

# 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 neither completely understood nor modeled 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 prestretch and various material models.

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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. However, 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 localized thinning deformations occur and, furthermore, that this process is *catastrophic*, as the deformations can neither be controlled nor restrained once they have started.

Here, we unveil the physical meaning of catastrophic thinning, based on energy minimization 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 a 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 we show that they are not sustainable but that they give the path to final breakdown; we generalize the results to include prestretch.

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 nonhomogeneous thinning localization 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 nondimensional measure [4] of the electric field:

$$E = \sqrt{\frac{\epsilon V}{\mu h}}, \quad (1)$$

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 an  $E_c$  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

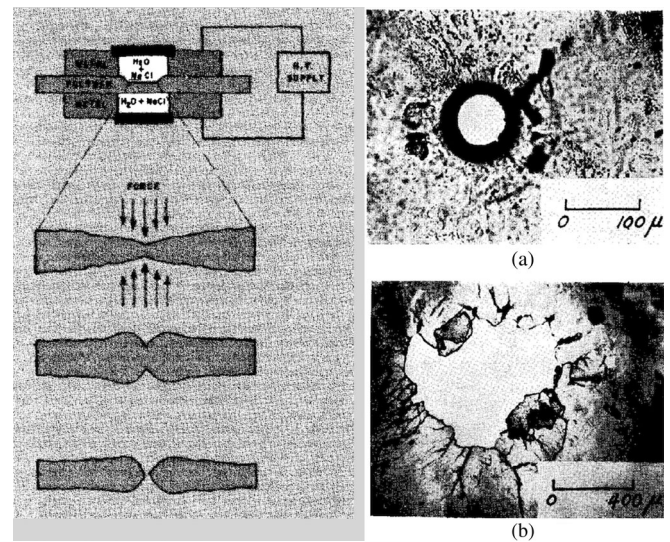


FIG. 1. (Right panels a,b) First recorded experimental evidence of catastrophic localization of thinning deformations (pull-in instability). Reprinted from [3], with the permission of AIP Publishing. (Left panel) Sketch of the experimental setting and qualitative description of the localizations.

prestretch (for one-side constrained films) play a beneficial role in delaying the onset of instabilities [7–9].

However, despite the abundance of experimental data, the critical values of the electric field to date have not been predicted by 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 linearized [10–13] and nonlinear [14] inhomogeneous bifurcation modes on top of the homogeneous deformation, but they require lengthy calculations, do not explain the catastrophic nature of localized deformations, and do not quantify the beneficial effects of prestretch. The situation is even worse for one-side constrained films. So far, electrocreasing has been studied only with entirely numerical methods based on finite element method (FEM) simulations: in the absence of prestretch, 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 prestretch 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 minimizers in a general class of homogeneous and nonhomogeneous deformations. For both unconstrained and constrained films, with and without prestretch and for a quite general class of incompressible materials, we obtain the following simple *unifying formula* for the critical electric field:

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

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 prestretches imposed prior to attachment to the rigid substrate. Above  $E_c$ , no stable configurations exist, neither homogeneous *nor* nonhomogeneous. 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 and 3.

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

For small stretches ( $\lambda_1, \lambda_2 \leq 1.5$ ), the constitutive response can be modeled 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 prestretch ( $\lambda_1 = \lambda_2 = 1$ ), this formula gives

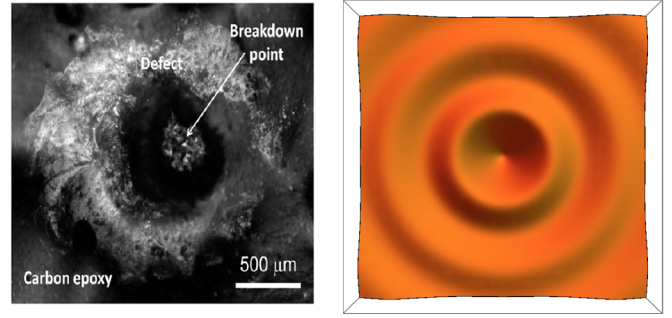


FIG. 2. (Left panel) Experimental creasing instability for non-prestretched one-side constrained films. Reprinted from [30], with the permission of AIP Publishing. (Right panel) Predicted failure precursor.

$E_c = \sqrt{2/3} = 0.816$ , less than 4% off the value  $E_c \approx 0.85$  obtained experimentally [see Ref. [6], Fig. 3(b)] and closer than the estimate  $E_c = 1.03$  obtained by FEM simulations [see Ref. [6], Fig. 4(b)]. For small values of uniaxial prestretch ( $\lambda_1 = \lambda > 1$ ,  $\lambda_2 = 1$ ), 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 prestretch were measured as beneficial in increasing the critical electric field [8,9], an effect that has not yet been explained theoretically or numerically. To demonstrate the ability of formula (2) to reproduce 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. Figure 4 shows the good agreement reached between our theory and the experiments (see the Supplemental Material [15] for the calibration of the model).

We then consider unconstrained films to study the onset of the pull-in instability; see the sketch in the inset of Fig. 5. For equibiaxially stretched films ( $\lambda_1 = \lambda_2 = \lambda$ ) in the absence of dead loads, the homogeneous extension of the neo-Hookean elastomer is described by  $E = \sqrt{\lambda^{-2} - \lambda^{-8}}$  (see Ref. [4] and the Supplemental Material

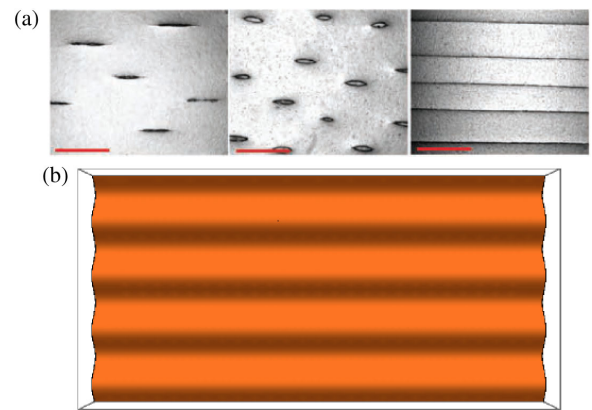


FIG. 3. (a) Alignment of creases in the direction of higher prestretch as voltage increases for one-side constrained films. Reprinted from [8], with the permission of John Wiley & Sons, Inc. (b) Failure precursors with  $\lambda_1 = 2$ ,  $\lambda_2 = 1$ .

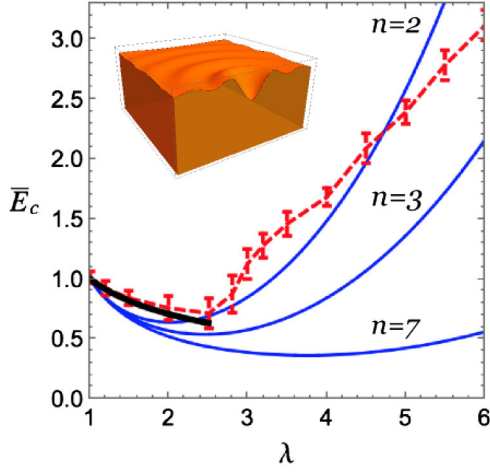


FIG. 4. Creasing instability after uniaxial prestretch. Comparison of experiments (the dashed line) [8] and theory (the solid curves), plotted for different values of  $n$ , the number of links in the Arruda-Boyce model). Here,  $\bar{E}_c = E_c/\sqrt{2/3}$ ; see the Supplemental Material [15] for model calibration. A thick solid curve shows the previous qualitative trend based on dimensional or FEM analysis [8].

[15]). It reaches  $E_c(\lambda) = \sqrt{2/3}\lambda^{-1}$  for  $\lambda = 3^{1/6}$ , where  $E_c = \sqrt{2/3}^{2/3} = 0.680$ , falling squarely within the range of the experimental values 0.678–0.686 in the absence of prestretch [5]. Thus, in the 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 prestretched by applying dead loads prior to the voltage; then

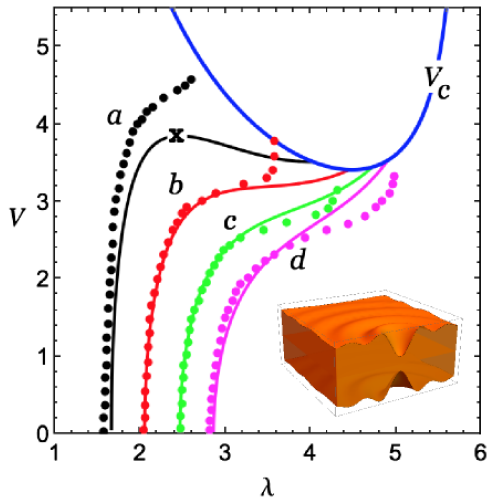


FIG. 5. Pull-in instability in equibiaxially strained films prestretched by dead loads. A comparison of experimental (dots) [7] and theoretical (solid curves) critical voltages (in kilovolts). Curves  $a$ ,  $b$ ,  $c$ , and  $d$  correspond to dead loads weighing 20, 25.5, 31, and 35.6 g, respectively: 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 (see the Supplemental Material [15] for details on the calibration). The Hessian approach can predict only one failure, denoted by  $x$ .

experiments show that *giant areal gains* can be achieved [7,31,32]. Theoretical models based on the Hessian approach fail to predict the actual gain, particularly for higher dead loads. That is because the Hessian criterion detects the points where the voltage-stretch curve ceases to be increasing [4]. However, elastomers stiffen greatly at large strains, and sufficiently high dead loads will make the voltage-stretch curve monotonically increasing: this phenomenon can lead to the erroneous conclusion that electromechanical instability can be eliminated by high prestretch, in contrast to experiments [7,33]. Consider, for example, experiments on voltage-actuated silicone disks, prestretched by dead loads, as carefully conducted and described in Ref. [7]. Based on the experimental results reported for the purely mechanical behavior of VHB<sup>TM</sup> silicone, we obtain the homogeneous loading curves for different values of dead loads using a strain-stiffening model [15]. For lower prestretches (Fig. 5), the voltage-stretch curves have a peak, corresponding to the failure resulting from the Hessian condition, but this peak disappears for higher values of prestretch, and the Hessian condition is no longer violated. Nonetheless, the experimental plots have a maximum, clearly corresponding to failure; see the 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 the 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) modeling [4,34], provides a complete picture of the electric breakdown experienced by unconstrained and one-side constrained voltage-actuated thin dielectrics; see the summary in Table I.

Our theory is based on energy minimization arguments [14]. *Stable* equilibrium configurations minimize the electroelastic free energy  $\Psi = U - QV/2 - \mathcal{W}$ , where  $U$  is the elastic energy,  $Q$  the total charge on the electrodes,  $V$  the voltage, and  $\mathcal{W}$  the mechanical work [15], while surface energy terms are negligible [8]. When the membrane is thin and curvature effects are neglected [35], 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 localization instabilities (higher-order terms are required for postcritical analysis, which is beyond the scope of this work). Denote by  $S$  the constrained (lower) surface for creasing, and the midsurface for pull-in. Then  $\Psi = \int_S \psi da$ , where the surface energy density  $\psi$  is expanded as

$$\psi(\lambda_i, \nabla \lambda_3) = h\varphi(\lambda_i) + h^3[\alpha_1(\lambda_i)\lambda_{3,1}^2 + \alpha_2(\lambda_i)\lambda_{3,2}^2]. \quad (3)$$

The stretches  $\lambda_i$  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.



TABLE I. Experiments vs previous and present theories.

Prestretch (dead load)	Creasing			Pull-in				
	1	1–2.5	2.5–6	1	(20 g)	(25.5 g)	(31 g)	(36.5 g)
$E_c$ [or $V_c$ (kV)] experiments	0.85 [6]	✓ [8]	✓ [8]	0.678–0.686 [5]	✓ [7]	✓ [7]	✓ [7]	✓ [7]
$E_c$ [or $V_c$ (kV)] other theories	1.03 FEM [6]	x dim <sup>a</sup> [8]	x x	0.687 H <sup>b</sup> [4]	✓ H [7]	x x	x x	x x
$E_c$ [or $V_c$ (kV)] our theory	0.816	Fig. 4	Fig. 4	0.680	Fig. 5	Fig. 5	Fig. 5	Fig. 5

<sup>a</sup>For this stretch range only, a qualitative behavior was obtained in Ref. [8] through dimensional analysis.

<sup>b</sup>Hessian method.

When higher-order terms are not considered, only homogeneous configurations can be described. They are characterized by the Euler-Lagrange equations  $\partial\varphi/\partial\lambda_i = 0$  and their stability is assessed through the convexity of  $\varphi$ , which is the so-called Hessian approach [4,34]. When applied to pull-in, this approach cannot predict instability, neither for nonequibiaxial states of deformation nor for large deformations [7]. When applied to creasing (for undeformed or prestretched films), it predicts stability for any voltage, which is clearly contradicted by the experiments.

These shortcomings are addressed by considering the whole energy (3), which imposes more stringent requirements for the existence of minimizers. Indeed, they exist provided that  $\psi$  is convex in  $\nabla\lambda_3$ , meaning that the functions  $\alpha_1$  and  $\alpha_2$  must both be 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 minimizers exist (be they homogeneous or belonging to a wide class of nonhomogeneous ones). Immediately above  $E_c$ , inhomogeneous *failure precursors* become possible: as soon as they appear, they are energetically more favorable than the homogeneous state since they lower the total free energy, albeit without finding a minimum (Fig. 6): this explains the *catastrophic nature* of localized thinning. Experiments on one-side constrained films show that, when the film is not prestretched, localization mainly takes place in circular spots, whereas, when the film is prestretched in one direction prior to bonding to the rigid substrate, the resulting thinning localizations align in the direction of higher prestretch [8]; see Figs. 2 and 3. Both findings are easily covered by our theory.

Indeed, linearizing the Euler-Lagrange equation based on Eq. (3), we find that failure precursors of the type  $\lambda_3(x_1, x_2) = \lambda_3^0 + w(x_1, x_2)$ , with  $\lambda_3^0$  being a constant and  $w$  small, solve [15]

$$\alpha_1 w_{,11} + \alpha_2 w_{,22} - \Gamma w = 0, \quad (4)$$

where  $\alpha_1$ ,  $\alpha_2$ , and  $\Gamma$  are functions of the underlying stretch and the electric field. For almost incompressible materials,  $\Gamma$  is always positive, whereas, as we have seen,  $\alpha_1$  and  $\alpha_2$  become negative above the critical voltage defined by Eq. (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 prestretched 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 Letter, 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 [33] stretched natural rubber using sprayed-on electric charges. In 1955, Stark and Garton [36] detected a previously unrecognized form of breakdown due to the mechanical deformation of electrically irradiated thin films of cross-linked polythene. The first experimental

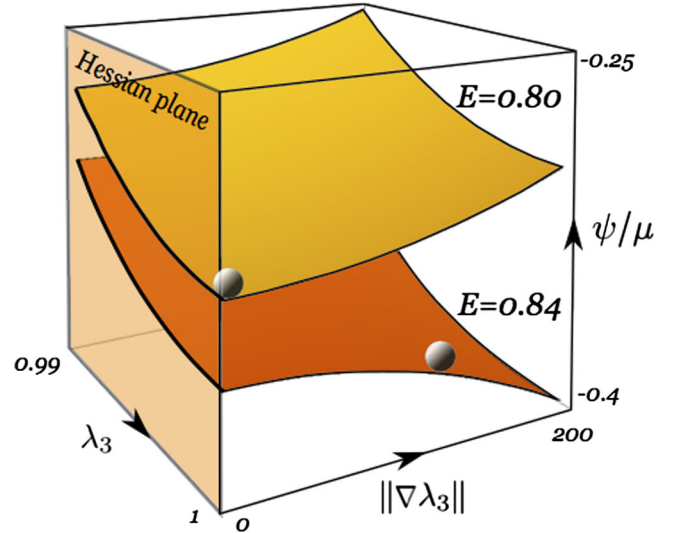


FIG. 6. Electroelastic free energy  $\psi/\mu$  for a one-side constrained film without prestretch ( $\lambda_1 = \lambda_2 = 1$ ). The film is a slightly compressible neo-Hookean material of thickness  $h = 0.01L$ , where  $L$  is a typical lateral length scale and  $\kappa/\mu = 1000$ , with  $\kappa$  being the initial bulk modulus [see Eqs. (7) and (8) in the Supplemental Material [15]]. For  $E < E_c = 0.816$ , the homogeneous solution with  $\lambda_3 = 1$  is the unique energy minimizer; see the upper surface. Above  $E_c$ , the energy is still convex in  $\lambda_3$ , but it becomes nonconvex in  $\nabla\lambda_3$ ; see the lower surface. Failure precursors are then energetically favored, but no energy minimizers exist. In the shaded Hessian plane (at  $\|\nabla\lambda_3\| = 0$ ), Hessian stability takes place; there, the energy is convex in  $\lambda_3$  and the failure threshold cannot be captured.

evidence that dielectric breakdown is due to strong thinning localization was by Blok and LeGrand [1] 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 localizing 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. A great majority of technological applications based on dielectric elastomers are dependent on whether wrinkling may or may not be anticipated by electromechanical instability, e.g., tunable adhesion, open-channel microfluidics, etc. [37,38], and on-demand fluorescent patterning [39]. By redefining and furthering the concept of the electromechanical instability of dielectric films, this Letter implies that new experimental campaigns and new analytical studies based on formula (2) are now required to generate a finer physical picture of the catastrophic thinning phenomenon.

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