## Supplementary material for "Creasing to cratering instability in polymers under ultrahigh electric fields"

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**Figure s1.** The shear modulus of the PDMS elastomer as a function of the crosslinker concentration (a), and the PDMS-film thickness as a function of the spin speed for PDMS with various crosslinker concentrations (b). The shear moduli were measured by uniaxial tensile tests with a Micro-Strain Analyzer (TA Instruments, USA) under a loading rate  $5 \times 10^{-5}$  s<sup>-1</sup>. The film thicknesses were measured by Dektak 150 Stylus Profiler (Bruker AXS, USA).



**Figure s2.** The deformation and electric potential distribution in a block of a polymer subject to an applied voltage. The model is taken to deform under plain-strain conditions. An analytical relation between the deformation and the applied voltage can be derived as  $\Phi/(H\sqrt{\mu/\varepsilon}) = \sqrt{1-(h/H)^4}$ . The result from the finite element model matches well with the analytical solution.



Figure s3. We perturb the surface of the flat film with a sinusoidal deflection  $\delta = \xi \sin kx$ . The film is taken to be incompressible, so that the change in the elastic energy from the flat state to the perturbed state per unit thickness of the region is [28]

$$\Delta \Pi_{elastic} = \int_0^{2n\pi/k} \mu \frac{\delta^2}{H} dx$$
 (s1)

The change in the electrostatic potential energy per unit thickness of the region is [28]

$$\Delta \Pi_{electric} = \int_{0}^{2n\pi/k} \left[ -\frac{\varepsilon \Phi^2}{2(H-\delta)} + \frac{\varepsilon \Phi^2}{2H} \right] dx$$
(s2)

To the leading order of the perturbation amplitude, the changes in the potential energy is

$$\Delta \Pi = \Delta \Pi_{elastic} + \Delta \Pi_{electric} = \frac{n\pi\xi^2}{Hk} \left[ \mu - \frac{\varepsilon}{2} \left(\frac{\Phi}{H}\right)^2 \right]$$
(s3)

Setting Eq. (s3) to zero, the critical electric field can be calculated as  $E_c = \sqrt{2\mu/\varepsilon}$ .

**Video s1.** Video segments illustrating the evolution of instability structures in a substrate-bonded PDMS film subject to a ramping voltage with a rate of  $10Vs^{-1}$ .