Nonlinear Electroelastic Deformations of Dielectric Elastomer Composites

Oscar Lopez-Pamies

In collaboration with: Victor Lefèvre

Work supported by the National Science Foundation (DMS, CMMI)
Dielectric elastomers

Energy Harvesters

Actuators

Examples: (pretty much) any elastomer

Disadvantages: finitely deformable — short response times — light — inexpensive — biocompatible

Disadvantage: require very large electric fields for actuation (in the order of 100 MV/m)

Idea: add high-dielectric (or conducting) particles to make dielectric composites that can finitely deform under moderate electric fields

SRI (2009); Kofod et al. (2007); Carpi et al. (2010); Aschwanden & Stemmer (2006)
**Dielectric elastomer composites**

Matrix: PU  
- dielectric constant: 10  
- shear modulus: 10 MPa

Particles: PAA decorated o-CuPc  
- dielectric constant: $10^4$  
- shear modulus: 1 GPa

Microstructure: isotropic distribution of roughly spherical nano-particles at 7% vol. fraction

**Competition of effects:** adding high-dielectric particles increases the overall dielectric constant of the composite, but it may also make it stiffer, and hence less deformable

- Initial conjecture in the literature (Li, 2003; Tian et al., 2012): **Enhancement is due to the heightened role of field fluctuations due to the nonlinearity of elastomers**

- Alternative conjecture (Lewis, 2004; Lopez-Pamies et al., 2014): **Enhancement is due to interphasial phenomena including free charges**
Problem Formulation
(elastic dielectrics)
Problem setting: 2-phase particulate material

Kinematics

Undeformed $\Omega$

Deformed $\Omega^*$

$W_m(F, E)$

$W_p(F, E)$

$x = \chi(X)$

Total nominal stress

$S = \frac{\partial W}{\partial F}(X, F, E)$

Lagrangian electric displacement

$D = -\frac{\partial W}{\partial E}(X, F, E)$

Deformation Gradient

$F = \frac{\partial x}{\partial X}$

Lagrangian Electric Field $E$

Local constitutive behavior

where

$W(X, F, E) = [1 - \theta(X)]W_m(F, E) + \theta(X)W_p(F, E)$

Indicator function characterizing the initial microstructure

$\theta(X) = \begin{cases} 
1 & \text{if } X \in \text{particle} \\
0 & \text{otherwise}
\end{cases}$
Problem setting: macroscopic response

Definition: relation between the volume averages of $S$, $D$ and $F$, $E$

Macroscopic deformation gradient & electric field

$$
\overline{F} = \frac{1}{|\Omega|} \int_{\Omega} F \, dX \\
\overline{E} = \frac{1}{|\Omega|} \int_{\Omega} E \, dX
$$

Macroscopic nominal stress & electric displacement

$$
\overline{\dot{S}} = \frac{1}{|\Omega|} \int_{\Omega} S \, dX \\
\overline{\dot{D}} = \frac{1}{|\Omega|} \int_{\Omega} D \, dX
$$

Variational Characterization

$$
\overline{S} = \frac{\partial \overline{W}}{\partial \overline{F}} (\overline{F}, \overline{E}, c) \\
\overline{D} = -\frac{\partial \overline{W}}{\partial \overline{E}} (\overline{F}, \overline{E}, c)
$$

$$
\overline{W} (\overline{F}, \overline{E}, c) = \min_{\overline{F} \in \mathcal{K}} \max_{\overline{E} \in \mathcal{E}} \int_{\Omega} W (X, F, E) \, dX
$$

Hill (1972)  Lopez-Pamies (2014)
Problem setting: macroscopic response

**Definition:** relation between the volume averages of $S$, $D$ and $F$, $E$

**Macroscopic deformation gradient & electric field**

$$\bar{F} = \frac{1}{|\Omega|} \int_{\Omega} F \, dX$$

$$\bar{E} = \frac{1}{|\Omega|} \int_{\Omega} E \, dX$$

**Macroscopic nominal stress & electric displacement**

$$\bar{S} = \frac{1}{|\Omega|} \int_{\Omega} S \, dX$$

$$\bar{D} = \frac{1}{|\Omega|} \int_{\Omega} D \, dX$$

**Variational Characterization**

$$\bar{W}(\bar{F}, \bar{E}, c) = \min_{\bar{F} \in \mathcal{K}} \max_{\bar{E} \in \mathcal{E}} \int_{\Omega} W(X, F, E) \, dX$$

**Euler-Lagrange equations**

$$\text{Div } S = 0 \quad \text{and} \quad \text{Div } D = 0$$

**BC’s**

$$\Phi = -\bar{E} \cdot X$$

$$x = \bar{F}X$$
Two strategies to generate exact solutions

I. **Iterated homogenization** (Lopez-Pamies, 2014): for an *infinitely poly-disperse* particulate microstructure \( W = W(\mathbf{F}, \mathbf{E}, c) \) is given implicitly by the *nonlinear* first-order Hamilton-Jacobi pde

\[
\frac{c}{c} \frac{\partial W}{\partial c} - W - \left\langle \max_{\alpha} \min_{\beta} \left[ \alpha \cdot \frac{\partial W}{\partial \mathbf{F}} \cdot \mathbf{\xi} + \beta \frac{\partial W}{\partial \mathbf{E}} \cdot \mathbf{\xi} - W_m (\mathbf{F} + \alpha \mathbf{\xi}, \mathbf{E} + \beta \mathbf{\xi}) \right] \right\rangle = 0
\]

subject to the initial condition \( W(\mathbf{F}, \mathbf{E}, 1) = W_p(\mathbf{F}, \mathbf{E}) \)

II. **Hybrid finite elements** (Lefèvre & Lopez-Pamies, 2016): periodic homogenization of an infinite medium made out of the repetition of a unit cell containing a random distribution of a *finite number* of particles

\[
W(\mathbf{F}, \mathbf{E}, c) = \min_{\mathbf{u} \in \mathcal{U}} \max_{\Phi \in \mathcal{F}} \max_{p \in \mathcal{P}} \int_Y \left\{ p [\det \mathbf{F(u)} - 1] - \hat{W}^*(\mathbf{X}, \mathbf{F(u)}, p, \mathbf{E(\Phi)}) \right\} \, d\mathbf{X}
\]
Solutions for

*Ideal Elastic Dielectrics*

with

*Isotropic Microstructures*
Isotropic ideal elastic dielectric composites

\[ W_m(F, E) = \begin{cases} \frac{\mu}{2} [I_1 - 3] - \frac{\varepsilon}{2} I_5^E & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \]

\[ W_p(F, E) = \begin{cases} \frac{\mu_p}{2} [I_1 - 3] - \frac{\varepsilon_p}{2} I_5^E & \text{if } J = 1 \\ +\infty & \text{otherwise} \end{cases} \]

Here

\[ I_1 = F \cdot F \]
\[ J = \det F \]
\[ I_5^E = F^{-T} E \cdot F^{-T} E \]

- In view of the overall (constitutive and geometric) isotropy and incompressibility of these composites, their effective free-energy function is of the form

\[ \overline{W}(\overline{F}, \overline{E}, c) = \begin{cases} \overline{W}(\overline{I}_1, \overline{I}_2, \overline{I}_4, \overline{I}_5, \overline{I}_6, c) & \text{if } \overline{J} = 1 \\ +\infty & \text{otherwise} \end{cases} \]

with

\[ \overline{I}_1 = \overline{F} \cdot \overline{F} \]
\[ \overline{I}_4 = \overline{E} \cdot \overline{E} \]
\[ \overline{I}_2 = \overline{F}^{-T} \cdot \overline{F}^{-T} \]
\[ \overline{I}_5^E = \overline{F}^{-T} \overline{E} \cdot \overline{F}^{-T} \overline{E} \]
\[ \overline{J} = \det \overline{F} \]
\[ \overline{I}_6^E = \overline{F}^{-1} \overline{F}^{-T} \overline{E} \cdot \overline{F}^{-1} \overline{F}^{-T} \overline{E} \]
Solution from iterated homogenization

Let us rewrite the pde for $\overline{W} = \overline{W}(\overline{F}, \overline{E}, c)$ in the classical form of H-J equations

$$\frac{\partial \overline{W}}{\partial c} + \mathcal{H} \left( \overline{F}, \overline{E}, c, \overline{W}, \frac{\partial \overline{W}}{\partial \overline{F}}, \frac{\partial \overline{W}}{\partial \overline{E}} \right) = 0, \quad \overline{W}(\overline{F}, \overline{E}, 1) = W_p(\overline{F}, \overline{E})$$

with Hamiltonian

$$\mathcal{H} = -\frac{1}{c} \overline{W} - \frac{1}{c} \left\langle \max_{\alpha} \min_{\beta} \left[ \alpha \cdot \frac{\partial \overline{W}}{\partial \overline{F}} \xi + \beta \frac{\partial \overline{W}}{\partial \overline{E}} \cdot \xi - W_m(\overline{F} + \alpha \otimes \xi, \overline{E} + \beta \xi) \right] \right\rangle$$

- H-J equations typically exhibit non-uniqueness of non-smooth (Lipschitz cont.) solutions
- The physically correct solution is the so-called viscosity solution
- Numerical scheme

  "space" discretization over a Cartesian grid making use of a monotone scheme and WENO finite difference of fifth order

  explicit "time" integration of fifth order

Crandall & Lions (1983, 1984)  
Osher & Sethian (1988); Jiang & Shu (1996)
IH sample results: \( c = 0.05, \quad \mu_p = 10^2 \mu, \quad \varepsilon_p = 10^2 \varepsilon \)
IH sample results: $c = 0.05$, $\mu_p = 10^2 \mu$, $\varepsilon_p = 10^2 \varepsilon$

Linear dependence on $\bar{I}_1, \bar{I}_4, \bar{I}_5$

independence of $\bar{I}_2, \bar{I}_6$

for all $c$, $\mu$, $\varepsilon$, $\mu_p$, $\varepsilon_p$ and all $\bar{F}$, $\bar{E}$
Solutions via a hybrid FE formulation

With help of the partial Legendre transform

\[ \hat{W}^*(X, F, p, E) = \max_J \left\{ p(J - 1) - \hat{W}(X, F, J, E) \right\} \]

the effective free-energy function can be written as

\[ \overline{W}(\bar{F}, \bar{E}, c) = \min_{u \in U} \max_{\Phi \in \Phi} \max_{p \in P} \int_Y \left\{ p[\det F(u) - 1] - \hat{W}^*(X, F(u), p, E(\Phi)) \right\} dX \]

for \( Y \)-periodic microstructures

\[ u(X) = (\bar{F} - I)X + \tilde{u}(X), \text{ where } \tilde{u} \text{ is } Y-\text{periodic, } \tilde{u}(0) = 0. \]
\[ \Phi(X) = -E \cdot X + \tilde{\Phi}(X), \text{ where } \tilde{\Phi} \text{ is } Y-\text{periodic, } \tilde{\Phi}(0) = 0, \]
\[ p \text{ is } Y-\text{periodic} \]

We look for FE solutions making use of conforming Crouzeix-Raviart-type elements — stable and convergent!
Sample “isotropic” microstructures and meshes

**Example:** isotropic distribution of 5% vol. fraction of three families of spherical particles

**Example:** isotropic distribution of 5% vol. fraction of monodisperse spherical particles
**FE sample results:** $c = 0.05$, $\mu_p = 10^2 \mu$, $\varepsilon_p = 10^2 \varepsilon$

Linear dependence on $\bar{I}_1, \bar{I}_4, \bar{I}_5$ independence of $\bar{I}_2, \bar{I}_6$ for all $c$, $\mu$, $\varepsilon$, $\mu_p$, $\varepsilon_p$ and all $\bar{F}$, $\bar{E}$
An approximate closed-form solution

\[
\bar{W}(\bar{F}, \bar{E}, c) = \begin{cases} 
\frac{\tilde{\mu}}{2} [\bar{I}_1 - 3] + \frac{\tilde{m}_K - \tilde{\varepsilon}}{2} \bar{I}_4^E - \frac{\tilde{m}_K}{2} \bar{I}_5^E & \text{if } \bar{J} = 1 \\
+\infty & \text{otherwise}
\end{cases}
\]

Here, \(\tilde{\mu}, \tilde{\varepsilon}, \tilde{m}_K\) denote the effective shear modulus, permittivity, and electrostriction constant in the \textit{limit of small deformations and moderate electric fields}.

\[
\tilde{\mu} = \int_{\Omega} 2\mu(X)K_{12rs}\Gamma_{r12,s}dX
\]
\[
\tilde{\varepsilon} = \int_{\Omega} \frac{1}{3}\varepsilon(X)\gamma_{m,m}dX
\]
\[
\tilde{m}_K = \int_{\Omega} 2\varepsilon(X)\Gamma_{r12,s}K_{rspq}\gamma_{p,1}\gamma_{q,2}dX
\]

where \(\Gamma\) and \(\gamma\) are solutions of the \textit{one-way coupled linear pdes}

\[
\begin{cases}
\left[\mu(X)K_{ijmn}\Gamma_{mkl,n} + \frac{1}{2}\delta_{ij}q_{kl}\right]_{,j} = 0 & \text{for } X \in \Omega, \quad \Gamma_{ikl} = \delta_{ik}X_l & \text{for } X \in \partial\Omega \\
\Gamma_{mkl,m} = 0
\end{cases}
\]

and
\[
[\varepsilon(X)\gamma_{i,j}]_{,i} = 0 & \text{for } X \in \Omega, \quad \gamma_{i} = X_i & \text{for } X \in \partial\Omega.
\]
Solutions for Non-Gaussian Elastomers Isotropically Filled with Non-Linear Elastic Dielectric Particles
Non-Gaussian elastomers; non-linear particles

\[ W_m(F, E) = \begin{cases} 
\psi(I_1) - \frac{\varepsilon}{2} I_5^E & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases} \]

\[ W_p(F, E) = \begin{cases} 
\frac{\mu_p}{2}[I_1 - 3] - S(I_5^E) & \text{if } J = 1 \\
+\infty & \text{otherwise}
\end{cases} \]

Here:
\[ I_1 = F \cdot F \\
J = \det F \\
I_5^E = F^{-T}E \cdot F^{-T}E \]

Examples:
(Lopez-Pamies, 2010)
\[ \psi(I_1) = \frac{3^{1-\alpha_1}}{2\alpha_1} \mu_1 [I_1^{\alpha_1} - 3^{\alpha_1}] + \frac{3^{1-\alpha_2}}{2\alpha_2} \mu_2 [I_1^{\alpha_2} - 3^{\alpha_2}] \]

Langevin (1904); Debye (1929)
\[ S(I_5^E) = \frac{\varepsilon_0}{2} I_5^E + \frac{p_s^2}{3(\varepsilon_p - \varepsilon_0)} \left[ \ln \left( \sinh \left( \frac{3(\varepsilon_p - \varepsilon_0) \sqrt{I_5^E}}{p_s} \right) \right) - \ln \left( \frac{3(\varepsilon_p - \varepsilon_0) \sqrt{I_5^E}}{p_s} \right) \right] \]

- **Numerically**: exact solutions can be generated via the HJ equation & the hybrid FE formulation
- **Analytically**: approximate solutions can be generated via a nonlinear comparison medium method
Comparison with electrostriction experiments

Matrix: PU
- Dielectric constant: 10
- Shear modulus: 10 MPa

Particles: PAA decorated o-CuPc
- Dielectric constant: $10^4$
- Shear modulus: 1 GPa

Microstructure: isotropic distribution of roughly spherical nano-particles at 7% vol. fraction

Graph:
- Matrix (Experiment)
- Composite (Experiment)
- Matrix (Theory)
- Composite (Sph. Theory)
- Composite (Sph. FE)

Huang et al. (2005); Lefèvre & Lopez-Pamies (2016)
Interphasial phenomena?

Experiment

$Lefèvre & Lopez$-
$Pamies (2016)$