$ to $10^{-11}$. For the kind of deformations of interest in this work, the results indicate that concentrations $c \leq 10^{-8}$ are sufficiently small to be representative of an infinitesimal particle. Accordingly, in this work we set the particle concentration at

$$c = \frac{125\pi}{48} \times 10^{-9},$$

(49)

corresponding to a spherical inclusion of radius $a = 1$ in a cube of side $L = 800$.

Having identified the geometry of the block and of the particle, we now turn to their discretization. We first note that there is no need to mesh the particle in order to model exactly its rigid behavior, but that instead it suffices to spatially fix the particle/matrix interface in its undeformed configuration. We further note that the geometric and constitutive symmetry of the problem allows to perform the calculations in just one octant of the cube. A mesh generator code is utilized to construct the 3D geometry for such an octant, as depicted in Fig. 2. Small elements are placed near the rigid particle at uniform angular intervals of $3^\circ$, while the radial length is gradually increased toward the outer boundary. In all, the mesh consists of 18,900 brick elements with 675 elements on a radial plane and 28 layers along the radial direction. This discretization was selected after various mesh refinements were tried to assess sufficient mesh convergence. In selecting an appropriate type of finite element, we tested 8-node linear and 20-node quadratic hybrid elements, where the pressure is treated as a further degree of freedom in order to be able to handle the incompressibility of the Neo-Hookean rubber exactly (in a numerical sense). Although both elements generated similar results, a close inspection revealed that more consistent behaviors with the known Einstein–Smallwood solution at very small loads were obtained with the 20-node element model. We thus make use here of higher-order 20-node elements for the analysis. Since the computations are carried out using the FE package ABAQUS, we make use in particular of the C3D20H hybrid elements available in this code (see ABAQUS Version 6.11 Documentation).

4.2. Computation of the overall elastic response

As already discussed within the more general context of Section 2, the overall elastic response of the above-defined reinforced block of Neo-Hookean rubber amounts to computing the total elastic energy per unit undeformed volume when the outer boundary of the block is subjected to the affine displacement boundary condition (6). Similar to the analytical approach presented above, here it also proves convenient to restrict attention — without loss of generality — to isochoric pure stretch loadings of the form (13). In terms of these stretch variables and based on the parametric study performed for decreasing values of particle concentration $c \in [10^{-6}, 10^{-11}]$, the effective stored-energy function computed from the FE
model turns out to be of the expected asymptotic form
\[
W_{\text{FE}}^e(\lambda_1, \lambda_2, c) = \frac{\mu}{2} \left[ \frac{\lambda_1^2}{\lambda_1^2 + \lambda_2^2 + \frac{1}{\lambda_1 \lambda_2}} - 3 \right] + 2 \mu H^e(\lambda_1, \lambda_2) c + O(c^2),
\]
(50)

where it is worth remarking that this asymptotic behavior in the limit as \( c \to 0^+ \) is of identical polynomial form as that of the analytical solution (42). It is also important to emphasize that the correction term in (50) is in the order of 10^{-8} (i.e., in the order of the particle concentration (49)), and hence that the computation of \( W_{\text{FE}}^e \) must be carefully carried out in double precision in order to be able to accurately determine the correcting function \( H^e \).

A convenient manner to numerically implement the affine boundary conditions (6) with (13) is to follow radial straining paths in principal-logarithmic-strain space (\( \ln \lambda_i \)). Specifically, we set
\[
\lambda_1 = \lambda \quad \text{and} \quad \lambda_2 = \lambda^m
\]
(51)

(and hence \( \lambda_3 = (\lambda_1 \lambda_2)^{-1} = \lambda^{-(1 + m)} \)), where \( \lambda \) is a positive load parameter that takes the value of 1 in the undeformed configuration and \( m \in \mathbb{R} \). Any desired macroscopic deformation state \((\lambda_1, \lambda_2, \lambda_3 = (\lambda_1 \lambda_2)^{-1})\) can be accessed by marching along (starting at \( \lambda = 1 \)) radial paths (51) with appropriate fixed values of \( m \). Because of the overall isotropy and incompressibility of the response it actually suffices to consider \( \lambda \geq 1 \) and \( m \in [-0.5, 1] \), where \( m = -0.5 \) and \( m = 1 \) correspond to axisymmetric tension (or, equivalently, biaxial compression) and axisymmetric compression (or, equivalently, biaxial tension), respectively. Fig. 3 shows FE results for seven different loading paths with \( \lambda \geq 1 \) and values of \( m = -0.5, -0.25, 0, 0.25, 0.5, 0.75 \) and 1.0. Results are shown for the radial loading paths in principal-logarithmic-strain space in part (a), and for the correcting function
\[
H^e(\lambda_1, \lambda_2) = \frac{1}{c} \left[ \frac{1}{2 \mu} W_{\text{FE}}^e(\lambda_1, \lambda_2, c) - \frac{1}{4} \left( \frac{\lambda_1^2}{\lambda_1^2 + \frac{1}{\lambda_1 \lambda_2}} - 3 \right) \right],
\]
(52)
in stretch space in part (b). The entire correcting function \( H^e \) can be constructed by carrying out further computations with \( \lambda \geq 1 \) and \( m \in [-0.5, 1] \), and by exploiting the inherent symmetries \( H^e(\lambda_1, \lambda_2) = H^e(\lambda_2, \lambda_1) = H^e(\lambda_1, \lambda_2)^{-1} = H^e(\lambda_2, \lambda_1)^{-1} \).

5. Results and discussion

Figs. 4 and 5 present results for the overall response of Neo-Hookean rubber reinforced by a dilute isotropic distribution of rigid particles, as characterized by the analytical solution (42), and the FE simulations of the preceding section for the overall response of a Neo-Hookean block of rubber reinforced by a single rigid spherical particle. For clarity of presentation, results are shown for the correcting functions \( H \) and \( H^e \) instead of the stored-energy functions \( W \) and \( W^e \) themselves.

Fig. 4(a) and (b) provide a full 3D comparison between the analytical and FE solutions in \( \lambda_1 - \lambda_2 \)-space. To aid the visualization of the quantitative differences, parts (c) through (f) of the figure also provide 2D views of both solutions along various fixed deformation paths: parts (c) and (d) display results for axisymmetric tension \((\lambda_1 = \lambda_2 = \lambda \leq 1)\) and
Fig. 4. Comparison of the analytical solution (42) for the overall response of Neo-Hookean rubber reinforced by a dilute isotropic distribution of rigid particles with the FE solution (50) for the overall response of a Neo-Hookean block of rubber reinforced by a single rigid spherical particle. The results are shown for the correcting functions $H$ (part (a)) and $H^{\text{FE}}$ (part (b)) in terms of the stretches $\lambda_1$ and $\lambda_2$ in $\lambda_1$-$\lambda_2$-space, as well as along various fixed deformation paths: (c) $\lambda_1 = \lambda_2 = \lambda < 1$, (d) $\lambda_1 = \lambda_2 = \lambda > 1$, (e) $\lambda_1 = \lambda_2 = 1$, and (f) $\lambda_1 = \lambda_2 = \lambda^{0.5}$. Compression ($\lambda_1 = \lambda_2 = \lambda \geq 1$), whereas parts (e) and (f) display results for pure shear ($\lambda_1 = \lambda, \lambda_2 = 1$) and a further intermediate deformation path ($\lambda_1 = \lambda_2 = \lambda^{0.5}$). In all these plots, the solid line corresponds to the analytical solution, while the dashed line denotes the FE results.

An immediate observation from Fig. 4 is that the FE results are in good qualitative and quantitative agreement with the analytical solution (42) for all loading conditions. More specifically, the FE results are practically identical to the analytical
solution up to sufficiently large deformations after which they start to exhibit a consistently stiffer behavior. The largest discrepancy between the two results occurs along axisymmetric compression (shown in Fig. 4(d)), but even in this case the quantitative difference is less than 7% at the maximum stretch of $\bar{\varepsilon}_1 = \bar{\varepsilon}_2 = 2.5$ reached with the FE model.

To further explore the connections between the analytical and FE solutions, Fig. 5 provides plots for $H$ and $H^{FE}$ as functions of the principal invariants $T_1 = \bar{\varepsilon}_1^2 + \bar{\varepsilon}_2^2 + \bar{\varepsilon}_3^2$ and $T_2 = \bar{\varepsilon}_1^2 + \bar{\varepsilon}_2^2 + \bar{\varepsilon}_3^2$. Part (a) of the figure shows $H$ and $H^{FE}$ for fixed values of the first invariant $T_1 = 4, 6, 8$, and 10 as functions of $T_2$. Here, it is appropriate to recall that the constraint of incompressibility imposes a restriction on the physically allowable values of $T_1$ and $T_2$. Thus, for fixed $T_2 = 4$ and 6 the first invariant is restricted to take values in the ranges $T_1 \in [3.71, 4.52]$ and $T_1 \in [4.72, 9.34]$, respectively. For fixed $T_1 = 4, 6, 8$, and 10, the corresponding allowable values of the second invariant are $T_2 \in [3.71, 5.24], T_2 \in [4.72, 9.34], T_2 \in [5.53, 16.25], and T_2 \in [6.22, 25.20]$. These are the ranges of values utilized in the figure.

Similar to Fig. 4, Fig. 5 shows that indeed the FE results are in good agreement with the analytical solution, being slightly stiffer at large deformations. More importantly, Fig. 5 serves to illustrate that both solutions are approximately linear in the first invariant $T_1$ and independent of second invariant $T_2$. That is, in addition to being similar quantitatively, the analytical and FE solutions are essentially identical in their functional character.

The agreement between the FE and analytical solutions revealed by the above results is somewhat remarkable, given that they correspond to different microstructures: while the FE results correspond to the overall response of a block of rubber reinforced by a single rigid spherical particle, the analytical solution corresponds to the overall response of rubber reinforced by a specific class of isotropic distribution of rigid particles. Again, while in the case of the FE result the particle is isolated and hence does not interact with other particles, in the case of the analytical solution the underlying particles do interact with each other in such a manner that their stress is uniform. The close functional and quantitative agreement between the two results thus suggests that the interaction among particles does not play an important role in the overall nonlinear elastic response of dilute suspensions of rigid particles in rubber, even at large deformations. In turn, this suggests that different dilute isotropic distributions of particles exhibiting different particle interactions lead to similar overall responses.

6. An approximate closed-form solution for dilute suspensions

The evaluation of the effective stored-energy function (42) requires knowledge of the function $H$, which ultimately amounts to solving numerically the Eikonal pde (37) subject to the initial condition (38). In this section, we propose an approximate closed-form solution for (37)–(38), very close to the exact solution, which allows in turn to generate a closed-form approximation for (42).

The approximation is based on the observation that the function $H$ is linear in the invariant $T_1$ and independent of $T_2$ in the limiting regimes of small and large deformations; see remarks iii and iv in Section 3.2. For intermediate deformations, $H$ does depend nonlinearly on $T_1$ and on the second invariant $T_2$, but both these dependencies are exceptionally weak, as illustrated in Fig. 5. Thus, we can readily generate an approximate solution that agrees identically with the exact solution.
or, equivalently,

$$W(\lambda_1, \lambda_2, c) = \mu \frac{5}{8} [I_1 - 3] + \frac{5 \mu}{4} [I_1 - 3] c.$$  

Because of the properties of (53), the approximate solution (54) is identical to the exact solution (42) in the limit of small deformations — and hence recovers the classical Einstein–Smallwood result — and quantitatively very close to (42) for arbitrarily large deformations. In addition, the result (54) is functionally very similar to (42) in that it is linear in I_1, independent of I_2, and strongly elliptic (see Appendix C). The approximate solution (54) provides then a mathematically simple, quantitatively accurate, and functionally sound result — which can be utilized in lieu of (42) for all practical purposes — for the overall elastic response of Neo-Hookean solids reinforced by a dilute isotropic distribution of rigid particles under arbitrarily large deformations.

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Appendix A. The coefficients x_1, x_2, x_3

The coefficients x_1, x_2, x_3 in the pdes (24), (34), and (37) are given by

$$x_1(\lambda_1, \lambda_2) = \frac{5}{3} \frac{\lambda_1^5 + \lambda_2^5 - 2 \lambda_1^2 \lambda_2^2}{(\lambda_1 - \lambda_2)^2 (\lambda_2^2 - \lambda_1^2)} I_1 + \frac{2}{3} \frac{\lambda_1^3 + \lambda_2^3 - 2 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)^2 (\lambda_2^2 - \lambda_1^2)} I_3 + \frac{2}{3} \frac{\lambda_1^4 + \lambda_2^4 - 2 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)^2 (\lambda_2^2 - \lambda_1^2)} I_3 + \frac{2}{3} \frac{\lambda_1^4 + \lambda_2^4 - 2 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)^2 (\lambda_2^2 - \lambda_1^2)} I_3 + \frac{2}{3} \frac{\lambda_1^4 + \lambda_2^4 - 2 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)^2 (\lambda_2^2 - \lambda_1^2)} I_3.$$
\[\begin{align*}
\alpha_2(\bar{\tau}_1, \bar{\tau}_2) &= \frac{\bar{\tau}_1^2 \bar{\tau}_2^2}{3(\bar{\tau}_1^2 - \bar{\tau}_2^2)(\bar{\tau}_1^2 - \bar{\tau}_2^2)} \Gamma_F - \frac{2\bar{\tau}_1^2 \bar{\tau}_2^2 (\bar{\tau}_1^2 \bar{\tau}_2^2 - 2\bar{\tau}_1^2 \bar{\tau}_2^2 + 1)}{3(\bar{\tau}_1^2 - \bar{\tau}_2^2)^2 (\bar{\tau}_1^2 - \bar{\tau}_2^2)} \Gamma_E + \frac{(\bar{\tau}_1^2 + 1)\bar{\tau}_1^2 - 2\bar{\tau}_1^2 \bar{\tau}_2^2 - \bar{\tau}_1^2 \bar{\tau}_2^2 + \bar{\tau}_1^2 \bar{\tau}_2^2}{3(\bar{\tau}_1^2 - \bar{\tau}_2^2)^2 (\bar{\tau}_1^2 - \bar{\tau}_2^2)}.
\end{align*}\]

with

\[\begin{align*}
\Gamma_F &= \frac{1}{\sqrt{1 - \bar{\tau}_1^2}} \mathcal{E}_F \left[ \frac{\sqrt{1 - \bar{\tau}_1^2}}{2\sqrt{\bar{\tau}_1^2 - 1}} \ln \left( \frac{(\bar{\tau}_1^2 + \sqrt{\bar{\tau}_1^2 + 1})}{\bar{\tau}_1^2 - 1} \right) \right],
\Gamma_E &= \frac{1}{\sqrt{1 - \bar{\tau}_1^2}} \mathcal{E}_E \left[ \frac{\sqrt{1 - \bar{\tau}_1^2}}{2\sqrt{\bar{\tau}_1^2 - 1}} \ln \left( \frac{(\bar{\tau}_1^2 + \sqrt{\bar{\tau}_1^2 + 1})}{\bar{\tau}_1^2 - 1} \right) \right].
\end{align*}\]

where the functions \(\mathcal{E}_F\) and \(\mathcal{E}_E\) stand for, respectively, the elliptic integrals of first and second kind, as defined by

\[\mathcal{E}_F(\varphi; t) = \int_0^t [1 - t^2 \sin^2 \varphi]^{-1/2} \, d\varphi \quad \text{and} \quad \mathcal{E}_E(\varphi; t) = \int_0^t [1 - t^2 \sin^2 \varphi]^{1/2} \, d\varphi.\]

By direct inspection we remark that

\[\begin{align*}
\alpha_1(\bar{\tau}_1, \bar{\tau}_2) &= \alpha_2(\bar{\tau}_2, \bar{\tau}_1),
\alpha_3(\bar{\tau}_1, \bar{\tau}_2) &= \alpha_3(\bar{\tau}_2, \bar{\tau}_1),
\alpha_1(\bar{\tau}_1, \bar{\tau}_2) &= \alpha_1(\bar{\tau}_1, (\bar{\tau}_1 \bar{\tau}_2)^{-1}),
\alpha_2(\bar{\tau}_1, \bar{\tau}_2) &= \alpha_1(\bar{\tau}_1, (\bar{\tau}_1 \bar{\tau}_2)^{-1}) + \frac{\alpha_3(\bar{\tau}_1, (\bar{\tau}_1 \bar{\tau}_2)^{-1})}{\bar{\tau}_1^2 \bar{\tau}_2^2},
\alpha_3(\bar{\tau}_1, \bar{\tau}_2) &= -\frac{2\alpha_1(\bar{\tau}_1, (\bar{\tau}_1 \bar{\tau}_2)^{-1})}{\bar{\tau}_1^2 \bar{\tau}_2^2} - \frac{\alpha_3(\bar{\tau}_1, (\bar{\tau}_1 \bar{\tau}_2)^{-1})}{\bar{\tau}_1^2 \bar{\tau}_2^2},
\alpha_1(\bar{\tau}_1, \bar{\tau}_2) &= \alpha_1((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2) + \frac{\alpha_3((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{\bar{\tau}_1^2 \bar{\tau}_2^2},
\alpha_2(\bar{\tau}_1, \bar{\tau}_2) &= -\frac{2\alpha_2((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{\bar{\tau}_1^2 \bar{\tau}_2^2} - \frac{\alpha_3((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{\bar{\tau}_1^2 \bar{\tau}_2^2},
\alpha_3(\bar{\tau}_1, \bar{\tau}_2) &= \frac{3\alpha_1((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{24(\bar{\tau}_1 - 1)^2} + \frac{\alpha_2((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{8(1 - \bar{\tau}_1)^{3/2}} \ln \left( 1 + \frac{\sqrt{1 - \bar{\tau}_1^6}}{\bar{\tau}_1^2} \right),
\alpha_3(\bar{\tau}_1, \bar{\tau}_2) &= \frac{3\alpha_1((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{12(\bar{\tau}_1 - 1)^2} + \frac{\alpha_2((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2)}{4(1 - \bar{\tau}_1)^{3/2}} \ln \left( 1 + \frac{\sqrt{1 - \bar{\tau}_1^6}}{\bar{\tau}_1^2} \right).\end{align*}\]

We further remark that

\[\frac{1}{3} \leq \alpha_1(\bar{\tau}_1, \bar{\tau}_2) \leq 0, \quad \frac{1}{3} \leq \alpha_2(\bar{\tau}_1, \bar{\tau}_2) \leq 0, \quad 0 \leq \alpha_3(\bar{\tau}_1, \bar{\tau}_2) \leq \frac{1}{6} \quad \forall \bar{\tau}_1, \bar{\tau}_2 > 0.\]

**Appendix B. The function \(H\)**

In this appendix, we sketch out the main properties of the function \(H : [(a, b) \in \mathbb{R}^2 : a, b > 0] \to \mathbb{R}\) defined by the Eikonal initial-value problem (37)–(38) and provide details on its numerical computation. We begin by recognizing from relations (59)–(61) that Eqs. (37)–(38) admit solutions such that

\[H(\bar{\tau}_1, \bar{\tau}_2) = H(\bar{\tau}_2, \bar{\tau}_1) = H(\bar{\tau}_1, (\bar{\tau}_1 \bar{\tau}_2)^{-1}) = H((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_1) = H((\bar{\tau}_1 \bar{\tau}_2)^{-1}, \bar{\tau}_2) \quad \forall \bar{\tau}_1, \bar{\tau}_2,\]

as required by the last of the conditions (26). A direct implication (see, e.g., Chapter 4 in the monograph by Ogden, 1997) of
the symmetry properties (63) is that $H$ may be written in the polynomial form

$$H(\ell_1, \ell_2) = \sum_{p,q=0}^{\infty} k_{pq}(\ell_1-1)^p(\ell_2-1)^q,$$

(64)

where it is emphasized that the coefficients $k_{pq}$ are not entirely independent but constrained by conditions (63). Substituting the representation (64) in (37)–(38) and taking the limit of small deformations as $\ell_1 \to 1$ and $\ell_2 \to 1$ leads to a hierarchy of systems of algebraic equations for the unknown coefficients $k_{pq}$. These systems are linear and hence have a unique solution, however, they do not appear to admit a simple recurrence solution and must therefore be solved successively one at a time. For the first four sets of equations, the solutions read as

$$k_{00} = 0, \quad k_{10} = k_{01} = 0, \quad k_{20} = k_{02} = k_{11} = \frac{5}{2}, \quad k_{30} = k_{03} = -\frac{5}{2}, \quad k_{21} = k_{12} = -\frac{55}{14}.$$

(65)

According to the result (64) with (65), the point $\ell_1 = \ell_2 = 1$ corresponds to a local minimum of $H$ at which $H(1,1) = 0$. Now, from the pde (37) it is easy to deduce that the value of the function $H$ evaluated at any critical point (i.e., any point $\ell_1, \ell_2$ at which $\partial H / \partial \ell_1 = \partial H / \partial \ell_2 = 0$) must be necessarily zero. These two results entail then that the point $\ell_1 = \ell_2 = 1$ is the only critical point of the function $H$ defined by (37)–(38), that this point corresponds to its global minimum, and hence that

$$H(1,1) = 0 \quad \text{and} \quad H(\ell_1, \ell_2) > 0 \quad \forall \ell_1, \ell_2 \neq 1,$$

(66)

as required by the first two conditions (26).

It also follows from (64) with (65) that in the limit of small deformations as $\ell_1 \to 1$ and $\ell_2 \to 1$, the function $H$ is indeed given explicitly by relation (39) in the main body of the text. In the opposite limit of infinitely large deformations as $\ell_1 \to +\infty$, it is not difficult to recognize that Eqs. (37)–(38) admit the explicit asymptotic solution

$$H(\ell_1, \ell_2) = \frac{3}{4} \ell_1^{-2} \left[ \frac{9}{16} (\ln 4 + 2 \ln 2) + 4 \ln \ell_1 \ell_2^{-1} + O(\ell_1^{-2}).ight.$$

(67)

From the symmetry condition $H(\ell_1, \ell_2) = H(\ell_2, \ell_1)$ it follows that

$$H(\ell_1, \ell_2) = \frac{3}{4} \ell_2^{-2} \left[ \frac{9}{16} (\ln 4 + 2 \ln 2) + 4 \ln \ell_1 \ell_1^{-1} + O(\ell_2^{-2}),ight.$$  

(68)

for $\ell_2 \to +\infty$. Moreover, the asymptotic solution of Eqs. (37)–(38) for the case when $\ell_1 \to 0$ is given by

$$H(\ell_1, \ell_2) = \frac{3}{4} \ell_1^{2} + \frac{3}{4} \ell_2^{2} + O(\ell_1 \ln \ell_1),$$

(69)

and from the symmetry $H(\ell_1, \ell_2) = H(\ell_2, \ell_1)$ we also then have that

$$H(\ell_1, \ell_2) = \frac{3}{4} \ell_2^{-1} + \frac{3}{4} \ell_1^{-1} + O(\ell_1 \ln \ell_2),$$

(70)

for infinitely large deformations with $\ell_2 \to 0$. Combining results (67)–(70) it readily follows that in the limit of infinitely large deformations (as $\ell_1 \to 0, +\infty$ and/or $\ell_2 \to 0, +\infty$) the function $H$ defined by Eqs. (37)–(38) is given explicitly, to leading order, by relation (40) in the main body of the text.

The numerical solution of the initial-value problem (37)–(38) for $H$ can be generated in a number of different ways using finite differences. We found it more efficient to consider the problem in the alternative set of variables $\tilde{\ell}_1 = \ell_1 / \ell_2$ and $\tilde{\ell}_2 = \ell_1 / \ell_2$, instead of in terms of the principal stretches $\ell_1$ and $\ell_2$ directly. The advantage of these variables is twofold: (i) the finite-difference discretization can be performed on a simple Cartesian grid with unilateral boundaries $\tilde{\ell}_1 \geq 1$ and $\tilde{\ell}_2 \geq 1$, and (ii) the initial condition (38) in $\tilde{\ell}_1 - \tilde{\ell}_2$-space is given at the constant value of $\tilde{\ell}_2 = 1$. The commercial package Wolfram Mathematica 8.0 was utilized to discretize and solve the equations. In spite of the quadratic nonlinearity of the pde (37), we note that the initial-value problem (37)–(38) admits only one solution that is consistent with the required conditions (63) and (66).

Appendix C. Conditions for strong ellipticity and polyconvexity of $W$

Explicit necessary and sufficient conditions for an isotropic incompressible stored-energy function to be strongly elliptic have been provided by Zee and Sternberg (1983). When the stored-energy function is written in the form $W = W(\ell_1, \ell_2)$ with $\ell_3 = \ell_1^{-1} \ell_2^{-1}$, as done in the present work, the conditions read as

$$\beta_i > 0 \quad (i = 1, 2, 3),$$

$$w_1 + 2\beta_1 \beta_i > 0 \quad (i = 1, 2, 3; \text{ no summation}),$$

$$\ell_2^{-1} \sqrt{w_2 + 2\beta_2 \beta_3} + \ell_3^{-1} \sqrt{w_3 + 2\beta_3 \beta_1} > \ell_1^{-2} (w_1 - 2\ell_1 \beta_1) > 0,$$
An incompressible stored-energy function $\mathbf{W}$ is strongly elliptic, it suffices to show that the function $H$ is strongly elliptic. This follows from the facts that the Neo-Hookean term $\mu/2[\mathcal{E}^2 + 3\mathcal{L}^2 - 3]$ in (42) is strongly elliptic and that the sum of strongly elliptic functions is strongly elliptic. Now, by making use of the explicit asymptotic expressions (39) and (40), it is straightforward to show analytically that $H$ satisfies all nine conditions (71) for small and large deformations. For arbitrary deformations, it is also straightforward to show — albeit by numerical means — that $H$ satisfies conditions (71), and hence that the effective stored-energy function (42) is strongly elliptic. By the same token, we note that the approximate solution (54) for $W$ is strongly elliptic, since the underlying approximation (53) for $H$ is strongly elliptic.

An incompressible stored-energy function $\mathbf{W} = \mathbf{W}(\mathcal{X}, \mathcal{X}_2)$ is said to be polyconvex if it can be written in the form

$$\mathbf{W} = \mathbf{W}(\mathbf{F}, \mathbf{F}^T),$$

with $\mathbf{W}(\cdot, \cdot)$ convex. The constitutive restriction (77) of polyconvexity is a stronger constitutive restriction than that of strong ellipticity (71) — in fact, polyconvexity implies strongly ellipticity — that was introduced by Ball (1977) to prove existence theorems in finite elasticity. Unlike strong ellipticity (see Geymonat et al., 1993), however, polyconvexity has not yet been given a strict physical interpretation and therefore its enforcement, although mathematically desirable, is still physically arguable.

For the case under study here, it is a trivial matter to deduce from its explicit asymptotic form (46) — after recognizing that $\mathbf{T}_1 = \mathbf{F} \cdot \mathbf{F}$ and $\mathbf{T}_2 = \mathbf{F}^T \cdot \mathbf{F}^T$ — that the effective stored-energy function (42) is not convex in $\mathbf{F}^T$ and hence not polyconvex.

References


Lopez-Pamies, O., 2008. Linear comparison estimates for the finite deformation of hyperelastic solids reinforced by ellipsoidal particles. Unpublished work.


