



PROPAGATION OF MICROSCALE MATERIAL UNCERTAINTY IN A CLASS OF HYPERELASTIC FINITE DEFORMATION STORED ENERGY FUNCTIONS

Tarek I. Zohdi

Department of Mechanical Engineering

6195 Etcheverry Hall

University of California, Berkeley, California, 94720-1740, USA

zohdi@newton.berkeley.edu fax. 512-642-6163

Abstract. Whenever effective material responses are determined, there is a degree of microscale uncertainty. For example in the case of overall isotropic responses, we have uncertainties in the effective constitutive parameters, such as $\mu^* + \Delta\mu^*$ and $\kappa^* + \Delta\kappa^*$. In this communication bounds on the resulting uncertainty in a class of polyconvex macroscale finite deformation stored energy functions, which employ the uncertain material parameters, are determined.

1. Stored energy representations. In many applications involving compressible polymeric materials, finite deformations are expected. Representative examples of such materials are polyethelenes, which have the largest volume use of any plastic in the world. Typically, they are prepared by chemically treating a vulcanized polymers, for example catalytic polymerization of ethylene. Such processes can result in various degrees of inhomogeneity, typically in the form of inclusions, throughout the material. For macroscale analyses at finite deformations of such materials, a widely used class of mathematical representations for the constitutive response are compressible hyperelastic stored energy functions. Analogous to the familiar infinitesimal strain case, an elastic material is called hyperelastic if there exists a stored energy function, W , that is only a function of the mechanical deformation, and that $\mathbf{S} = 2 \frac{\partial W}{\partial \mathbf{C}} = \frac{\partial W}{\partial \mathbf{E}}$ or $\mathbf{P} = \frac{\partial W}{\partial \mathbf{F}}$, where $\mathbf{F} = \nabla_{\mathbf{X}} \mathbf{x}$ is the deformation gradient, $\mathbf{u} = \mathbf{x} - \mathbf{X}$, \mathbf{X} are referential coordinates and \mathbf{x} are current coordinates, J is the Jacobian of \mathbf{F} , $J = \det \mathbf{F}$, $\mathbf{C} \stackrel{\text{def}}{=} \mathbf{F}^T \cdot \mathbf{F}$ is the right Cauchy-Green strain tensor, $\mathbf{E} \stackrel{\text{def}}{=} \frac{1}{2}(\mathbf{C} - \mathbf{1})$ is the Green-Lagrange strain tensor, $\mathbf{P} = \boldsymbol{\sigma} \cdot \mathbf{F}^{-T} J$ is the first Piola-Kirchhoff stress, $\boldsymbol{\sigma}$ is the Cauchy stress and where $\mathbf{S} = J \mathbf{F}^{-1} \cdot \boldsymbol{\sigma} \cdot \mathbf{F}^{-T}$ is the second Piola-Kirchhoff stress. In addition to being material frame indifferent, any admissible stored energy function must obey five criteria: (1) $\mathbf{C} = \mathbf{1} \Leftrightarrow \mathbf{F} = \mathbf{1} \Leftrightarrow (I_C = II_C = 3, III_C = 1) \Leftrightarrow W = 0$, where (I_C, II_C, III_C) are the principal invariants of \mathbf{C} , (2) $W \geq 0$, (3) $W \rightarrow \infty \Leftrightarrow \det \mathbf{F} \rightarrow 0$ or $\det \mathbf{F} \rightarrow \infty$, (4) $\mathbf{S} = \mathbf{0}$ for $\mathbf{C} = \mathbf{1}$ and (5) the material constants in a finite deformation material law must give hyperelastic responses with known material constants, for example in isotropic cases, the Lamé parameters, $\lambda^* (= \kappa^* - \frac{2\mu^*}{3})$ and μ^* , when perturbed around the undeformed configuration. A relatively simple family of representations which satisfy the previously mentioned requirements are compressible Mooney-Rivlin models

$$W(\mathbf{C}) = \underbrace{K_1^*(\bar{I}_C - 3) + K_2^*(\bar{II}_C - 3)}_{\text{incompressible part}} + \underbrace{\frac{\kappa^*}{2}(\sqrt{III_C} - 1)^2}_{\text{compressible part}}, \quad (1)$$

where $K_1^* + K_2^* = \frac{\mu^*}{2}$. For this class of models, the first and second invariants of \mathbf{C} , I_C and II_C , have been scaled by roots of the third invariant, $\bar{I}_C = I_C III_C^{-\frac{1}{3}} = I_C J^{-\frac{2}{3}}$ and $\bar{II}_C = II_C III_C^{-\frac{2}{3}} = II_C J^{-\frac{4}{3}}$, to insure that they contribute nothing to the compressible part of the response. We remark that when $K_1 = \frac{\mu}{2}$ and $K_2 = 0$, the material is called a compressible Neo-Hookean material. Compressible Neo-Hookean and compressible Mooney-Rivlin material response functions are known to be polyconvex, and thus possess desirable mathematical (existence) properties (Ball [1]). For general overviews see Ciarlet [2] or Ogden [3]. Without loss of generality, for the remainder of the analysis, we consider a convex combination of the material constants, $K_1^* + K_2^* = \phi \frac{\mu^*}{2} + (1 - \phi) \frac{\mu^*}{2} = \frac{\mu^*}{2}$, $0 \leq \phi \leq 1$.

2. Microscale uncertainties. In reality, for microheterogeneous materials, such as a chemically treated vulcanized polymer, the coefficients in Equation 1 are the effective bulk and shear moduli, $3\kappa^* \langle \frac{tr \boldsymbol{\epsilon}}{3} \rangle_\Omega = \langle \frac{tr \boldsymbol{\sigma}}{3} \rangle_\Omega$ and $2\mu^* \langle \boldsymbol{\epsilon}' \rangle_\Omega = \langle \boldsymbol{\sigma}' \rangle_\Omega$, which are the Lamé parameters of $\langle \boldsymbol{\sigma} \rangle_\Omega = \mathbf{IE}^* : \langle \boldsymbol{\epsilon} \rangle_\Omega$, where $\langle \cdot \rangle_\Omega \stackrel{\text{def}}{=} \frac{1}{|\Omega|} \int_\Omega \cdot d\Omega$, and where $\boldsymbol{\sigma}$ and $\boldsymbol{\epsilon}$ are the Cauchy stress and infinitesimal strain tensor fields within a statistically representative volume element (RVE) of volume $|\Omega|$. The quantity, \mathbf{IE}^* , is known as the effective property, and is the elasticity tensor used in usual structural scale analyses. For the effective response to be meaningful, i.e. statistically representative, it must be computed over a sample containing a large amount of material. In other words, the sample must be so large, that for further enlargements \mathbf{IE}^* does not change. If one were to attempt to perform a direct numerical simulation of a truly statistically representative volume element of material, incorporating all of the microscale details, an extremely fine spatial discretization mesh, for example that of a finite element mesh, would be required in order to accurately resolve the microstructural fields. In order to give an idea of the immensity of the computational problem, consider that in materials possessing particulate microstructure, a cubical sample of dimensions $0.0001m \times 0.0001m \times 0.0001m$, *which may still not even be statistically representative*, can typically contain on the order of 1000-10000 randomly distributed particles or pores. For three dimensional problems, thousands of numerical degrees of freedom per particle are needed to deliver numerically accurate solutions, and thus the resulting system of equations would contain several million numerical unknowns. This is the primary reason that effective property bounds are popular. Rigorously speaking, the simplest upper and lower bounds on the effective response are the Hill-Reuss-Voigt bounds [4], [6], [5]: $\langle \mathbf{IE}^{-1} \rangle_\Omega^{-1} \leq \mathbf{IE}^* \leq \langle \mathbf{IE} \rangle_\Omega$. The notation means that the difference tensors $(\langle \mathbf{IE} \rangle_\Omega - \mathbf{IE}^*)$ and $(\mathbf{IE}^* - \langle \mathbf{IE}^{-1} \rangle_\Omega^{-1})$ are positive definite. For isotropic macroscopic responses with isotropic phases this implies $\langle \kappa^{-1} \rangle_\Omega^{-1} \leq \kappa^* \leq \langle \kappa \rangle_\Omega$ and $\langle \mu^{-1} \rangle_\Omega^{-1} \leq \mu^* \leq \langle \mu \rangle_\Omega$, where κ and μ are the spatially variable bulk and shear moduli of material. In 1963, such bounds were improved by Hashin and Shtrikman [7], [8], for isotropic materials with an isotropic effective response, resulting in the following

$$\begin{array}{c}
\boxed{
\begin{array}{ccc}
\kappa_1 + \frac{v_2}{\frac{1}{\kappa_2 - \kappa_1} + \frac{3(1-v_2)}{3\kappa_1 + 4\mu_1}} \leq \kappa^* \leq \kappa_2 + \frac{1-v_2}{\frac{1}{\kappa_1 - \kappa_2} + \frac{3v_2}{3\kappa_2 + 4\mu_2}}, & & \\
\text{bulk modulus H/S lower bound} & & \text{bulk modulus H/S upper bound} \\
\mu_1 + \frac{v_2}{\frac{1}{\mu_2 - \mu_1} + \frac{6(1-v_2)(\kappa_1 + 2\mu_1)}{5\mu_1(3\kappa_1 + 4\mu_1)}} \leq \mu^* \leq \mu_2 + \frac{(1-v_2)}{\frac{1}{\mu_1 - \mu_2} + \frac{6v_2(\kappa_2 + 2\mu_2)}{5\mu_2(3\kappa_2 + 4\mu_2)}}, & & \\
\text{shear modulus H/S lower bound} & & \text{shear modulus H/S upper bound}
\end{array}
}
\end{array} \tag{2}$$

where κ_1, μ_1 and κ_2, μ_2 are the bulk and shear moduli for the phases, while v_2 is the second phase volume fraction. Such bounds are the tightest possible on isotropic effective responses, with isotropic two phase microstructures, where only the volume fractions and phase contrasts of the constituents are known. Therefore, we have an interval of uncertainty in the effective constitutive parameters, for example in the isotropic case, $\mu^* + \Delta\mu^*$ and $\kappa^* + \Delta\kappa^*$, which produces uncertainty in finite deformation stored energy functions that employ such parameters

$$W = \frac{\phi(\mu^* + \Delta\mu^*)}{2}(\bar{I}_C - 3) + \frac{(1-\phi)(\mu^* + \Delta\mu^*)}{2}(\bar{II}_C - 3) + \frac{(\kappa^* + \Delta\kappa^*)}{2}(\sqrt{\bar{III}_C} - 1)^2. \tag{3}$$

Due to thermodynamic restrictions, the bulk and shear moduli for any admissible system must be positive, thus $\mu^* + \Delta\mu^* > 0$ and $\kappa^* + \Delta\kappa^* > 0$, even if $\Delta\mu^* < 0$ and $\Delta\kappa^* < 0$. Due to the positivity of the components of the stored energy function, we have the following

$$\begin{array}{c}
\left| \frac{W^*(\kappa^* + \Delta\kappa^*, \mu^*) - W^*(\kappa^*, \mu^*)}{W^*(\kappa^*, \mu^*)} \right| = \frac{\frac{|\Delta\kappa^*|}{2}(\sqrt{\bar{III}_C} - 1)^2}{\underbrace{\frac{\mu^* \phi}{2}(\bar{I}_C - 3) + \frac{\mu^*(1-\phi)}{2}(\bar{II}_C - 3)}_{\geq 0} + \underbrace{\frac{\kappa^*}{2}(\sqrt{\bar{III}_C} - 1)^2}_{\geq 0}} \leq \frac{|\Delta\kappa^*|}{\kappa^*}, \\
\left| \frac{W^*(\kappa^*, \mu^* + \Delta\mu^*) - W^*(\kappa^*, \mu^*)}{W^*(\kappa^*, \mu^*)} \right| = \frac{\frac{|\Delta\mu^*| \phi}{2}(\bar{I}_C - 3) + \frac{|\Delta\mu^*|(1-\phi)}{2}(\bar{II}_C - 3)}{\underbrace{\frac{\mu^* \phi}{2}(\bar{I}_C - 3) + \frac{\mu^*(1-\phi)}{2}(\bar{II}_C - 3)}_{\geq 0} + \underbrace{\frac{\kappa^*}{2}(\sqrt{\bar{III}_C} - 1)^2}_{\geq 0}} \leq \frac{|\Delta\mu^*|}{\mu^*}.
\end{array} \tag{4}$$

The maximum range of uncertainty in the effective moduli are the differences in the upper and lower Hashin-Shtrikman bounds, which, when divided by the smallest normalizing factors, the lower bounds, lead to

$$\boxed{
\begin{aligned}
\left| \frac{W^*(\kappa^* + \Delta\kappa^*, \mu^*) - W^*(\kappa^*, \mu^*)}{W^*(\kappa^*, \mu^*)} \right| &\leq \frac{|\Delta\kappa^*|}{\kappa^*} \leq \frac{\kappa_2 + \frac{1-v_2}{\kappa_1 - \kappa_2} + \frac{3v_2}{3\kappa_2 + 4\mu_2}}{\kappa_1 + \frac{v_2}{\frac{1}{\kappa_2 - \kappa_1} + \frac{3(1-v_2)}{3\kappa_1 + 4\mu_1}}} - 1, \\
\left| \frac{W^*(\kappa^*, \mu^* + \Delta\mu^*) - W^*(\kappa^*, \mu^*)}{W^*(\kappa^*, \mu^*)} \right| &\leq \frac{|\Delta\mu^*|}{\mu^*} \leq \frac{\mu_1 + \frac{v_2}{\frac{1}{\mu_2 - \mu_1} + \frac{6(1-v_2)(\kappa_1 + 2\mu_1)}{5\mu_1(3\kappa_1 + 4\mu_1)}}}{\mu_2 + \frac{(1-v_2)}{\frac{1}{\mu_1 - \mu_2} + \frac{6v_2(\kappa_2 + 2\mu_2)}{5\mu_2(3\kappa_2 + 4\mu_2)}}} - 1.
\end{aligned}
} \tag{5}$$

As a numerical example, consider Polyethelene ($\kappa = 3.06$ GPa and $\mu = 0.26$ GPa) with inhomogeneities due that occur due to processing. Shown in Figure 1 are the behavior of the bounds in Box 5 as a function of volume fraction. Clearly, as the phase contrast is increased the bounds increase.

3. Concluding remarks. The corresponding second Piola-Kirchhoff stress tensor is

$$\mathbf{S} = 2 \left(K_1^* III_C^{-\frac{1}{3}} \mathbf{1} + K_2^* III_C^{-\frac{2}{3}} (I_C \mathbf{1} - \mathbf{C}) + \left(\frac{\kappa^*}{2} (III_C - III_C^{\frac{1}{2}}) - \frac{K_1^*}{3} I_C III_C^{-\frac{1}{3}} - \frac{2K_2^*}{3} II_C III_C^{-\frac{2}{3}} \right) \mathbf{C}^{-1} \right), \tag{6}$$

with the Cauchy stress being derived from $\boldsymbol{\sigma} = \frac{1}{J} \mathbf{F} \cdot \mathbf{S} \cdot \mathbf{F}^T$. Defining $\Delta_{\mu^*} \mathbf{S} \stackrel{\text{def}}{=} \mathbf{S}(\kappa^* + \Delta\kappa^*, \mu^*) - \mathbf{S}(\kappa^*, \mu^*)$ we have $\Delta_{\mu^*} \mathbf{S} = \Delta\kappa^* (III_C - III_C^{\frac{1}{2}}) \mathbf{C}^{-1}$, and $\Delta_{\mu^*} \mathbf{S} \stackrel{\text{def}}{=} \mathbf{S}(\kappa^*, \mu^* + \Delta\mu^*) - \mathbf{S}(\kappa^*, \mu^*)$

$$\Delta_{\mu^*} \mathbf{S} = \Delta\mu^* \left(\phi \left(III_C^{-\frac{1}{3}} \mathbf{1} - \frac{1}{3} I_C III_C^{-\frac{1}{3}} \mathbf{C}^{-1} \right) + (1 - \phi) \left(III_C^{-\frac{2}{3}} (I_C \mathbf{1} - \mathbf{C}) - \frac{2}{3} II_C III_C^{-\frac{2}{3}} \mathbf{C}^{-1} \right) \right). \tag{7}$$

By a direct transformation we have $\Delta_{\kappa^*} \boldsymbol{\sigma} \stackrel{\text{def}}{=} \boldsymbol{\sigma}(\kappa^* + \Delta\kappa^*, \mu^*) - \boldsymbol{\sigma}(\kappa^*, \mu^*) = \frac{1}{J} \mathbf{F} \cdot (\Delta\kappa^* (III_C - III_C^{\frac{1}{2}}) \mathbf{C}^{-1}) \cdot \mathbf{F}^T$, and $\Delta_{\mu^*} \boldsymbol{\sigma} \stackrel{\text{def}}{=} \boldsymbol{\sigma}(\kappa^*, \mu^* + \Delta\mu^*) - \boldsymbol{\sigma}(\kappa^*, \mu^*) = \frac{1}{J} \mathbf{F} \cdot \Delta_{\mu^*} \mathbf{S} \cdot \mathbf{F}^T$

$$\Delta_{\mu^*} \boldsymbol{\sigma} = \frac{1}{J} \mathbf{F} \cdot \Delta_{\mu^*} \left(\phi \left(III_C^{-\frac{1}{3}} \mathbf{1} - \frac{1}{3} I_C III_C^{-\frac{1}{3}} \mathbf{C}^{-1} \right) + (1 - \phi) \left(III_C^{-\frac{2}{3}} (I_C \mathbf{1} - \mathbf{C}) - \frac{2}{3} II_C III_C^{-\frac{2}{3}} \mathbf{C}^{-1} \right) \right) \cdot \mathbf{F}^T. \tag{8}$$

Unfortunately, no general rigorous bounds, as for the energy, are available on the components of the perturbed stresses.

REFERENCES

1. Ball, J. M. (1977). Convexity conditions and existence theorems in nonlinear elasticity. *Arch. Rational Mech. Anal.* **63**, 337-403.
2. Ciarlet, P. G. (1993). *Mathematical elasticity*. Elsevier.
3. Ogden, R. W. (1999). *Nonlinear elasticity*. Dover reissue.
4. Hill, R. (1952). The Elastic Behaviour of a Crystalline Aggregate. *Proc. Phys. Soc. (Lond.)* **A65**, 349-354.
5. Voigt, W. (1889). Über die Beziehung zwischen den beiden Elastizitätskonstanten isotroper Körper. *Wied. Ann.* **38**, 573-587.
6. Reuss, A. (1929). Berechnung der Fließgrenze von Mischkristallen auf Grund der Plastizitätsbedingung für Einkristalle. *Z. angew. Math. Mech.* **9**, 49-58.
7. Hashin, Z. and Shtrikman, S. (1962). On some variational principles in anisotropic and nonhomogeneous elasticity. *Journal of the Mechanics and Physics of Solids.* **10**, 335-342.
8. Hashin, Z. and Shtrikman, S. (1963). A variational approach to the theory of the elastic behaviour of multiphase materials. *Journal of the Mechanics and Physics of Solids.* **11**, 127-140.

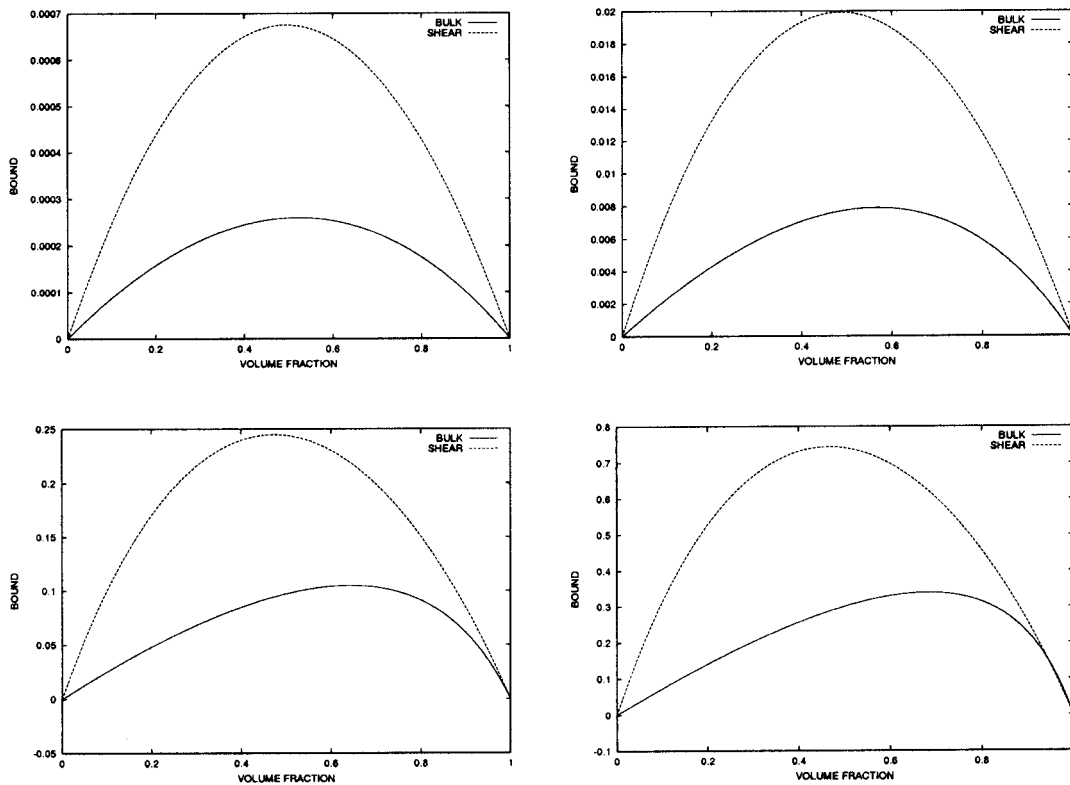


Figure 1: The behavior of the bounds as a function of volume fraction. Clockwise, starting from the top left: $\frac{\kappa_2}{\kappa_1} = \frac{\mu_2}{\mu_1} = 1.25$, $\frac{\kappa_2}{\kappa_1} = \frac{\mu_2}{\mu_1} = 2$, $\frac{\kappa_2}{\kappa_1} = \frac{\mu_2}{\mu_1} = 5$ and $\frac{\kappa_2}{\kappa_1} = \frac{\mu_2}{\mu_1} = 10$.