Surface Tension Effects on Surface Instabilities of Dielectric Elastomers

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Abstract

Dielectric elastomers have recently been proposed for various biologically-relevant applications, in which they may operate in fluidic environments where surface tension effects may have a significant effect on their stability and reliability. Here, we present a theoretical analysis coupled with computational modeling for a generalized electromechanical analysis of surface stability in dielectric elastomers accounting for surface tension effects. For mechanically deformed elastomers, significant increases in critical strain and instability wavelength are observed for small elastocapillary numbers. When the elastomers are deformed electrostatically, both surface tension and the amount of pre-compression are found to substantially increase the critical electric field while decreasing the instability wavelength.

Keywords: Wrinkling Instability, Elastocapillary, Perturbation Analysis

1. Introduction

Because soft materials like gels and elastomers often operate under states of compression, there have been significant efforts to examine their mechanical stability under such loading (Biot, 1963; Gent and Cho, 1999; Biot and Romain, 1965; Tamaki et al., 1987; Gent, 2005; Hong et al., 2009). Recently, efforts have focused on examining the stability of surfaces due to the formation of instabilities such as wrinkles and creases under compression (Cao and Hutchinson, 2012; Li et al., 2012; Jin et al., 2014; Cai et al., 2012; Hong and Gao, 2013; Weiss et al., 2013). The studies of wrinkling instabilities have typically been analytical in nature, based on linear perturbation analysis, which enables predictions of critical strains and instability wavelengths in these soft structures. These studies have also been insightfully combined with experiments (Cai et al., 2010, 2012; Jin et al., 2015), and also numerical (finite element) analyses (Cao and Hutchinson, 2012; Jin and Suo, 2015; Wang and Zhao, 2014, 2016).

More recently, linear perturbation analyses have been applied to an interesting class of soft, active materials - dielectric elastomers (DEs). These materials have drawn significant interest due to the large deformations they can undergo resulting from applied electrostatic loading (Carpi et al., 2010; Brochu and Pei, 2010; Biddiss and Chun, 2008; Mirmakhrai et al., 2007). Furthermore, DEs often form surface instabilities like creasing, wrinkling and cratering, which have been studied via experiment, theory and computation (Su, 2010; Plante and Dubowsky, 2006; Zhou et al., 2008; Wang et al., 2012; Shivapooja et al., 2013, Wang et al., 2011; Wang and Zhao, 2013; Park et al., 2012, 2013), and also using analytic techniques based on stability analyses (Zhao and Suo, 2007, 2009) and the nonlinear field theory of Suo et al. (2008).

While DEs have drawn significant interest in recent years, many promising technological innovations, such as generating electromechanical motion during magnetic resonance imaging (Carpi et al., 2008), operating soft, underwater robots (Rus and Tolley, 2015; Kim et al., 2013), manipulating microfluidic flow (Holmes et al., 2013), focusing a tunable lens (Carpi et al., 2011), the preparation of bioinspired surfaces (Shivapooja et al., 2013), underwater grippers (Laschi et al., 2012) and soft body locomotion (Marchese et al., 2014), involve operation of the DE in fluidic environments, where elastocapillary effects due to surface tension can become prominent (Roman and Bico, 2010; Liu and Feng, 2012). While prior works have examined either the effects of surface tension alone on surface instabilities in elastomers (Chen et al., 2012) and hydrogels (Kang and Huang, 2010), or the coupled effects of surface tension and electric fields on surface instabilities in constrained DE films (Wang and Zhao, 2013; Seifi and Park, 2016; Wang et al., 2016), a general stability analysis of compressed DE films subject to both surface tension and electric fields has not been performed.

The objective of this work is to present a theoretical analysis coupled with computational modeling for surface tension effects on surface instabilities in DEs, with particular emphasis on surface instability wavelengths, and critical strains or electric fields needed to induce the surface instabilities. We consider first the case of surface tension effects on a mechanically compressed DE film, then move on to consider the general case of electrostatic loading on...
The nominal stress pressibility:

\[ s_{iK} = \mu F_{iK} - \pi H_{iK} \]  

where \( H = F^{-T} \), \( \mu \) is the shear modulus, and \( \pi = \pi(x) \) is the Lagrange multiplier to enforce the constraint of incompressibility: \( J = \det F = 1 \). The nominal stress satisfies the equilibrium equation:

\[ \frac{\partial s_{iK}}{\partial X_K} = 0 \]  

(4)

On the surface of the body the film is subjected to the Young-Laplace boundary condition \((\text{Saksono and Peric} \ 2006)\) with surface tension \( \gamma \):

\[ s_{iK} N_K dA = 2\gamma \kappa n_i da \]  

(5)

where \( N \) is the unit normal vector to the surface of the body in the undeformed state, \( n \) is the unit normal vector in the current state, and \( \kappa \) is the mean curvature of the surface.

We now add a perturbation into the state of finite deformation with position \( x^0(x) \), deformation gradient \( F_{iK}^0(x) \) and nominal stress \( s_{iK}^0(x) \). If the deformation in the \( z \)-direction is constrained, only 2D deformation is considered. The deformation gradient of this state is

\[ F^0 = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & 1/\lambda & 0 \\ 0 & 0 & 1 \end{bmatrix} \]  

(6)

Therefore the corresponding position is \( x^0 = F^0 x \). Now by adding a small displacement perturbation of \( x \), we write the perturbed state as \( x = x^0 + \delta x \). Both state \( x^0(x) \) and the perturbed state \( x^0(x) + \delta x(x) \) satisfy the same governing equations in (1) and (2). The incremental form of the deformation gradient reduces to

\[ \delta F_{iK} = \frac{\partial \delta x_i}{\partial X_K} \]  

(7)

and the equilibrium equation to

\[ \frac{\partial \delta s_{iK}}{\partial X_K} = 0 \]  

(8)

where the incremental nominal stress can be obtained using Taylor’s expansion at \( F^0 \) of the stress in (3):

\[ s_{iK} = \mu \delta F_{iK} - \pi H^0_{iL} H^0_{jK} \delta F_{jL} \]  

(9)

Inserting (9) into (8) we can obtain the following equation:

\[ (\mu \delta_{ij} - \delta_{KL} H^0_{iL} H^0_{jK}) \frac{\partial^2 \delta x_i}{\partial X_K \partial X_L} - H^0_{iK} \frac{\partial \pi}{\partial X_K} = 0 \]  

(10)

The perturbation of the incompressibility condition becomes

\[ H^0_{iK} \delta F_{iK} = 0 \]  

(11)

where the perturbation of the boundary-condition is:

\[ s_{iK} N_K = 2\gamma \kappa H^0_{iK} N_K + 2\gamma \kappa H^0_{iK} N_K \]  

(12)

Finally, the boundary condition on the top the film reduces to

\[ s_{iK} N_K = 2\gamma \kappa H^0_{iK} N_K \]  

(13)

where the term involving \( \kappa \) in (12) drops out as \( \kappa = 0 \). At the bottom of the film, we have the boundary conditions:

\[ s_{12} = 0 \quad \text{and} \quad \hat{x}_2 = 0 \]  

(14)
and therefore the nominal stress of (3) becomes

\[ s^e = J \sigma^e \mathbf{H} \]

where \( E_2 = \Phi/h_f \) is the applied electric field where \( h_f = H_f/\lambda^{pre} \) is the height of the pre-compressed film.

The electric field \( \mathbf{E} \) generates an extra stress \( s^e \) inside the film. The nominal electric stress can be calculated as

\[ s^e = J \sigma^e \mathbf{H} \]

where \( \sigma^e = \epsilon \mathbf{E} \otimes \mathbf{E} - \frac{1}{2} | \mathbf{E} |^2 \mathbf{I} \).

Also here \( \mathbf{H} = (\mathbf{F}^0)^{-T} \) where \( \mathbf{F}^0 \) is the deformation gradient that describes the post-compressed motion. Therefore we can obtain the nominal electric stress as

\[ s^e = J \left[ - \frac{1}{2} \epsilon E_2^2 \right. \]

The equilibrium equation becomes

\[ \frac{\partial s_{ik}}{\partial X_K} = 0 \]

With relation \( 19 \) and the fact that the film remains flat before the onset of surface wrinkling, we have the approximation \( \mathbf{F}^0 \approx \mathbf{I} \) and therefore \( (\mathbf{F}^0)^{-T} = \mathbf{H} \approx \mathbf{I} \), we can obtain \( \frac{\partial s_{ik}}{\partial X_K} \approx 0 \). Thus the the stress equilibrium reduces to:

\[ \frac{\partial s_{ik}}{\partial X_K} = 0 \]

Now similar to the previous case, we perturb the post-compressed state of deformation, of displacement \( x_i^0 \) and deformation gradient \( \mathbf{F}^0 \). The incremental state of stress becomes:

\[ \delta s_{ik} = \mu F_{Km}^{\text{pre}} F_{Lm}^{\text{pre}} x_i^0 + \pi H_{ik}^0 x_i^0 \mathbf{F}_{jL}^0 - H_{ik}^0 \delta \]

and the equilibrium equation becomes:

\[ \frac{\partial \delta s_{ik}}{\partial X_K} = 0 \]

where it can be rewritten as:

\[ (\mu F_{Km}^{\text{pre}} F_{Lm}^{\text{pre}} \delta s_{ij} + \pi H_{ik}^0 H_{jK}^0 H_{kl}^0 - H_{ik}^0 H_{jk}^0) \frac{\partial^2 x_i}{\partial X_K \partial X_L} - H_{ik}^0 \frac{\partial \delta}{\partial X_K} = 0 \]

After rearrangement of electric stress the boundary condition on top of the film is:

\[ \delta s_{ik} N_K = 2 \gamma \kappa n_i - \delta s_{ik} N_K \]

where according to \( 19 \) we have \( \delta s_{ik} = \delta \sigma_{ij} \) as a consequence of the fact that \( \mathbf{H} \approx \mathbf{I} \). By assuming the fact that the DE film undergoes a sinusoidal undulation with small amplitude \( \delta \) at the onset of wrinkling, the electric field then is \( (E_2)_0 = \Phi/(h_f + \delta) \), and therefore the linear approximation of the electrical stress using Taylor’s series becomes:

\[ \delta s_{ik} = \left[ \epsilon E_2^2 \frac{\delta}{(h_f + \delta)} \right. \]

And finally the boundary conditions at the bottom of the film is fixed:

\[ \delta x_1(X, H_f) = \delta x_2(X, H_f) = 0 \]

Figure 2: A perfectly flat film subject to compression, electric field and surface tension \( \gamma \). (a) Undeformed configuration; (b) Compressed to a given strain \( \varepsilon^{\text{pre}} \), after which voltage \( \Phi(t) \) is applied to the top surface while \( \Phi = 0 \) on the bottom surface; (c) The formation of wrinkles with critical wavelength \( l_c \) at critical electric potential \( \Phi_c \).

2.2. Dielectric Elastomer with Surface Tension and Electric Field Effects

We now consider the general problem of interest, that of a DE film that is subject to compressive loading, while accounting for both surface tension and electrostatic loading via electric fields. This is illustrated in Figure [1]. In this case, the reference state is a pre-compressed plane strain film, subject to the deformation gradient

\[ \mathbf{F}^{\text{pre}} = \begin{bmatrix} \lambda^{\text{pre}} & 0 & 0 \\ 0 & 1/\lambda^{\text{pre}} & 0 \end{bmatrix} \]

For this film, the net deformation gradient between the current state and the undeformed state is \( \mathbf{F}_{ik} \mathbf{F}_{Km}^{\text{pre}} \), and the free energy of equation in [2] becomes

\[ W(\mathbf{F}) = \frac{\mu}{2} F_{ik} F_{Km}^{\text{pre}} F_{iL} F_{Lm}^{\text{pre}} - \pi(\text{det} \mathbf{F} - 1) \]

and therefore the nominal stress of (3) becomes

\[ s_{ik} = \frac{\mu}{2} F_{ik}^{\text{pre}} F_{Km}^{\text{pre}} F_{iL} F_{Lm}^{\text{pre}} - \pi H_{ik} \]

At this state (Figure [2]b), an electric potential \( \Phi \) is applied on top of the film along with the surface tension \( \gamma \). When the elastomer is at the flat state, the electric field in the elastomer film is homogeneous, with the electric field vector as

\[ \mathbf{E} = \begin{bmatrix} 0 \\ E_2 \end{bmatrix} \]
3. Linear Perturbation Analysis for Wrinkles

3.1. Compression with elastocapillary effect

To determine the onset of wrinkling, we require the existence of non-trivial solutions to the eigenvalue problem corresponding to the incremental boundary value problems derived in the previous section. Separated solution exists in the incremental boundary value problem with the perturbation in the following form:

\[
\dot{x}_1(X_1, X_2) = f_1(X_2) \sin(KX_1)
\]
\[
\dot{x}_2(X_1, X_2) = f_2(X_2) \cos(KX_1)
\]
\[
\pi(X_1, X_2) = f_3(X_2) \cos(KX_1)
\] (28)

Substituting these equations into (10), we obtain

\[
f''''_2 - K^2 \left( \lambda^{-4} + 1 \right) f''_2 + K^4 \lambda^{-4} f = 0
\] (29)

The derivatives of \( f_2 \) are with respect to \( X_2 \) and \( K = 2\pi/L \) is the wave number. The wavelength in the reference state \( L \) relates to wavelength in current \( l \) by state by \( L = l/\lambda \). The ODE in (29), accompanied by the boundary conditions in (13) and (14), leads to an eigenvalue problem, of which the non-trivial solutions correspond to the wrinkling state. The corresponding algebraic equation of the following form then can be obtained

\[
A \begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \end{bmatrix} = 0
\] (30)

The existence of a non-trivial solution to the perturbed boundary value problem requires

\[
\det A = 0
\] (31)

where matrix \( \det A \) can be written as a function of \( \det A = f(\lambda, KH_f, \gamma/(\mu H_f)) \). The explicit expression of this matrix is given as follows:

\[
A_{11} = -\frac{\gamma KH_f}{\lambda \mu} - H_f K \left( \lambda^2 + \frac{1}{\lambda^2} \right)
\]
\[
A_{12} = -\frac{\gamma KH_f}{\lambda \mu} + H_f K \left( \lambda^2 + \frac{1}{\lambda^2} \right)
\]
\[
A_{13} = -\frac{\gamma H_f K^2}{\mu \lambda} - 2KH_f
\]
\[
A_{14} = -\frac{\gamma H_f K^2}{\mu \lambda} + 2KH_f
\]
\[
A_{21} = A_{22} = \frac{2KH_f}{\lambda^2}
\]
\[
A_{23} = A_{24} = \lambda^2 KH_f + \frac{KH_f}{\lambda^2}
\] (32)

By solving the eigenvalue problem in (31), we obtain the relation between stretch \( \lambda \), and therefore the compressive strain \( \varepsilon = 1 - \lambda \) with the wavelength \( l \) (Figure 3). For each curve, the minimal critical strain \( \varepsilon_c = 1 - \lambda_c \) reaches a minimum for wrinkles of certain wavelength \( l_c \). The relation of these critical values with elastocapillary number \( \gamma/(\mu H_f) \) is plotted in Figure 3.

When surface tension is neglected (\( \gamma = 0 \)), we recover Biot’s value of wrinkling at \( \varepsilon_w \approx 0.46 \) as shown in Figure 3 (Biot, 1963). When surface tension is present (\( \gamma > 0 \)), the strain \( \varepsilon \) first decreases and then increases as the normalized wavelength \( l/H_f \) increases (Figure 4). The key effect of surface tension is that it inhibits bifurcation, as both the critical strain \( \varepsilon_c \) and corresponding normalized wavelength \( l_c/H_f \) increase with increasing \( \gamma/(\mu H_f) \) (Figure 4). These results are consistent with previous studies on surface tension effects on surface instabilities (Chen et al., 2012), and are expected since a smaller wavelength \( l/H_f \) implies a larger energy penalty in terms of surface energy (Shenoy and Sharma, 2001).

In addition, we find that for large elastocapillary numbers, the critical strain approaches a limiting value of \( \varepsilon_c \approx 0.85 \). The largest change in critical strain occurs for small elastocapillary numbers, i.e. \( 0 \leq \gamma/(\mu H_f) \leq 2 \), where the change in \( \varepsilon_c \) is more than 34% from \( \varepsilon_c = 0.46 \)
Figure 4: (a): Critical strain $\varepsilon_c$ vs. elasto-capillary number $\gamma/(\mu Hf)$; (b): Critical wavelength $l_c/H_f$ vs. elasto-capillary number $\gamma/(\mu Hf)$

for $\gamma/(\mu Hf) = 0$ to $\varepsilon_c \approx 0.7$ for $\gamma/(\mu Hf) = 2$, while for $\gamma/(\mu Hf) > 2$ the increase in $\varepsilon_c$ is less than 18% (Figure 4).

The solutions to the critical wavelength are different from the critical strain in that there is no limit to the wavelength. As with the critical strain in Figure 4, a rapid increase in normalized wavelength is observed for small elastocapillary numbers. For $\gamma/(\mu Hf) > 4$, where the change in critical strain is not significant, the normalized wavelength continues increasing in a linear fashion.

We also verified the theoretical model by performing dynamic, nonlinear finite element (FE) calculations using the methodology for electro-elastocapillary phenomena in DEs previously developed by Seifi and Park (2016), while neglecting the electrostatic effects, thus considering a purely mechanical problem. All numerical simulations using open source simulation code Tahoe (2015) using standard 4-node, bilinear quadrilateral finite elements within a two-dimensional, plane strain approximation. Simulations were performed on a elastomer film with length $L_f = 40$ and height of $H_f = 4$ with a mesh size of $d = 1/16$.

In the FE simulations, we measured the critical strain $\varepsilon_c$ as soon as the wrinkling pattern on surface appears, see for example (Figure 5). For the comparisons to the critical strain in Figure 4(a), the FE results match the theoretical model very closely. For the normalized wavelength $l_c/H_f$ in Figure 4(b), the FE results match also closely match the theoretical model. However, we were not able to obtain the wavelength of the wrinkles for $\gamma/(\mu Hf) > 2$ where the film is more than 70% compressed due to the computational expense needed in modeling very long films.

3.2. Compressed film subject to surface tension and electric field

To account for electromechanical coupling on the compression-induced instability, we substitute equations in (28) into (24) giving the following differential equation:

$$f'''' + K^2 \left( (\lambda_{pre}^{\upsilon})^4 + 1 \right) f'' + K^4 (\lambda_{pre}^{\upsilon})^4 f = 0. \tag{36}$$

This equation along with the boundary conditions in (25) and (27) gives the second algebraic equation:

$$\begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \end{bmatrix} = 0 \tag{37}$$

The existence of a non-trivial solution requires:

$$\det \mathbf{B} = 0 \tag{38}$$

where the matrix $\det \mathbf{B}$ can be written as a function of dimensionless parameters $\det \mathbf{B} = g(K H_f, \tilde{E} \sqrt{\epsilon/\mu, \gamma/(\mu Hf)})$ and the stretch $\lambda_{pre}^{\upsilon}$ is a prescribed constant and where $\tilde{E} = \Phi/H_f$ is the nominal
electric field. The explicit expression of the matrix $\mathbf{B}$ is:

$$B_{11} = -\frac{(\lambda^{pre})^2 \Phi^2 e}{\mu H_f^2} - \frac{\gamma H_f K^2}{\mu (\lambda^{pre})^2} - 2KH_f$$

$$B_{12} = -\frac{(\lambda^{pre})^2 \Phi^2 e}{\mu H_f^2} - \frac{\gamma H_f K^2}{\mu (\lambda^{pre})^2} + 2KH_f$$

$$B_{13} = -\frac{(\lambda^{pre})^2 \Phi^2 e}{\mu H_f^2} - \frac{\gamma H_f K^2}{\mu (\lambda^{pre})^2}$$

$$B_{14} = -\frac{(\lambda^{pre})^2 \Phi^2 e}{\mu H_f^2} - \frac{\gamma H_f K^2}{\mu (\lambda^{pre})^2}$$

$$B_{21} = B_{22} = -\frac{(\lambda^{pre})^2 KH_f}{(\lambda^{pre})^2}$$

$$B_{23} = B_{34} = -\frac{2KH_f}{(\lambda^{pre})^2}$$

$$B_{31} = (\lambda^{pre})^2 KH_f \left(-e^{-KH_f(\lambda^{pre})}\right)$$

$$B_{32} = (\lambda^{pre})^2 KH_f \left(e^{KH_f(\lambda^{pre})}\right)$$

$$B_{33} = KH_f \left(-e^{-KH_f(\lambda^{pre})}\right), B_{34} = KH_f \left(e^{KH_f(\lambda^{pre})}\right)$$

$$B_{41} = e^{-KH_f(\lambda^{pre})}, B_{42} = e^{KH_f(\lambda^{pre})}$$

$$B_{43} = e^{-KH_f(\lambda^{pre})}, B_{44} = e^{KH_f(\lambda^{pre})}$$

The solution to the eigenvalue problem in (38) gives the critical nominal electric field $\tilde{E}$ for various elastocapillary numbers. For instance, Figure 6 shows the calculated nominal electric-field $\tilde{E}$ for inducing the wrinkling instabilities in DEs for a uniaxial compression of $\epsilon^{pre} = 0.2$. The normalized nominal electric-field first decreases and then increases as the wavelength increases. The lowest electric-field in each curve gives the critical nominal electric-field $\tilde{E}_c$ for wrinkling instability.

We have recovered the analytic result in Wang and Zhao (2013) for wrinkling instability of a film without pre-compression $\epsilon^{pre} = 0$, as shown in Figure 7. The results in Figure 7 also demonstrate that pre-compressing the DE film leads to a significant increase in the electric field that is needed to induce the surface wrinkling instability, i.e. a near doubling of the voltage is needed as the initial compressive strain increases from 0 to 40%. Pre-stretch has been widely observed to decrease the nominal electric field required for pull-in instability (Zhao and Suo 2007), thus compression induces the opposite effect, that of increasing the nominal electric field $\tilde{E}$ required for electromechanical instability.

Increasing the surface tension, and thus the elastocapillary number has a similar effect, in that the critical nominal electric field needed to induce surface wrinkling increases substantially for a given pre-compression as the
elastocapillary number increases. Again, a near-doubling of the critical electric field is needed as the elastocapillary number increases from 1 to 100. These results were verified using the nonlinear FE method of Seifi and Park (2016). To study the surface instability of a pre-compressed film subjected to electric field, we first statically compress a film with length of $L_f = 160$ and height $H_f = 4$ with mesh size $d = 1/16$ to various strains of $\varepsilon_{pre} = 0.1, 0.2, 0.3$ and 0.4, all of which are smaller than Biot’s wrinkling strain $\varepsilon_w = 0.46$. Once the film is compressed, an electric potential on top of the film is applied (Figure 2-b) in conjunction with surface tension $\gamma$. The electric potential $\Phi$ was then increased linearly with time until the surface wrinkling instability occurs, where an illustration of the resulting surface wrinkling instability that is observed is shown in Figure 8. The normalized critical nominal electric field $E_c\sqrt{\varepsilon_H}$ is then measured as soon as the wrinkles appears on the surface. In Figure 7, this critical electric field is plotted using symbols as a function of elastocapillary number $\gamma/({\mu}H_f)$. The FE results are in a good agreement with our perturbation analysis.

Finally, Figure 9 shows that for a given elastocapillary number, the wrinkling wavelength decreases with increasing DE film compressive strain. This trend is opposite from Figure 4, where the wavelength increases with increasing elastocapillary number. This is because the pre-compressive strain $\varepsilon_{pre}$ reduces the surface area. This reduction in surface area also reduces the surface energy, which results in a decrease in the critical wavelength.

4. Conclusions

In conclusion, we have presented a theoretical model augmented with computational analysis to examine the surface instabilities of dielectric elastomers accounting for surface tension effects. Surface tension is found to significantly impact the nature of the surface instabilities in the DEs, both through increasing the electric fields required to induce the instability, as well as in increasing the wavelength of the resulting instability. We note that in the purely mechanical compression of soft materials, creases always occur before wrinkles regardless of the surface tension value (Chen et al., 2013). However, in electromechanically coupled soft materials like DEs, wrinkles occur before creases when the elastocapillary number is larger than one (Wang and Zhao, 2013). Indeed, here we have focused on the analysis for elastocapillary numbers larger than one, as illustrated in Figs. 8 and 9.

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