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Catastrophic thinning of dielectric elastomers

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We provide an energetic insight into the catastrophic nature of thinning instability in soft electroactive elastomers. This phenomenon is a major obstacle to the development of giant actuators; yet it is not completely understood nor modelled accurately. In excellent agreement with experiments, we give a simple formula to predict the critical voltages for instability patterns; we model their shape and show that reversible (elastic) equilibrium is impossible beyond their onset. Our derivation is fully analytical, does not require finite element simulations and can be extended to include pre-stretch and various material models.

Consider a thin dielectric plate with conducting faces: when will it break if a voltage is applied? If it is rigid it will break once its dielectric strength is overcome by the voltage. But what if it is highly stretchable, like the elastomers used for soft actuators, stretchable electronics, or energy harvesters? The precise answer to that question is not known. Experiments show that it will break when highly localised thinning deformations occur, and further, that this process is catastrophic, as the deformations cannot be controlled nor restrained once they have started.

Here we unveil the physical meaning of catastrophic thinning, based on energy minimisation arguments; we derive a unifying and simple formula giving very accurate predictions of the voltage thresholds for the creasing instability (one-side constrained plates, with one compliant electrode on one face and a rigid electrode on the other face) and the pull-in instability (unconstrained plates with fully compliant electrodes glued on both faces); we calculate the shape of the instability patterns at their onset and show that they are not sustainable but give the path to final breakdown; we generalise the results to include pre-stretch.

Typically thin dielectric elastomers are highly deformable (isotropic, incompressible) polymeric or silicone electroelastic films, brushed with conductive carbon grease [1]. With an applied electric field the attractive Coulomb forces between the electrodes compress the thickness of the film, which expands in its plane. At first the rectangular film deforms homogeneously into another rectangle, until a critical voltage is reached. Then, a sudden, irreversible and non-homogeneous thinning localisation occurs (usually accompanied by a spark and a popping sound [2]), anticipating the dielectric breakdown of the film, forming holes and significantly reducing its capacitance (Fig.1).

Call $E$ the following non-dimensional measure [4] of the electric field

$$E = \sqrt{\frac{\epsilon V}{\mu h}},$$

and $E_c$ its critical value. Here $V$ is the applied voltage, $\epsilon$ is the dielectric permittivity of the elastomer, $\mu$ its initial shear modulus and $h$ its initial thickness. For unconstrained plates, experiments [5] give $E_c$ in the range $0.678 - 0.686$ at the onset of pull-in instability, corresponding to a contraction of 30–34% in the layer’s thickness. For one-side constrained plates, experiments [6] reveal that $E_c \approx 0.85$. Finally, extensive experimental studies document that dead-loads (for unconstrained films) and pre-stretch (for one-side constrained films) play a beneficial role in delaying the onset of instabilities [7,8].

However, despite the abundance of experimental data,
so far the critical values of the electric field have not been predicted by the theory in an entirely satisfying manner. Many papers establish a connection between pull-in and snap-through instabilities for unconstrained films [4] using the so-called Hessian method, but when dead-loads are applied these predictions fail to account for the actual delay of instability, a key factor for technological applications [7]. Indeed, dead-loads can suppress snap-through instability but not the catastrophic thinning leading to failure. For unconstrained films there were attempts at introducing linearised [10] [13] and non-linear [14] inhomogeneous bifurcation modes on top of the homogeneous deformation, but they require lengthy calculations, do not explain the catastrophic nature of localised deformations and do not quantify the beneficial effects of pre-stretch. The situation is even worse for one-side constrained films. So far, electro-creasing was only studied with entirely numerical methods based on finite element method (FEM) simulations: in the absence of pre-stretch they lead to an estimate of $E_c \approx 1.03$, which is more than 20% off the experimental mark [6] of $E_c \approx 0.85$. There are no theoretical predictions available when pre-stretch is applied.

Our analysis [15] does not require the machinery of classical bifurcation methods. It provides a new paradigm for understanding electromechanical instability, which we find corresponds to a threshold where the electroelastic energy does not possess minimisers in a general class of homogeneous and non-homogeneous deformations. For both unconstrained and constrained films, with and without pre-stretch and for a quite general class of incompressible materials, we obtain the following simple unifying formula for the critical electric field:

$$E_c = \frac{2}{\sqrt{3}} \sqrt{\frac{W'(I)}{\mu}} \min\left(\frac{1}{\lambda_1}, \frac{1}{\lambda_2}\right)$$

(2)

where $\lambda_1$ and $\lambda_2$ are the principal stretches in the plane of the thin layer, $W$ is the elastic energy density and $I = \lambda_1^2 + \lambda_2^2 + (\lambda_1 \lambda_2)^{-2}$ is the first invariant of deformation (and hence, $\mu = 2W'(3)$). For unconstrained films, the principal stretches are determined by the homogeneous solution; for constrained films, they are the fixed pre-stretches imposed prior to attachment to the rigid substrate. Above $E_c$, no stable configurations exist, neither homogeneous nor non-homogeneous. As soon as $E_c$ is attained, failure precursors appear, opening the way for a catastrophic failure of the film. The initial pattern of these precursors is a permanent signature for the subsequent inelastic processes, see Figs. 2 [3].

To assess how formula (2) predicts the onset of catastrophic thinning we first consider one-side constrained films for the creasing instability, see sketch in the insert of Fig 4.

For small stretches ($\lambda_1, \lambda_2 \leq 1.5$) the constitutive response can be modelled as neo-Hookean: $W = \mu(I - 3)/2$, and formula (2) further simplifies to $E_c = \sqrt{2/3} \min(1/\lambda_1, 1/\lambda_2)$. Without pre-stretch ($\lambda_1 = \lambda_2 = 1$) this formula gives $E_c = \sqrt{2/3} = 0.816$, less than 4% off the value $E_c \approx 0.85$ obtained experimentally (Fig 3b) and closer than the estimate $E_c = 1.03$ obtained by FEM simulations (Fig 3b). For small values of uniaxial pre-stretch ($\lambda_1 = \lambda > 1$, $\lambda_2 = 1$), experiments [8] report an initial reduction of the critical electric field, in agreement with the prediction of formula (2) here, $E_c = \sqrt{2/3} \lambda^{-1}$. Beyond this initial negative effect, larger values of pre-stretch were measured as beneficial in increasing the critical electric field [8, 9], an effect that did not receive theoretical or numerical explanations so far. To demonstrate the ability of formula (2) in reproducing this effect, we use a material model $W$ which accounts for the strain-stiffening induced by the limit chain extensibility of the elastomer in large deformations. Fig. 4 shows the good agreement reached between our theory and experiments (see [15] for the calibration of the model).

We then consider unconstrained films to study the onset of the pull-in instability, see sketch in the insert of Fig 5. For equi-biaxial stretched films ($\lambda_1 = \lambda_2 = \lambda$) in the absence of dead-loads, the homogeneous extension of the neo-Hookean elastomer is described by $E = \sqrt{2/3} \lambda^{-4}$ (4), see also [15]. It reaches $E_c(\lambda) = \sqrt{2/3} \lambda^{-3}$ for $\lambda = 3^{1/6}$, where $E_c = \sqrt{2/3} \lambda^{-3} = 0.680$, falling square within the range of the experimental val-

![FIG. 2. Left: Experimental creasing instability for non pre-stretched one-side constrained films (taken from [10]). Right: Predicted failure precursor.](image)

![FIG. 3. a) Alignment of creases in the direction of higher pre-stretch as voltage increases for one-side constrained films (taken from [9]). b) Failure precursors with $\lambda_1 = 2$, $\lambda_2 = 1$.](image)
TABLE I. Experiments Vs previous and present theories. †For this stretch range only a qualitative behaviour was obtained in [8] through dimensional analysis. ‡Hessian method.

<table>
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<th>Pre-stretch (dead-load)</th>
<th>Creasing</th>
<th>Pull-in</th>
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<tr>
<td></td>
<td>1</td>
<td>1−2.5</td>
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<tr>
<td>$E_c$ (or $V_c$ [kV])</td>
<td></td>
<td></td>
</tr>
<tr>
<td>experiments</td>
<td>$0.85$</td>
<td>$\checkmark$</td>
</tr>
<tr>
<td>$E_c$ (or $V_c$ [kV])</td>
<td></td>
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<tr>
<td>other theories</td>
<td>$1.93$</td>
<td>$\times$</td>
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<tr>
<td>$E_c$ (or $V_c$ [kV])</td>
<td></td>
<td></td>
</tr>
<tr>
<td>our theory</td>
<td>$0.816$</td>
<td>$\checkmark$</td>
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FIG. 4. Creasing instability after uniaxial pre-stretch; comparison of experiments (dashed line) [8] and theory (solid curves, plotted for different values of $n$, the number of links in the Arruda-Boyce model). Here $E_c = E_c/\sqrt{3/\lambda}$, see [15] for model calibration. Thick solid curve: previous qualitative trend based on dimensional/FEM analysis [8].

FIG. 5. Pull-in instability in equi-biaxially strained films pre-stretched by dead-loads; comparison of experimental (dots) [7] and theoretical (solid curves) critical voltages (in kV). Curves $a,b,c,d$ correspond to dead-loads weighing 20g, 25.5g, 31g, 35.6g, respectively: they describe homogeneous paths until failure and are modeled by the solid curves. Their intersection with the $V_c$ curve (given by Eq. (2)) corresponds to catastrophic thinning (details on calibration in [15]). The Hessian approach can only predict one failure, denoted by $x$.

TABLE I. Experiments Vs previous and present theories. †For this stretch range only a qualitative behaviour was obtained in [8] through dimensional analysis. ‡Hessian method.

uses $0.678−0.686$ in the absence of pre-stretch [5]. Thus, in absence of dead-loads, our analysis is close to the Hessian approach, which gives an estimate of $E_c = 0.687$ [4].

Things change drastically when the film is pre-stretched by applying dead-loads prior to the voltage; then experiments show that giant areal gains can be achieved [7, 17, 18]. Theoretical models based on the Hessian approach fail to predict the actual gain, in particular for higher dead-loads. That’s because the Hessian criterion detects the points where the voltage-stretch curve ceases to be increasing [4]. But elastomers stiffen greatly at large strains, and sufficiently high dead-loads will make the voltage-stretch curve monotonic increasing: this phenomenon can lead to the erroneous conclusion that electromechanical instability can be eliminated by high pre-stretch, in contradiction with experiments [7, 19]. Consider for example experiments on voltage actuated silicone disks, pre-stretched by dead-loads, as carefully conducted and described in [7]. Based on the experimental results reported for the purely mechanical behaviour of VHB silicone, we obtain the homogeneous loading curves for different values of dead-loads using a strain-stiffening model [15]. For lower pre-stretches (Fig.5), the voltage-stretch curves have a peak, corresponding to failure according to the Hessian condition, but this peak disappears for higher values of pre-stretch and the Hessian condition is no longer violated. Nonetheless, the experimental plots have a maximum, clearly corresponding to failure, see last upper experimental dots in Fig.5.

Our theory predicts failure whenever the homogeneous loading curves intersect the critical threshold curve described by formula (2). It gives a clear correspondence between experimental and theoretical thresholds, as seen in Fig.5.

We may thus conclude that the simple formula (2), coupled to that based on snap-through (Hessian) modelling [4, 20], provides a complete picture of the electric breakdown experienced by unconstrained and one-side constrained voltage-actuated thin dielectrics, see summary in Table 1.
while surface energy terms are negligible [8]. When the membrane is thin and curvature effects are neglected [21], the electric field in the film can be approximated as \( V/(h\lambda_3) \), where \( \lambda_3 \) is the thickness stretch. With these assumptions, the total free energy depends only on the deformation.

Thinness of the dielectric film leads to a Taylor expansion of \( \Psi \) in powers of \( h \). Truncating \( o(h^3) \) terms gives the minimal tools for detecting the onset of localisation instabilities (higher-order terms are required for post-critical analysis, beyond the scope of this work.) Denote by \( S \) the constrained (lower) surface for creasing, and the mid-surface for pull-in. Then \( \Psi = \int_S \psi \, da \) where the surface energy density \( \psi \) is expanded as

\[
\psi(\lambda_1, \nabla \lambda_3) = h \varphi(\lambda_1) + h^3 \left( \alpha_1(\lambda_1) \lambda_3^2 + \alpha_2(\lambda_1) \lambda_3^2 \right).
\]

(3)

The stretches \( \lambda_1 \) are functions of the planar coordinates \((x_1, x_2)\), while the gradient \( \nabla \lambda_3 = (\lambda_{3,1}, \lambda_{3,2}) \) accounts for deformation inhomogeneities.

When higher order terms are not considered, only homogeneous configurations can be described. They are characterised by the Euler-Lagrange equations \( \partial \varphi / \partial \lambda_1 = 0 \) and their stability is assessed through convexity of \( \varphi \): which is the so-called Hessian approach [4,20]. When applied to pull-in this approach cannot predict instability, neither for non-equibiaxial states of deformation, nor for large deformations [2]. When applied to creasing (for undeformed or pre-stretched films) it predicts stability for any voltage, which is clearly contradicted by experiments.

These shortcomings are addressed by considering the whole energy [3], that imposes more stringent requirements for the existence of minimisers. Indeed, they exist provided that \( \psi \) is convex in \( \nabla \lambda_3 \), meaning that the functions \( \alpha_1, \alpha_2 \) must be both positive. This is the case as long as the electric field is less than the critical threshold \( E_c \) defined by [2] [15].

Above \( E_c \) the total free energy becomes concave in \( \nabla \lambda_3 \) and no energy minimisers exist (be they homogeneous or belonging to a wide class of non-homogeneous ones). Immediately above \( E_c \), inhomogeneous failure precursors become possible: as soon as they appear, they are energetically more favourable than the homogeneous state, since they lower the total free energy, albeit without finding a minimum (Fig.3): this explains the catastrophic nature of localised thinning. Experiments on one-side constrained films show that when the film is not pre-stretched, localisation mainly takes place in circular spots, whereas when the film is pre-stretched in one direction prior to bonding to the rigid substrate, the resulting thinning localisations align in the direction of higher pre-stretch [8], see Figs 2,3. Both findings are easily covered by our theory.

Indeed, linearising the Euler-Lagrange equation based on (3), we find that failure precursors of the type \( \lambda_3(x_1, x_2) = \lambda_3^0 + w(x_1, x_2) \), with \( \lambda_3^0 \) a constant and \( w \) small, solve

\[
\alpha_1 w_{,11} + \alpha_2 w_{,22} - \Gamma w = 0,
\]

(4)

where \( \alpha_1, \alpha_2 \) and \( \Gamma \) are functions of the underlying stretch and electric field. For almost incompressible materials \( \Gamma \) is always positive whereas, as we have seen, \( \alpha_1, \alpha_2 \) become negative above the critical voltage defined by [2]. For undeformed or equibiaxially stretched layers, \( \alpha_1 = \alpha_2 \) and they become negative together for \( E > E_c \), meaning that polar symmetric solutions of the Bessel type become possible, see Fig.2. When the film is pre-stretched with \( \lambda_1 > \lambda_2 \), for example, the first coefficient that becomes negative is \( \alpha_2 \), leading to sinusoidal solutions in the direction of least stretch, see Fig.3.

With this work we improved the current understanding of the catastrophic nature of electroelastic instabilities in thin films. This unsolved problem goes back as early as 1880, when Röntgen [19] stretched natural rubber using sprayed-on electric charges. In 1955, Stark and Garton [22] detected a previously unrecognized form of breakdown, due to mechanical deformation of electrically irradiated thin films of cross-linked polynyme. The first experimental evidence that dielectric breakdown is due to strong thinning localisation is by Blok and LeGrand [3] in 1969, who provided clear optical evidence (Fig.1) in voltage-controlled polymer films. They speculated that it is experimentally impossible to deform the entire area of the dielectric without using an immense stress, thus inferring the energetic convenience of localising deformations above a critical voltage. Here
we further their intuition and find a completely new paradigm for the analysis of electromechanical instabilities in dielectric films, both one-side constrained and unconstrained. The great majority of technological applications based on dielectric elastomers is dependent on whether wrinkling may or may not be anticipated by electromechanical instability, e.g. tunable adhesion, open-channel microfluidics, etc.\cite{23, 24} and on-demand fluorescent patterning \cite{25}. By re-defining and furthering the concept of electromechanical instability of dielectric films, the paper implies that new experimental campaigns and new analytical studies based on formula \cite{2} are now required to generate a finer physical picture of the catastrophic thinning phenomenon.

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