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Compression-induced failure of electroactive polymeric thin films

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The onset of compression induces wrinkling in actuation devices based on electroactive polymer thin films, which leads to a sudden decrease in performances and, eventually, to failure. Inspired by the classical tension field theory for thin membranes, we provide a general framework for the analysis of the insurgence of in-plane compressions. Our main result is the analytical deduction of a voltage-dependent domain of tensile configurations in the principal stretches plane. © *2011 American Institute of Physics*. [doi:10.1063/1.3568885]

The growing interest in electroactive polymers (EAPs) as actuator devices results from their relevant qualities in terms of lightness, small dimensions, low costs, flexibility, and fast response. A typical EAP device consists of a thin polymer sheet sandwiched between two compliant electrodes. The compressive Coulomb forces acting on the electrodes induce transversal dilatation that is used for actuation. Since typical devices are constituted by thin films constrained inside rigid frames, typical failure mechanisms of EAPs are determined by a sudden loss of equilibrium due to in-plane compression. The onset of deformation localizations has been analyzed in a series of interesting papers.[1–6] In this perspective, prestretch may provide a real improvement of the actuation device, as several papers show (e.g., Refs. 4, 7, and 8), detecting also the existence of an optimal prestretch value.

Here, we obtain explicit analytical results for Mooney–Rivlin incompressible materials describing the onset of compression instability. While analytical and numerical results about this phenomenon were already obtained (see, e.g., Refs. 1 and 4) under restrictive assumptions on the deformation homogeneity and device geometry, a general framework for the analysis of this effect is still missing.

Our results take inspiration on the tension field theory for thin elastic membranes.[9,10] The main ingredient of the theory is the existence of a *natural width* which assigns a threshold to one in-plane principal stretch as a function of the other one. This threshold separates compressed and tensile states. Accordingly, the space of principal in-plane stretches can be decomposed into a domain where both principal stresses are positive (*tensile configurations*), a domain where only one principal stress is positive (*wrinkled configurations*), and the remaining region where no principal stress is positive. Here, we extend these results to the analysis of electroactivated membranes. As we show, for sufficiently high values of the assigned voltage, the tensile region reduces to an "island" that shrinks as the voltage is increased. We then deduce the existence of a loading threshold (critical voltage) such that for larger value of the electric load no tensile configuration is possible. As we show under particular boundary conditions, this thresholds leads to the typical pull-in instability. The amplitude of the safe stretch region and the critical threshold strongly depend on the material properties. Roughly speaking, *the stiffer, the safer*.

Our model can be extended to general constitutive hypotheses and does not assume homogeneous deformations. The proposed approach may provide a useful tool for the design of electroactive materials, a very attractive area of scientific and technological research. By considering two specific boundary value problems, amenable of fully analytical results, we show the effectiveness of the proposed model in order to describe the main physical phenomena observed in the wrinkling of EAPs.

We now collect the main equations for a continuum body under electromechanical loading. We refer the reader to Ref. 11 and to the references therein for details.

Let $\boldsymbol{f}$ be the deformation carrying the continuum body $\mathcal{B}$ (reference configuration) to the current configuration $\mathcal{B}' = \boldsymbol{f}(\mathcal{B})$ and let $\mathbf{F}=\nabla \boldsymbol{f}$ be the deformation gradient. The left Cauchy–Green tensor is $\mathbf{B}=\mathbf{F}\mathbf{F}^T$ and we indicate with $\boldsymbol{e}_i$ and λ_i^2 the eigenvectors and eigenvalues of $\mathbf{B}$, where the λ_i are the principal stretches. $\boldsymbol{D}$ and $\boldsymbol{E}$ are the electric displacement and the electric field in the current configuration $\mathcal{B}'$, respectively. For a linear, homogeneous and isotropic dielectric material $\boldsymbol{D}=\varepsilon \boldsymbol{E}$, where $\varepsilon=\varepsilon_0\varepsilon_d$ with ε_0 the permittivity of free space and ε_d the dielectric constant of the material.

Under the simplifying assumption that the dielectric constant does not depend on the deformation and that the material moduli do not depend on the electric field, we may consider the additivity of the total stress, with the (current) Cauchy stress tensor $\mathbf{T}$ (see, Ref. 11) assigned as the sum of the elastic stress tensor $\mathbf{T}^{el}$ and the electric Maxwell stress tensor, $\mathbf{T}=\mathbf{T}^{el}+\mathbf{T}^{M}$. Despite the approach here proposed is general and does not require specific constitutive assumptions on the elastic response of the polymer, for the sake of clarity we consider an incompressible ($\det \mathbf{F}=\lambda_1\lambda_2\lambda_3=1$), isotropic, elastic Mooney–Rivlin material

$$\mathbf{T}^{el} = -\pi\mathbf{I} + 2c_1\mathbf{B} - 2c_2\mathbf{B}^{-1}, \tag{1}$$

where $c_1 \geq 0$ and $c_2 \geq 0$ are constant material parameters and π is an undetermined pressure. The electric part of the stress (Maxwell stress) can be expressed by (see again Ref. 11)

$$\mathbf{T}^{M} = \varepsilon\left[\boldsymbol{E}\otimes\boldsymbol{E} - \tfrac{1}{2}(\boldsymbol{E}\cdot\boldsymbol{E})\mathbf{I}\right]. \tag{2}$$

Thus, we have

$$\mathbf{T} = -p\mathbf{I} + 2c_1\mathbf{B} - 2c_2\mathbf{B}^{-1} + \varepsilon\boldsymbol{E}\otimes\boldsymbol{E} \tag{3}$$

where $p=\pi+\varepsilon E^2/2$. Let us now introduce the concept of *tensile stretches region*. Consider an isotropic, incompressible thin sheet whose upper and lower faces are bonded to

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compliant electrodes with applied voltage V. As reference configuration, we consider a right cylindrical region with flat mid-surface Ω, normal to $\boldsymbol{e}_3$, and constant thickness h. We embrace the *membrane approximation* which asserts that the bending stiffness is zero and that any in-plane compressive stress immediately leads to the membrane buckling. Moreover, according with most common application schemes of EAPs we assume that Ω remains flat after deformation. We also assume that the deformation is normal preserving, with the fibers along the thickness direction which deform respecting incompressibility: $\lambda_3=(1/\lambda_1\lambda_2)$.

The application of a voltage on the electrodes determines the onset of an electric field $\boldsymbol{E}$ which should be rigorously calculated by solving the corresponding electromechanical equilibrium problem (see, e.g., Ref. 11). On the other hand, since each electrode is an equipotential surface, we assume that the electric field remains perpendicular to Ω. Thus, if a voltage V is applied to the electrodes, the electric field is $\boldsymbol{E}=(V/h\lambda_3)\boldsymbol{e}_3$. Under these hypotheses (3) gives the principal stresses

$$t_1=-p+2c_1\lambda_1^2-2c_2\lambda_1^{-2},$$

$$t_2=-p+2c_1\lambda_2^2-2c_2\lambda_2^{-2},$$

$$t_3=-p+2c_1\lambda_1^{-2}\lambda_2^{-2}+2(k_V-c_2)\lambda_1^2\lambda_2^2, \tag{4}$$

where, as proposed in Ref. 1, we introduce the electric energy density, which represents our activation parameter

$$k_V=\frac{\varepsilon V^2}{2h^2}. \tag{5}$$

The undetermined multiplier p can be deduced by imposing the boundary condition $t_3=0$ on the upper and lower faces: $p=2c_1\lambda_1^{-2}\lambda_2^{-2}+2(k_V-c_2)\lambda_1^2\lambda_2^2$. After substitution in (4), the in-plane principal stresses are

$$t_1=2[c_1(\lambda_1^2-\lambda_1^{-2}\lambda_2^{-2})-c_2(\lambda_1^{-2}-\lambda_1^2\lambda_2^2)-k_V\lambda_1^2\lambda_2^2],$$

$$t_2=2[c_1(\lambda_2^2-\lambda_1^{-2}\lambda_2^{-2})-c_2(\lambda_2^{-2}-\lambda_1^2\lambda_2^2)-k_V\lambda_1^2\lambda_2^2]. \tag{6}$$

We are now in position to introduce the central idea of natural width in simple tension developed within the context of tension field theory of thin elastic membranes (e.g., Ref. 10 and references therein). Consider a state of local uniaxial stress in direction $\boldsymbol{e}_1$; under the assumption $t_2=t_3=0$, the transverse stretch in direction $\boldsymbol{e}_2$ assumes a specific value called *natural width* in tension, which is constitutively dependent on the stretch λ_1

$$\lambda_2=\nu(\lambda_1,k_v). \tag{7}$$

Since for $\lambda_2=\nu(\lambda_1,k_v)$ it is $t_2=0$, any attempt to reduce the transverse stretch under this value can be shown to require the application of a compressive stress, leading to the formation of wrinkles. While in the classical tension field theory without electric actions, it results $\lambda_2=\nu(\lambda_1)=\lambda_1^{-1/2}$, in the present case the natural width depends on the applied voltage and in view of Eq. (6) (Ref. 1) takes the form

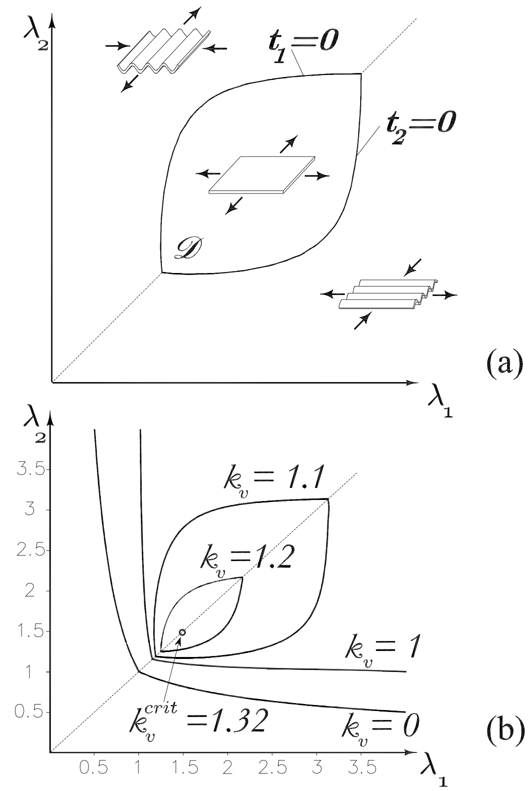

FIG. 1. (a) Representation of the tensile region $\mathcal{D}$. (b) Dependence of the tensile region on the activation parameter k_v. Here, $c_1=1$ and $c_2=1$. The critical value k_v^{crit} is the activation threshold that leads to the disappearance of tensile states.

$$\lambda_2=\nu(\lambda_1,k_v)=\lambda_1^{-1/2}\left[\frac{c_1+c_2\lambda_1^2}{c_1+c_2\lambda_1^2-k_V\lambda_1^2}\right]^{1/4}. \tag{8}$$

Analogous considerations hold for uniaxial tension in direction $\boldsymbol{e}_2$, so that the condition $t_1=0$ gives the natural width in the transverse direction $\boldsymbol{e}_1$

$$\lambda_1=\nu(\lambda_2,k_v)=\lambda_2^{-1/2}\left[\frac{c_1+c_2\lambda_2^2}{c_1+c_2\lambda_2^2-k_V\lambda_2^2}\right]^{1/4}. \tag{9}$$

One may deduce that for given voltage V, the membrane is in tension when $\lambda_1>\nu(\lambda_2,k_v)$ and $\lambda_2>\nu(\lambda_1,k_v)$, otherwise the membrane undergoes a compression-induced instability. Thus, there exists a voltage-dependent region in the principal stretches space [see Fig. 1(a)]

$$\mathcal{D}(k_v)=\{(\lambda_1,\lambda_2):\lambda_1>\nu(\lambda_2,k_v),\lambda_2>\nu(\lambda_1,k_v)\}, \tag{10}$$

that collects the possible values of (λ_1,λ_2) corresponding to tensile states. Wrinkling arises for combinations of the principal stretches which do not belong to $\mathcal{D}$.

As depicted in Fig. 1, the domain $\mathcal{D}$ is symmetric with respect to the straight line $\lambda_1=\lambda_2$ and its boundary is given by the states for which $t_1(\lambda_1,\lambda_2,k_v)=0$, where $\lambda_2>\lambda_1$ and by the states for which $t_2(\lambda_1,\lambda_2,k_v)=0$, where $\lambda_1>\lambda_2$. The upper and lower vertices where the boundary of $\mathcal{D}$ intersects the line $\lambda_1=\lambda_2$ represent equibiaxial, unstressed configurations with $t_1=t_2=0$.

Due to the symmetry of $\mathcal{D}$ with respect to the line $\lambda_1=\lambda_2$, let us focus on the curve $t_2=0$, where $\lambda_1>\lambda_2$. When the applied voltage is zero, then $\nu(\lambda_1,0)=\lambda_1^{-1/2}$. In this case, which corresponds to the curve $k_V=0$ in Fig. 1(b), the domain $\mathcal{D}$ is unbounded.

We will now discuss how the region $\mathcal{D}$ changes during an increasing path of the voltage starting from zero. The lower relevant threshold which is met is given by the voltage corresponding to $k_V = c_2$. For $c_1 \geq 0$ and $c_2 \geq 0$, a straightforward analysis shows that if $k_V < c_2$ the domain $\mathcal{D}$ remains unbounded at infinity; in other words in this case the curve $t_2 = 0$ has one single intersection with the line $\lambda_1 = \lambda_2$. As soon as the voltage overcomes the threshold for which $k_V = c_2$ the domain $\mathcal{D}$ becomes bounded since the function ν has a vertical asymptote in correspondence of the stretch

$$\lambda_1 = \lambda^* = \sqrt{\frac{c_1}{k_V - c_2}}. \tag{11}$$

In this case a simple analysis shows that the upper and lower intersections of $\mathcal{D}$ with the line $\lambda_1 = \lambda_2$ are both greater than 1 and lower than λ^*.

By further increasing the activation parameter k_V, the two vertices approach each other until they coalesce for a critical voltage: the domain $\mathcal{D}$ degenerates in a single point and, for values of the voltage greater than this critical value, the region $\mathcal{D}$ simply does not exist anymore. We denote by k_V^{crit} the activation energy at the critical voltage and with λ^{crit} the corresponding stretch threshold [see Fig. 1(b)].

We now apply our analysis to homogeneously deformed EAP sheets under two different boundary conditions. In order to get explicit analytic solutions we consider the special case of Neo–Hookean materials, i.e., $c_2 = 0, c_1 = \mu/2$, where μ is the shear modulus. Here, Eq. (8) gives

$$\lambda_2 = \nu(\lambda_1, k_V) = \lambda_1^{-1/2}\left(\frac{\mu}{\mu - 2k_V\lambda_1^2}\right)^{1/4}. \tag{12}$$

In this case the two vertices of the region $\mathcal{D}$, with $\lambda_1 = \lambda_2 = \lambda$, are the solutions of $2k_v\lambda^8 - \mu\lambda^6 + \mu = 0$. These vertices coalesce for $k_V = k_V^{\mathrm{crit}} = 3\mu/2^{11/3}$, which corresponds to an equibiaxial strain $\lambda_1 = \lambda_2 = \lambda^{\mathrm{crit}} = 2^{1/3}$.

Consider the case of an EAP membrane under an assigned voltage V and without prestretch on the edges (upper scheme of Fig. 2). By imposing that $t_1 = t_2 = t_3 = 0$, we obtain that the equilibrium solutions correspond to the intersections of the boundary of $\mathcal{D}$ with the line $\lambda_1 = \lambda_2$, represented with filled circles in Fig. 2. As a consequence, we may interpret the vertices of $\mathcal{D}$ as the stretches corresponding to the present situation. Observe that for given activation k_V there are two equilibrium solutions. Moreover, the stretch of the equilibrium solution corresponding to the upper vertex decreases as k_V grows; accordingly, the membrane thickness grows with increasing voltage and we may argue that this equilibrium solution is unstable.

Based on the previous analysis, we deduce that when we increase k_V the system stays in the equibiaxially stretched configuration corresponding to the lower vertex of $\mathcal{D}$. When k_V reaches k_V^{crit} in correspondence of $\lambda = \lambda_{\mathrm{crit}}$, the region disappears: after this threshold no equilibrium solution is possible. This effect represents what is denoted in the literature as pull-in instability (see Ref. 1). Observe that the maximum activation value grows with the stiffness of the material.

Consider now the case where a prestretch $\lambda_2 = \hat{\lambda}$ is prescribed in direction $\boldsymbol{e}_2$ of a square EAP membrane (lower scheme of Fig. 2). The homogeneous equilibrium solution is obtained by requiring $t_1 = t_3 = 0$ (empty circles in Fig. 2). In this case, for given k_V the value of the natural width in direction $\boldsymbol{e}_1$ is given by Eq. (8) as $\lambda_1 = \nu(\hat{\lambda}, k_V)$. The boundary of $\mathcal{D}$ here represents the equilibrium solutions in the prestretched case.

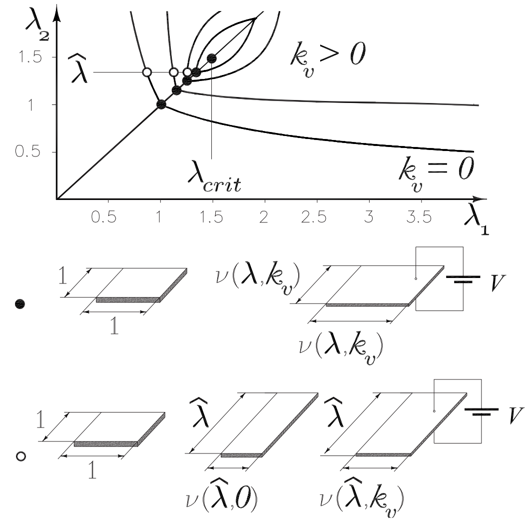

FIG. 2. Equilibrium solutions under the hypothesis of homogeneous deformation for different values of the activation parameter k_v. Empty and filled circles represent the equilibrium states for the prestretched and the nonprestretched cases, respectively (values of parameters as in Fig. 1).

Observe that the system loses its equilibrium for an activation k_V (see Fig. 2) such that the straight line $\lambda_2 = \hat{\lambda}$ corresponds to one of the two vertices of the tensile region. Thus, we have that the largest activation $k_V = k_V^{\mathrm{crit}}$ is attained if we choose a prestretch $\hat{\lambda}_2 = \lambda^{\mathrm{crit}}$. We recall that the existence of an optimal prestretch is also experimentally deduced Eq. (6) and it was already theoretically justified in Ref. 1.

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