Fine tuning the electro-mechanical response of dielectric elastomers

Giuseppe Zurlo,[1,a)] Michel Destrade,[1,b)] and Tongqing Lu[2,c)]
[1]*School of Mathematics, Statistics and Applied Mathematics, NUI Galway, University Road, Galway, Ireland*
[2]*State Key Lab for Strength and Vibration of Mechanical Structures, Department of Engineering Mechanics, Xi'an Jiaotong University, Xi'an 710049, China*

(Received 24 August 2018; accepted 2 October 2018; published online 16 October 2018)

We propose a protocol to model accurately the electromechanical behavior of dielectric elastomer membranes using experimental data of stress-stretch and voltage-stretch tests. We show how the relationship between electric displacement and the electric field can be established in a rational manner from these data. Our approach demonstrates that the *ideal dielectric model*, prescribing linearity in the purely electric constitutive equation, is quite accurate at low-to-moderate values of the electric field and that, in this range, the dielectric permittivity constant of the material can be deduced from stress-stretch and voltage-stretch data. Beyond the linearity range, more refined couplings are required, possibly including a non-additive decomposition of the electro-elastic energy. We also highlight that the presence of vertical asymptotes in voltage-stretch data, often observed in the experiments just prior to failure, should not be associated with strain stiffening effects but instead with the rapid development of electrical breakdown. *Published by AIP Publishing.*
https://doi.org/10.1063/1.5053643

A major challenge in the modelling of soft dielectric membranes with compliant electrodes comes from attempting to deduce by experimental tuning the total energy density function $\Omega(\mathbf{F},\boldsymbol{E})$, which depends on the deformation gradient $\mathbf{F}$ and on the Lagrangian electric field $\boldsymbol{E}$. An accurate knowledge of Ω is essential to formulate correctly the equations of electromechanical equilibrium, giving the Piola-Kirchhoff stress as $\mathbf{S} = \partial_{\mathbf{F}}\Omega$ and the Lagrangian electric displacement vector as $\boldsymbol{D} = -\partial_{\boldsymbol{E}}\Omega$. Then, the total Cauchy stress is as follows: $\boldsymbol{\sigma} = J^{-1}\mathbf{S}\mathbf{F}^{\top}$ with $J = \det\mathbf{F}$, and the Eulerian electric field and displacement vector are as follows: $\boldsymbol{e} = \mathbf{F}^{-\top}\boldsymbol{E}$ and $\boldsymbol{d} = \mathbf{F}\boldsymbol{D}$, respectively.

For isotropic and incompressible dielectrics, the electro-elastic energy depends on $(\mathbf{F},\boldsymbol{E})$ through five scalar invariants,[1] for instance: $I_1 = \mathrm{tr}\mathbf{C}$, $I_2 = \mathrm{tr}\mathbf{C}^{-1}$, $I_4 = \boldsymbol{E}\cdot\boldsymbol{E}$, $I_5 = \mathbf{C}^{-1}\boldsymbol{E}\cdot\boldsymbol{E}$, $I_6 = \mathbf{C}^{-1}\boldsymbol{E}\cdot\mathbf{C}^{-1}\boldsymbol{E}$, where $\mathbf{C} = \mathbf{F}^{\top}\mathbf{F}$ is the right Cauchy-Green tensor. Noting that $I_5 = ||\boldsymbol{e}||^2$, a fairly general and common assumption made in the literature is that the electro-elastic energy is split additively into

$$\Omega = W(I_1, I_2) - \psi\left(\sqrt{I_5}\right), \tag{1}$$

where W is a purely elastic term and ψ is an electro-elastic term. This expression encompasses, for example, the so-called *ideal dielectric model*[2] with $\psi(x) = \varepsilon x^2/2$, where ε is the dielectric constant, and the *polarization saturation model*[3] with $\psi(x) = (d_s^2/\varepsilon)\ln(\cosh(\varepsilon x/d_s))$, where d_s is the saturation value of the Eulerian electric displacement.

In this paper, we leave ψ unspecified and show that this function can be completely determined from independent sets of stress-stretch and voltage-stretch experimental data.

Assume that the dielectric membrane is a plate of uniform reference thickness H, with compliant electrodes on its upper and lower faces. For equi-biaxially deformed states, the deformation gradient is $\mathbf{F} = \mathrm{diag}(\lambda, \lambda, \lambda^{-2})$, where λ is the in-plane stretch, the Lagrangian electric field is $\boldsymbol{E} = (0,0,E)$, and the displacement vector is $\boldsymbol{D} = (0,0,D)$, where the non-vanishing components are along the direction perpendicular to the membrane mid-surface. Note that $E = V/H$, where V is the controlled voltage and that $e = \lambda^2 E = \sqrt{I_5}$ and $d = \lambda^{-2}D$.

Specialising the energy (1) to equi-biaxial states, we introduce $\omega(\lambda, E) = w(\lambda) - \psi(\lambda^2 E)$, where $w(\lambda) = W(2\lambda^2 + \lambda^{-4}, 2\lambda^{-2} + \lambda^4)$ for the purely elastic part. Electro-mechanical equilibrium in the presence of a pre-stretch λ_p reads $\partial_\lambda\omega(\lambda, E) = \partial_\lambda\omega(\lambda_p, 0)$ or

$$s(\lambda) - \lambda E\psi'(\lambda^2 E) = s(\lambda_p), \tag{2}$$

where $s(\lambda) = w'(\lambda)/2$ is the purely mechanical in-plane stress [and $s(\lambda_p)$ is the mechanical stress required to pre-stretch the plate]. Furthermore, the electric displacement reads $D = -\partial_E\omega = \lambda^2\psi'(E\lambda^2)$, which can be conveniently recast in the Eulerian form as

$$d = \psi'(e). \tag{3}$$

Remarkably, Eqs. (2) and (3) show that if the purely mechanical response of the polymer is known through the function $s(\lambda)$, then the relationship between d and e can be determined by an independent set of voltage-stretch data. More precisely, once various sets of experimental data $\{\lambda_p, \lambda, E\}$ are recorded, then the resulting curve $d(e)$ can be determined by interpolating the points

$$d = \frac{s(\lambda) - s(\lambda_p)}{\lambda E}, \quad e = \lambda^2 E. \tag{4}$$

We emphasise that because d depends only on e according to (3), the purely electrical constitutive equation in the Eulerian form must be independent of the specific values of λ, λ_p, and

a)giuseppe.zurlo@nuigalway.ie
b)michel.destrade@nuigalway.ie
c)tongqinglu@xjtu.edu.cn

E. If the data do not show this independence, then the assumption of additivity (1) must be abandoned.

The first step towards the derivation of the purely electric constitutive equation through (4) is to determine the mechanical response of the polymer. Here, we focus on the popular dielectric acrylic elastomer VHB4905 (3M Center, St. Paul, MN). This is a viscoelastic material, but here, we focus on rate-independent effects.

To provide a robust tuning of the material response, we report two sets of data, see Fig. 1. The first set comes from direct equi-biaxial tensile tests carried out in our Soft Machine Laboratory (Xi'an Jiaotong University): we used disks with initial radius $R = 17.5\,\text{mm}$ and thickness $H = 0.5\,\text{mm}$, with a loading rate of 2 mm/min. The second set of data is extracted from the paper by Huang *et al.*[4] (same R and H with a loading rate of 11.0 g/90 s, which was the slowest rate reported).

We notice that the material breaks before it can exhibit the strain-stiffening effect due to limiting chain extensibility. It is thus not appropriate to use a Gent or an Arruda-Boyce model[5] here, because their stiffening parameter cannot be determined properly. Instead, we find that a good fit is obtained with the following strain energy density (based on the I_1—model of Lopez-Pamies[6]):

$$W(I_1, I_2) = \frac{c_1}{2}(I_1^\alpha - 3^\alpha) + \frac{c_2}{2}(I_2^\beta - 3^\beta), \tag{5}$$

with $c_1 = 75\,400\,\text{Pa}$, $c_2 = 0.23\,\text{Pa}$, $\alpha = 0.87$, and $\beta = 2.2$, see the corresponding graph of $s(\lambda)$ in Fig. 1. The initial shear modulus for this model is computed as $\mu = 56\,869\,\text{Pa}$ from the formula $\mu := 2(\partial W/\partial I_1 + \partial W/\partial I_2)$ at $I_1 = I_2 = 3$.

We now move on to modelling the electromechanical behavior. We consider that the (E, λ) data from equi-biaxial voltage-stretch experiments conducted on VHB4905 disks

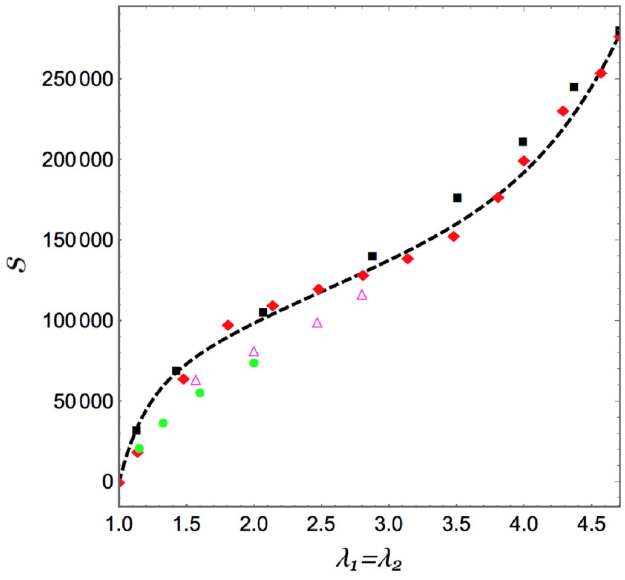

FIG. 1. Mechanical tuning of 3M VHB4905 in equibiaxial tension. Black squares taken from Ref. 4, and red diamonds refer to our own experiment. For information, we also give the stress-stretch pairs deduced from the voltage-stretch experiments of Ref. 7 (green circles) and Ref. 4 (magenta triangles) when the voltage is zero. The dashed curve indicates the best-fit function $s(\lambda)$.

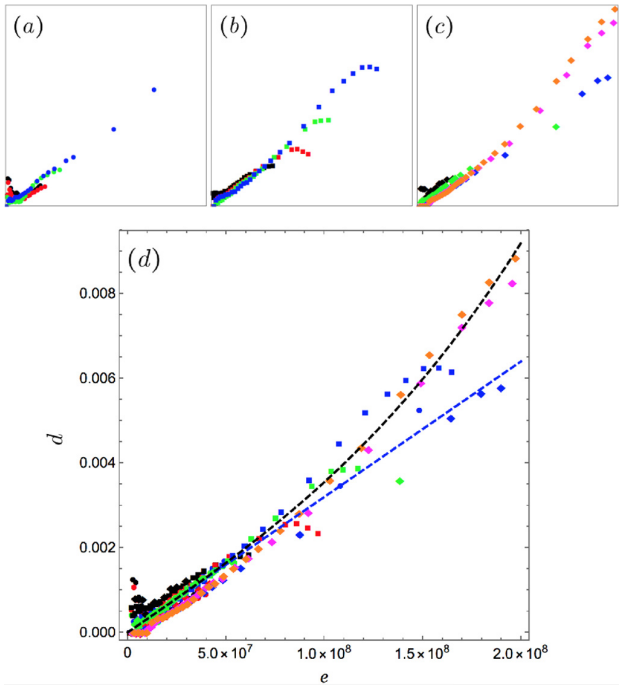

FIG. 2. Pairs (d, e) in (C/m^2, V/m) deduced from Eq. (4) once the mechanical response $s(\lambda)$ has been determined. The pairs characterise the purely electrical response of the polymer for three sets of experiments. Circle points in (a) are obtained from Ref. 7, where the colors black, red, blue, and green correspond to the pre-stretches $\lambda_p = 1.2$, 1.3, 1.6, and 2, respectively. Square points in (b) are obtained from Ref. 4, where the colors black, red, blue, and green correspond to the pre-stretches $\lambda_p = 1.6$, 2, 2.5, and 2.8, respectively. Diamond points in (c) are obtained from Ref. 8, where the colors black, red, blue, green, magenta, and orange correspond to the pre-stretches $\lambda_p = 1.2$, 1.6, 1.8, 2, 2.5, and 2.9, respectively. The points are all aggregated in the main picture (d), where the blue and black dashed curves represent linear and cubic interpolations of the data, respectively.

with 14 different levels of pre-stretch, taken from the articles.[4,7,8] Figure 2 shows the distributions of the pairs (d, e) deduced from (4) [now that $s(\lambda)$ is known].

The insets (a), (b), and (c) refer to Refs. 4, 7, and 8, respectively. The (d, e) pairs show a late "electric softening" behavior, in the sense that the d–e plots have a decreasing slope just prior to the electrical breakdown. In some cases, (b) in particular, breakdown occurs after the recording of a peak; in (a) and (c), it occurs before a peak can be recorded (this could be explained by how fast the final expansion occurs, and how difficult it is to control it and to measure its progression with increasing dead-loads).

The compound picture in Fig. 2 further suggests that the softening branches of the 14 sets of (d, e) pairs may be seen as *outliers* with respect to a collective "safe" behavior of the polymer. In particular, in the $0 \le e \le 10^8\,\text{V m}^{-1}$ window, this safe branch is fitted by the linear interpolation

$$d_{\text{lin}} = \varepsilon e, \quad \varepsilon = 3.2 \times 10^{-11}\ \text{Fm}^{-1}. \tag{6}$$

We thus conclude that, in this range, *VHB4905 behaves as an ideal dielectric*. Note that the value of the deduced dielectric constant is on the same order of magnitude but lower than the usual value $\approx 3.8 \times 10^{-11}\,\text{Fm}^{-1}$ proposed in the literature for this material, see, e.g., Refs. 4, 7, and 8.

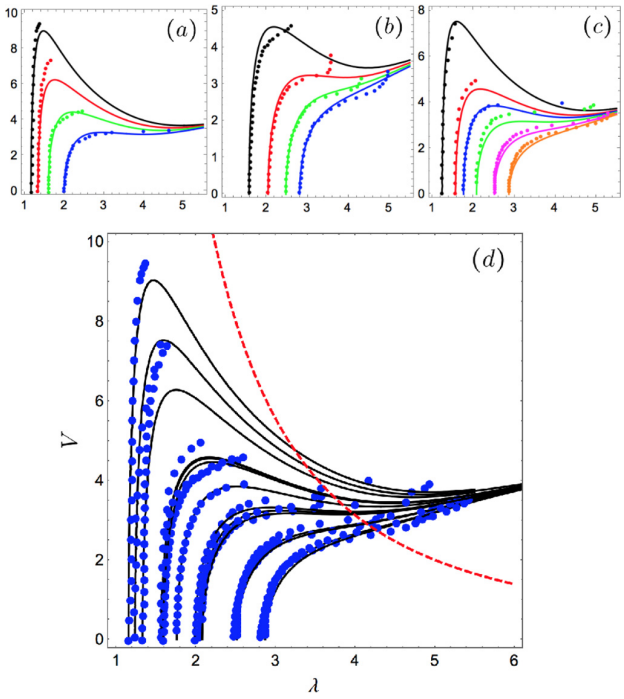

FIG. 3. Experimental data (dots) vs theory (solid curves) for all the pre-stretch levels taken into consideration in this manuscript. The three insets (*a*), (*b*), and (*c*) refer to the three sets of experiments,[4,7,8] respectively. The compound picture is reported in (*d*). Voltage is measured in kV. Solid curves are obtained by using the mechanical response (5) together with the cubic electrical response (7). The red dashed curve, corresponding to $e = 10^8\,\mathrm{V\,m^{-1}}$, represents the limit of applicability of the ideal dielectric model for VHB4905.

The ideal dielectric paradigm is widely used in the literature on dielectric elastomers, but its direct proof from actuation experiments is quite rare. Here, we used a rational approach to deduce its validity indirectly.

Beyond that range, the cubic approximation

$$d_{\mathrm{cub}} = \varepsilon e + a e^3, \tag{7}$$

with ε as above and $a = 3.5 \times 10^{-28}\,\mathrm{Fm^2\,V^{-3}}$ fits the safe branch better; it gives $\psi = \varepsilon I_5/2 + a I_5^2/4$ for the electro-elastic part of the free energy. Of course, it is debatable whether the cubic is able to capture the scatter observed in Fig. 2 at the higher values of the Eulerian electric field e. As we do not have enough data to conduct a statistical analysis, we leave this question open.

An alternative point of view is to conclude that the scatter is too large and that a more sophisticated form of free energy than in Eq. (1) must be adopted. We tried to replace ψ with linear combinations of I_4, I_5, and I_6 but they also lead to d being a function of e only, which excludes scatter in the d–e plots. Possibly, the additive split of a function of I_1 and I_2 only and a function of I_4, I_5, and I_6 only has to be abandoned to model the electro-mechanical behavior of VHB4905 over the entire range of stretches and voltages up to breakdown.

Electric softening occurs at high values of the electric field but without any matching behavior (such as strain-softening/stiffening) in the purely mechanical response of the polymer. On the other hand, we discard the possibility that—at least for this material—electric softening can be explained in terms of "polarization saturation" in the electrical response of the polymer,[3] because none of the d–e curves exhibit saturation. Instead, we propose that electric softening is simply a global manifestation of a local failure process. For this reason, this effect should not be regarded (nor modelled) as a constitutive feature of the material.

We finally show that the voltage-stretch response of the polymer can be described and modelled by using the purely mechanical response function $s(\lambda)$ and the purely electrical response (7). The resulting theoretical curves show an excellent prediction of the experimental data in a wide range of pre-stretches. Figure 3 shows the overall data of the 14 levels of prestretch from the three different articles,[4,7,8] together with our theoretical predictions.

The work of T.L. was supported by the NSFC (No. 11772249). G.Z. gratefully acknowledges the hospitality of the PMMH-ESPCI ParisTech and the support of the Italian Gruppo Nazionale di Fisica Matematica (GNFM). The authors thank the organisers of the 2016 EMI International Conference of ASCE in Metz, France, where preliminary ideas for this paper were discussed. They also thank Ruisen Yang for providing some of the experimental data in Fig. 1.

[1]L. Dorfmann and R. W. Ogden, "Nonlinear electroelasticity," Acta Mech. **174**, 167–183 (2005).
[2]X. H. Zhao and Z. G. Suo, "Method to analyze electromechanical stability of dielectric elastomers," Appl. Phys. Lett. **91**, 061921 (2007).
[3]B. Li, L. Liu, and Z. Suo, "Extension limit, polarization saturation, and snap-through instability of dielectric elastomers," Int. J. Smart Nano Mater. **2**, 59–67 (2011).
[4]J. Huang, T. Li, C. C. Foo, J. Zhu, D. R. Clarke, and Z. Suo, "Giant, voltage-actuated deformation of a dielectric elastomer under dead load," Appl. Phys. Lett. **100**, 041911 (2012).
[5]C. O. Horgan and G. Saccomandi, "A molecular-statistical basis for the Gent constitutive model of rubber elasticity," J. Elasticity **68**, 167–176 (2002).
[6]O. Lopez-Pamies, "A new I_1–based hyperelastic model for rubber elastic materials," C. R. Mec. **338**, 3–11 (2010).
[7]T. Lu, S. Cheng, T. Li, T. Wang, and Z. Suo, "Electromechanical catastrophe," Int. J. Appl. Mech. **8**, 1640005 (2016).
[8]T. Lu, J. Huang, C. Jordi, G. Kovacs, R. Huang, D. R. Clarke, and Z. Suo, "Dielectric elastomer actuators under equal-biaxial forces, uniaxial forces, and uniaxial constraint of stiff fibers," Soft Matter **8**, 6167 (2012).