Supplemental Material for

Creasing-Wrinkling Transition in Elastomer Films under Electric Fields

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Measurement of shear moduli of elastomers

The shear moduli of elastomers were measured by frequency sweep tests using a rheometer (TA instrument, USA). Cross-hatch steel parallel plates with diameter of 8 mm were used for the tests. During the tests, the applied strain was controlled to oscillate from 0 to 0.05. When the cross-linker concentration in Sylgard is lower or higher than 1.2%, the measured storage modulus of the elastomer is correspondingly lower or higher than the loss modulus, and therefore it is in liquid-like or solid-like state, respectively [**Figs. S1(a)-(d)**]. We chose elastomers in solid-like states (*i.e.*, cross-linker concentration higher than 1.2%) for studying the wrinkling-creasing transitions and measuring the critical electric fields of the instabilities [**Fig. S1(e)**]. Since we used quasi-static loading condition in the current paper, we regarded the storage moduli at near-zero frequency as the shear moduli of the elastomer [**Fig. S1(e)**]. The shear moduli of another elastomer Ecoflex were also measured using the same method [**Fig. S1(f)**].
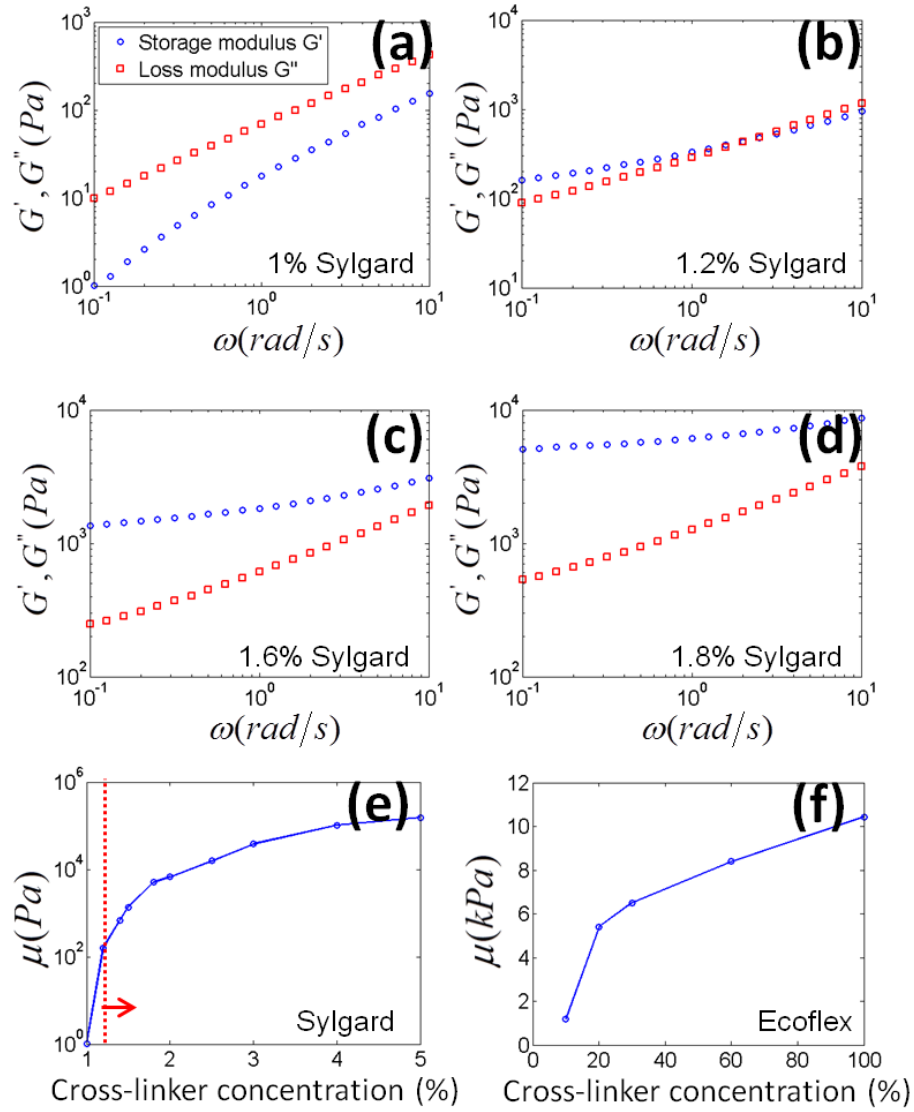
FIG. S1. The measured storage and loss moduli of Sylgard with cross-linker concentrations of 1%(a), 1.2%(b), 1.6%(c) and 1.8%(d). The shear moduli of Sylgard (e) and Ecoflex (f) as functions of cross-linker concentrations.

Analysis of wrinkling instability in elastomers under electric fields

The analysis of the wrinkling instability follows Ref [7]. We consider a region of the elastomer as shown in the inset of **Fig. 2(a)**. For simplicity, the rigid substrate is not included in the current analysis. We assume a voltage Φ is applied on the elastomer film with thickness H. When the elastomer is at the flat state, the electric field in the elastomer film is homogeneous, with the electric field vector as $\mathbf{E} = [0, E]^T$, where $E = \Phi/H$ is the applied electric field.

When the elastomer is at the initial wrinkled state, the surface of elastomer film undergoes a sinusoidal undulation with small amplitude δ. The electric field in the elastomer film can be approximately expressed as $\mathbf{E} = [0, E_\delta]^T$, where $E_\delta = \Phi/(H + \delta)$. The electrical stress in the elastomer can now be written as

$$\boldsymbol{\sigma}_E = \begin{bmatrix} -\dfrac{1}{2}\varepsilon E_\delta^2 & 0 \\ 0 & \dfrac{1}{2}\varepsilon E_\delta^2 \end{bmatrix} \tag{S1}$$

Since δ/H is small, $\varepsilon E_\delta^2/2$ can be linearly approximated as

$$\frac{1}{2}\varepsilon E_\delta^2 = \frac{\varepsilon}{2}\left(\frac{\Phi}{H}\right)^2 - \frac{\varepsilon\Phi^2}{H^2}\frac{\delta}{H} + O\left(\left(\frac{\delta}{H}\right)^2\right) \tag{S2}$$

For small deformation at the wrinkling initiation, the mechanical stress in the elastomer can be linearized as $\boldsymbol{\sigma}_M = \mu(\nabla\mathbf{u} + \nabla^T\mathbf{u}) - p\mathbf{I}$, where $\mathbf{u} = [u_x, u_y]^T$ is the displacement vector and p is the hydrostatic pressure.

Stress equilibrium in the elastomer reads $\nabla\cdot\boldsymbol{\sigma} = 0$, where $\boldsymbol{\sigma} = \boldsymbol{\sigma}_M + \boldsymbol{\sigma}_E$. From **Eqs. (S1)** and **(S2)**, we can approximately obtain $\nabla\cdot\boldsymbol{\sigma}_E = 0$, so the stress equilibrium can be reduced as

$$\mu\nabla^2\mathbf{u} - \nabla p = 0 \tag{S3}$$

Boundary conditions can be written as

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$$\sigma_{M\,yy} + \frac{\varepsilon}{2}E^2 - \frac{\varepsilon E^2}{H}\delta = \gamma u_{y,xx}, \quad \sigma_{M\,yx} = 0 \qquad \text{at } y=H \qquad (S4)$$

$$u_x = u_y = 0 \qquad \text{at } y=0 \qquad (S5)$$

Eqs. (S3-5) constitute a boundary value problem with a homogenous solution and perturbed solutions,

i.e., $\mathbf{u} = \mathbf{u}^h + \mathbf{u}^p$ and $p = p^h + p^p$. The homogenous solution is

$$\mathbf{u}^h = 0, \, p^h = \frac{\varepsilon}{2}E^2 \qquad (S6)$$

Eliminating the homogenous solution, the perturbed solutions are solved as follows.

The incompressibility condition of the elastomer $\nabla \cdot \mathbf{u} = 0$ permits a stream function $\varphi(x,\,y)$ such

that (note $\mathbf{u} = \mathbf{u}^p$)

$$u_x = \frac{\partial \varphi}{\partial y}, \quad u_y = -\frac{\partial \varphi}{\partial x} \qquad . \qquad (S7)$$

We assume the wrinkling instability has sinusoidal morphology; therefore, we are looking for a perturbed

solution for stream function and hydrostatic pressure with the form

$$\varphi(x,y) = \varphi(y)\cos(kx), \, p^p(x,y) = p^p(y)\sin(kx) . \qquad (S8)$$

The governing equation (Eq. S3) can be written as

$$\begin{cases} \mu\left(-k^2\frac{d\varphi(y)}{dy} + \frac{d^3\varphi(y)}{dy^3}\right) - kp^p(y) = 0 \\ \mu\left(-k^3\varphi(y) + k\frac{d^2\varphi(y)}{dy^2}\right) - \frac{dp^p(y)}{dy} = 0 \end{cases} \qquad (S9)$$

with boundary conditions as

$$2\mu k\frac{d\varphi(y)}{dy} = \left(Yk - \gamma k^3\right)\varphi(y) + p^p(y), \text{ and } \frac{d^2\varphi(y)}{dy^2} + k\varphi(y) = 0 \quad \text{at } y=H \qquad (S10)$$

$$\frac{d\varphi(y)}{dy} = \varphi(y) = 0 \qquad \text{at } y=0 \qquad (S11)$$

where $Y = \varepsilon E^2/H$. **Eq. (S9)** can be solved as

$$\varphi(y) = \frac{1}{4}e^{-ky}\left(2 + 2e^{2ky} + ky - e^{2ky}ky\right)c_1 + \frac{e^{-ky}\left(-1 + e^{2ky}\right)}{2k}c_2 + \frac{e^{-ky}\left(-1 + e^{2ky}\right)y}{4k}c_3$$
$$+ \frac{e^{-ky}\left(1 - e^{2ky} + ky + e^{2ky}ky\right)}{4k^2\mu}c_4$$

(S12)

$$p^p(y) = \frac{1}{2}e^{-ky}\left(1 - e^{2ky}\right)k^2\mu c_1 + \frac{1}{2}e^{-ky}\left(-1 + e^{2ky}\right)\mu c_3 + \frac{1}{2}e^{-ky}\left(1 + e^{2ky}\right)c_4$$

(S13)

The boundary conditions [**Eqs. (S10)** and **(S11)**] require

$$\begin{bmatrix} A_1 & A_2 \\ A_3 & A_4 \end{bmatrix}\begin{bmatrix} c_3 \\ c_4 \end{bmatrix} = 0$$

(S14)

$$A_1 = \frac{1}{2}e^{-Hk}\left(1 - Hk + e^{2Hk}\left(1 + Hk\right)\right)$$

(S15)

$$A_2 = \frac{e^{-Hk}\left(1 + e^{2Hk}\right)Hk}{2\mu}$$

(S16)

$$A_3 = \frac{1}{4}e^{-Hk}H\left[2k\mu\left(1 + e^{2Hk}\right) + Y\left(1 - e^{2Hk}\right) + k^2\gamma\left(-1 + e^{2Hk}\right)\right]$$

(S17)

$$A_4 = \frac{e^{-Hk}}{4k\mu}\left[Y\left(-1 + e^{2Hk}\right) - k\left(1 + e^{2Hk}\right)\left(2\mu + HY\right) + k^2\left(-1 + e^{2Hk}\right)\left(2H\mu - \gamma\right) + \left(1 + e^{2Hk}\right)Hk^3\gamma\right]$$

(S18)

The existence of roots requires the determinant of the coefficient matrix in **Eq. (S14)** to be equal

to zero. The resultant equation for the electric fields that satisfy the equilibrium condition is

$$\frac{\varepsilon E^2}{\mu} = 2Hk\frac{1 + 2e^{2Hk} + e^{4Hk} + 4e^{2Hk}H^2k^2}{-1 + e^{4Hk} - 4e^{2Hk}Hk} + \left(Hk\right)^2\frac{\gamma}{\mu H}$$

(S19)

As illustrated in **Fig. 2(a)**, the normalized electric field $E/\sqrt{\mu/\varepsilon}$ first decreases and then increases Hk,

and the minimum electric field that satisfies **Eq. (S19)** gives the critical electric field $E_{wrinkle}^c$ for the

wrinkling instability.

Next, we prove that the viscoelastic effect of the elastomer film is not necessary to be considered

to calculate the critical electric fields for the wrinkling instability. If the elastomer film is considered as a

viscoelastic solid with a constitutive law written as

$$\boldsymbol{\sigma}_M = 2\mu\mathbf{e} + 2\eta\dot{\mathbf{e}} - p\mathbf{I} \tag{S20}$$

where $\mathbf{e} = (\nabla\mathbf{u} + \nabla^T\mathbf{u})/2$ and η is the viscosity of the elastomer. We can repeat the above analysis by assuming

$$\varphi(x,y,t) = \varphi(y)\cos(kx)e^{\omega t}, \; p(x,y,t) = p(y)\sin(kx)e^{\omega t} \tag{S21}$$

where ω is the growth rate of the wrinkle amplitude. Therefore, we obtain the equivalent equation for **Eq. (S19)** as

$$\omega\eta + \mu = \frac{-1 + e^{4Hk} - 4e^{2Hk}Hk}{1 + 2e^{2Hk} + e^{4Hk} + 4e^{2Hk}H^2k^2}\frac{Y - k^2\gamma}{2k} \tag{S22}$$

which only replaces μ in **Eq. (S19)** with $\omega\eta + \mu$. For viscoelastic elastomers under electric fields, the critical electric field is the minimum electric field that satisfies zero growth rate of the wrinkling amplitude, i.e., $\omega = 0$ [7, 8]. Therefore, the critical electric field calculated from **Eq. (S22)** for a viscoelastic elastomer is the same as the one from **Eq. (S19)** for the corresponding elastic elastomer.

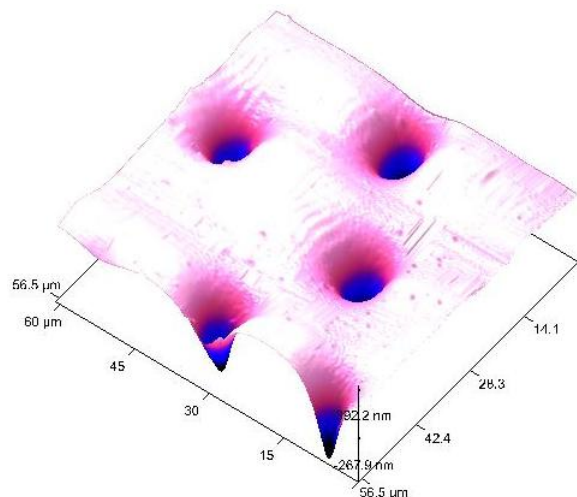## An evidence of the concave pattern of creasing instability

FIG. S2. Surface topography of a highly deformed Sylgard layer by electric field. A 2% Sylgard films with a thickness ~10 μm was subjected to an electric field much higher than the critical electric field for the creasing instability. As a result, the electric field highly deforms the polymer film after the creasing instability, inducing a permanently damaged pattern of concave craters. The pattern of craters was characterized by AFM, once the electric field was withdrawn.

Critical sizes of defects to nucleate creases under critical electric fields
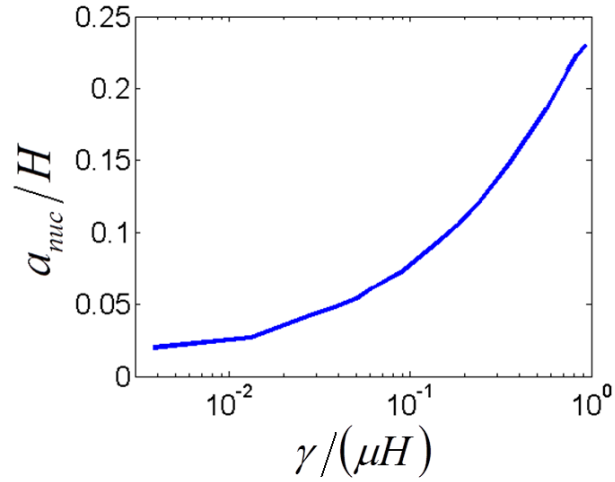

FIG. S3. The calculated critical sizes of defects to nucleate creases under critical electric fields. Elastomers with relatively high values of $\gamma/(\mu H)$ require defects of relatively large sizes to nucleate creases under the critical electric field. If the defects are not present on elastomer surfaces in the experiments, a higher applied electric field than the calculated critical field is required to initiate creases. Therefore, the theory may underestimate the observed electric fields for creasing instability in elastomers with relatively high values of $\gamma/(\mu H)$.

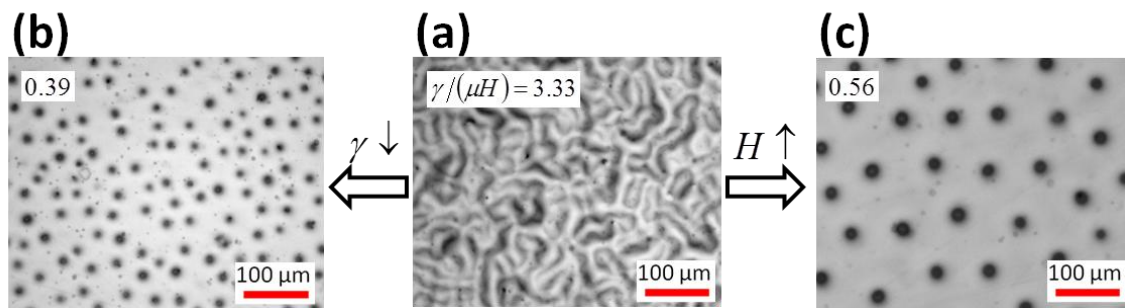## Creasing-wrinkling transition in Ecoflex

FIG. S4. Wrinkling-creasing instability transitions in Ecoflex films. Wrinkling instability is observed in an Ecoflex film with $\gamma = 0.04\,N/m$, $H = 10 \pm 2\mu m$, and $\mu \approx 1.2 kPa$ (a). By decreasing surface energy to $\gamma = 0.0047 N/m$ (b) or by increasing the film thickness to $H = 59 \pm 3\mu m$ (c), the creasing instability occurs instead.

Supplemental video

Video S1. The evolution of surface morphology of a Sylgard film subject to a DC voltage with a ramping rate of 10Vs^{-1}. The cross-linker concentration of the film is 1% [Fig. S1(a)] and the thickness is $10\pm2\mu m$.