

Mechanical constraints enhance electrical energy densities of soft dielectrics

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(Received 11 September 2011; accepted 30 September 2011; published online 25 October 2011)

Dielectrics are essential components in modern electronics and electric systems. When a sufficiently high voltage is applied on a layer of a dielectric, the dielectric will breakdown electrically. The breakdown limits the electrical energy density of the dielectric. We show that constraining the deformation of soft dielectrics can greatly enhance their breakdown electric fields and thus increase their electrical energy densities. The mechanical constraints suppress electromechanical instabilities, a major cause for electrical breakdowns in soft dielectrics. © 2011 American Institute of Physics. [doi:10.1063/1.3655910]

Soft dielectrics, most based on polymers and polymer gels, have been intensively studied in recent years due to their unique electrical and mechanical properties including high dielectric strength, fast charging rate, and mechanical flexibility and conformability. Soft dielectrics have been widely used as insulating matrix, organic capacitors, polymer actuators, generators, and refrigerators in various electrical devices and systems.^{1–7} In particular, intensive experimental and theoretical efforts have been focused on developing materials as soft dielectrics with high electric energy densities.^{8–10} In this letter, we propose a drastically different approach to achieve high-energy-density soft dielectrics. We show that the electrical energy densities of existent soft dielectrics can be significantly enhanced by proper mechanical constraints of the dielectrics.

The electrical energy density of a dielectric can be expressed as $u = \int DdE$, where E and D are the electric field and electric displacement of the dielectric. Electrical breakdown of the dielectric limits the maximum electrical energy density it can achieve, i.e., $u_{\max} = \int_0^{E_B} DdE$, where E_B is the breakdown electric field. Soft dielectrics usually behave as ideal dielectrics,¹¹ where E and D are linearly related by a dielectric constant ϵ . For ideal dielectrics, the maximum electrical energy density can be calculated as

$$u_{\max} = \epsilon E_B^2 / 2. \quad (1)$$

Previous studies have shown that electromechanical instabilities are one major cause for electrical breakdowns in soft dielectrics.^{8,12–15} Figure 1(a) illustrates one mode of electromechanical instability, the pull-in instability.¹² As a voltage is applied on a layer of a soft dielectric sandwiched between two compliant electrodes such as carbon grease or metal films, the soft dielectric reduces its thickness, so that the same voltage induces a higher electric field. The positive feedback may cause some region of the dielectric to thin down drastically, resulting in the electrical breakdown.^{12,13} Figure 1(b) illustrates another mode of electromechanical instability, the electro-creasing instability.¹⁶ As a voltage is

applied on a layer of a soft dielectric with compliant electrode on one side but mechanically constrained by a rigid electrode (e.g., carbon epoxy) on the other side, compressive stresses develop in the soft dielectric parallel to the layer. As the compressive stresses increase with the applied voltage, the deformable surface of the soft dielectric can fold against itself to form a crease. The electric field around the tip of the crease increases drastically, causing electrical breakdown.¹⁶

Since the electromechanical instabilities induce electrical breakdowns of soft dielectrics, the breakdown fields of the dielectrics are approximately the same as the critical fields for the instabilities, i.e., $E_C \approx E_B$. From previous studies, the critical electric field for the electromechanical instabilities can be expressed as^{8,12–14,16}

$$E_C = Z \sqrt{\frac{K}{\epsilon}}, \quad (2)$$

where K is a measurement of the stiffness of the polymer (e.g., Young's modulus or shear modulus) and Z a non-dimensional factor. The value of Z depends on the mode of the instability and the type of stiffness measurement. For example, if the polymer follows the neo-Hookean law with a shear modulus K , the value of Z is ~ 0.69 for the pull-in instability¹³ and ~ 1.03 for the electro-creasing instability.¹⁶ By substituting Eq. (2) into Eq. (1), we can get the maximum electrical energy density of an ideal soft dielectric that breaks down due to electromechanical instabilities,

$$u_{\max} = Z^2 K / 2. \quad (3)$$

Equation (3) indicates that the electrical energy density of a soft dielectric increases with its rigidity. This trend is consistent with previous experimental observations.^{8,12,16–18} However, it is highly desirable to increase energy densities of soft dielectrics while still maintaining their mechanical flexibility. Therefore, we propose to decouple u_{\max} and K by suppressing the electromechanical instabilities, i.e., to make $u_{\max} > Z^2 K / 2$.

As illustrated on Figs. 1(a) and 1(b), the electromechanical instabilities require deformation of the dielectrics. Based on this fact, we hypothesize that the electrical

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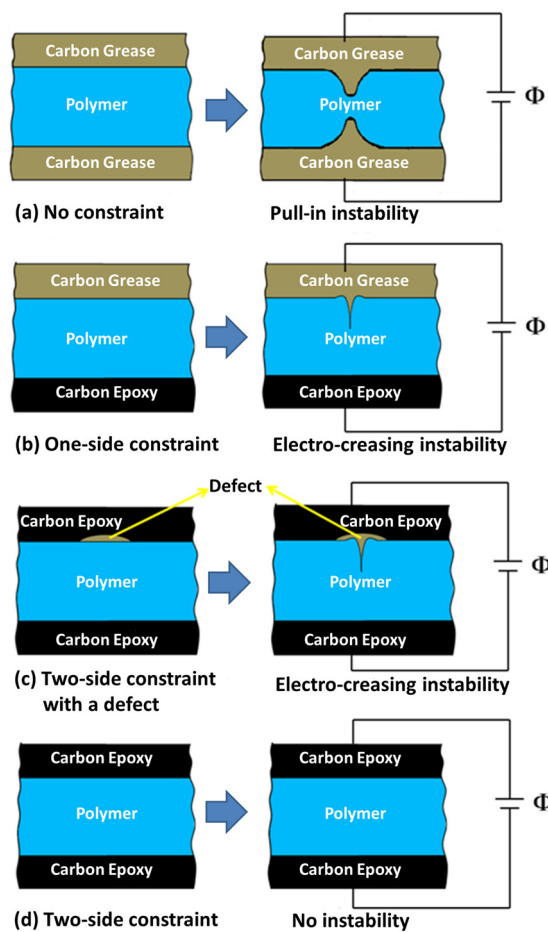


FIG. 1. (Color online) A film of a soft dielectric is under four types of mechanical constraints from electrodes: no constraint (a), one-side constraint (b), two-side constraint with a defect (c), and two-side constraint (d).

breakdown fields and energy densities of soft dielectrics can be significantly enhanced by constraining the deformation of the dielectrics. To test the hypothesis, we chose three types of widely used soft dielectrics: VHB tapes (3 M Inc.), Sylgard 527 silicone dielectric gel (Dow Corning Inc.), and Ecoflex silicone rubber (Smooth-on Inc.). The Sylgard 527 and Ecoflex were fabricated into thin films with a thickness of $200\ \mu\text{m}$ using the spin-coating method,^{16,18} and VHB tapes of $500\ \mu\text{m}$ were used as received. The films of soft dielectrics follow the neo-Hookean law at moderate deformation,^{15,16,19} and their shear moduli were measured by uniaxial tensile tests with a strain rate of $2.5 \times 10^{-4}\ \text{s}^{-1}$. The dielectric constants of soft dielectrics were taken from their data sheets.²⁰

The films of soft dielectrics were subject to different types of mechanical constraints by using either carbon grease or carbon epoxy as electrodes. The carbon grease deforms freely with the dielectric, but attaching a thick layer ($>1\ \text{mm}$) of carbon epoxy can mechanically constrain the surface of the film. Four types of mechanical constraints were employed: no constraint [Fig. 1(a)], one-side constraint [Fig. 1(b)], two-side constraint [Fig. 1(d)], and two-side constraint with a defect on the dielectric-carbon epoxy interface [Fig. 1(c)]. The defect was approximated by a spot of carbon grease on the interface [Fig. 1(c)]. The diameter of the electrodes is $8\ \text{mm}$ and the diameter of the defect is $\sim 1.5\ \text{mm}$.

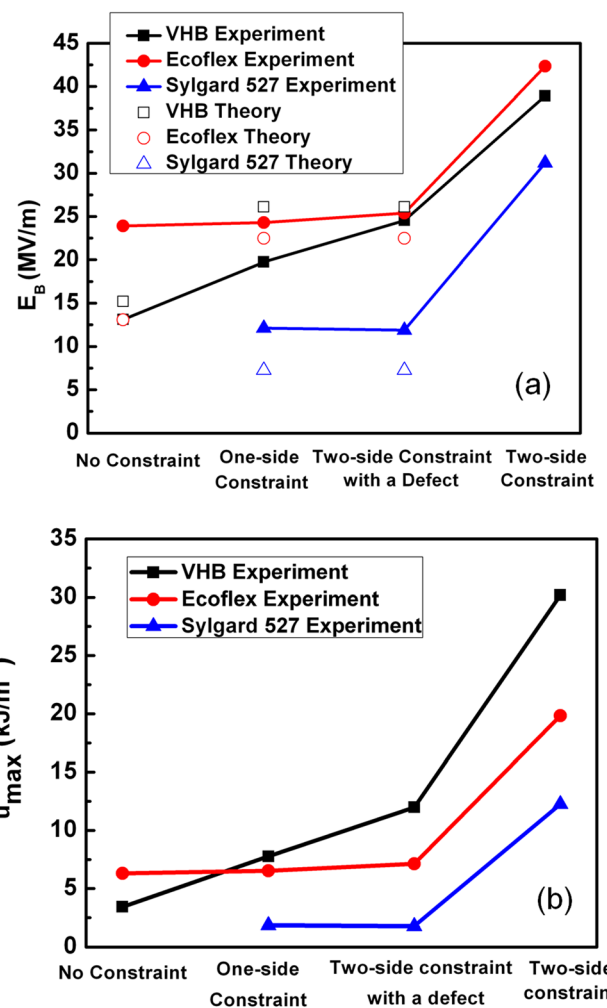


FIG. 2. (Color online) The breakdown fields (a) and electric energy densities (b) of soft dielectrics under various types of mechanical constraints.

A direct-current voltage was applied between the electrodes on the dielectric film by a high voltage supply (Matsusada, Japan) with a ramping rate of $500\ \text{V/s}$. Once the electric breakdown occurs, the voltage was recorded as Φ_B and the breakdown field was calculated as Φ_B/H , where H is the thickness of the film at the undeformed state. Here, it is assumed that the thickness of the dielectric film does not vary with the applied voltage, since the film-thickness changes in our experiments are observed to be less than 10%. The assumption may underestimate the breakdown fields of no-constraint films,¹³ but it has been widely used in calculating breakdown fields of dielectrics^{8,12,17} and it will not affect the physical pictures of the paper. For each soft dielectric under each type of mechanical constraint, at least 20 tests were carried out and the breakdown field was evaluated using the two-parameter Weibull analysis (e.g., Fig. S1).^{21,22}

As shown on Fig. 2(a), for each soft dielectric, the breakdown fields for no-constraint and one-side-constraint films are significantly lower than that of two-side-constraint film. This is due to the electrical breakdown induced by pull-in instability in no-constraint films [Fig. 1(a)] and by electro-creasing instability in one-side-constraint films [Fig. 1(b)]. On the other hand, the deformation and instabilities have been suppressed in the two-side-constraint films [Fig. 1(d)],

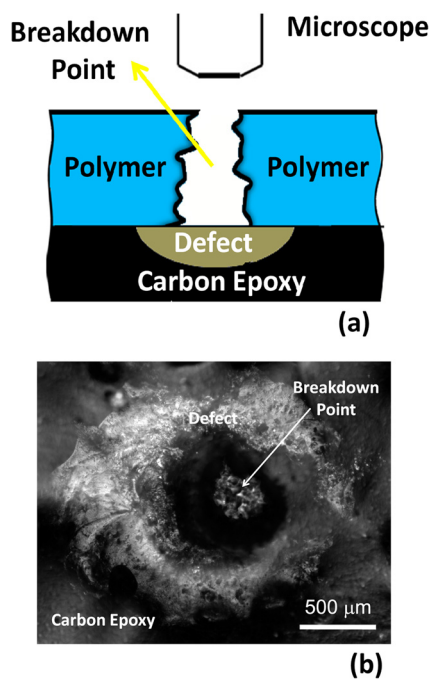


FIG. 3. (Color online) The surface of a soft dielectric film with two-side constraint and a defect after electrical breakdown.

which gives much higher breakdown fields. For example, the breakdown field for two-side-constraint VHB is 3 times of that for no-constraint VHB. Furthermore, the two-side-constraint films with defects have similar breakdown fields as those of one-side-constraint films [Fig. 2(a)], because the defects can also allow electro-creasing instability as illustrated on Fig. 1(c). After electrical breakdown, we further imaged the two-side-constraint films with defects to identify the breakdown locations. As shown on Fig. 3, we found that the electrical breakdown mostly occurs around the defects, regardless of the location of the defects (e.g., edge or center of the electrode). This is because the defect allows deformation of the film beneath it and thus accommodates the electro-creasing instability.

In addition, the critical electric fields for pull-in and electro-creasing instabilities have been calculated using Eq. (2) and plotted on Fig. 2(a). It can be seen that the theoretical values are on the same order as the measured instability-induced breakdown fields. The inconsistency may be due to random factors such as defects in the samples and experimental errors. On the other hand, the measured breakdown fields for two-side-constraint films are significantly higher than the theoretical critical fields. We further compare the breakdown fields of the soft dielectrics reported from data sheets²⁰ to those of two-side-constraint films in Table I. It is remarkable that the breakdown fields for two-side-constraint films are at least 2 times higher than the values from the data sheets.²⁰ These experimental and theoretical results prove that proper mechanical constraints can significantly enhance the breakdown fields of soft dielectrics by suppressing electromechanical instabilities.

TABLE I. Comparison of breakdown fields of two-side-constraint soft dielectrics and values reported from data sheets.

Breakdown field	VHB (MV/m)	Ecoflex (MV/m)	Sylgard 527 (MV/m)
Reported from data sheets	18	13.78	15
Two-side constraint	38.94	42.35	31.18

The electrical energy densities of soft dielectrics under various types of mechanical constraints were calculated with Eq. (1) and plotted in Fig. 2(b). It is evident that the two-side constraints significantly enhance the electrical energy densities of soft dielectrics. For example, the electrical energy density of VHB can be increased by 9 times by the two-side constraint, which suppresses the electromechanical instabilities.

In summary, we show that electromechanical instabilities including pull-in and electro-creasing instabilities is a major cause for electrical breakdowns of soft dielectrics. Proper mechanical constraints can greatly enhance the electrical breakdown fields and energy densities of soft dielectrics by suppressing the instabilities. We expect that the breakdown fields and energy densities may be further increased by increasing the adhesion strength between the soft dielectrics and rigid electrodes.

Funding for this research was provided by the NSF's Research Triangle MRSEC (DMR-1121107) and Pratt School of Engineering at Duke University.

¹L. A. Dissado and J. C. Fothergill, *Electrical Degradation and Breakdown in Polymers* (Peter Peregrinus Ltd, London, 1992), Vol. 11.

²H. G. Yaworski, R. Bukovnik, and IEEE, in *2001 IEEE/PES Transmission and Distribution Conference and Exposition, Vols 1 and 2: Developing New Perspectives* (2001), p. 837.

³B. J. Chu, X. Zhou, K. L. Ren, B. Neese, M. R. Lin, Q. Wang, F. Bauer, and Q. M. Zhang, *Science* **313**, 334 (2006).

⁴Q. M. Zhang, V. Bharti, and X. Zhao, *Science* **280**, 2101 (1998).

⁵R. Pelrine, R. Kornbluh, Q. B. Pei, and J. Joseph, *Science* **287**, 836 (2000).

⁶S. J. A. Koh, X. H. Zhao, and Z. G. Suo, *Appl. Phys. Lett.* **94**, 262902 (2009).

⁷B. Neese, B. Chu, S.-G. Lu, Y. Wang, E. Furman, and Q. M. Zhang, *Science* **321**, 821 (2008).

⁸X. Zhou, X. H. Zhao, Z. G. Suo, C. Zou, J. Runt, S. Liu, S. H. Zhang, and Q. M. Zhang, *Appl. Phys. Lett.* **94**, 162901 (2009).

⁹X. H. Zhao and Z. G. Suo, *Phys. Rev. Lett.* **104**, 178302 (2010).

¹⁰G. Finis and A. Claudi, *IEEE Trans. Dielectr. Electr. Insul.* **14**, 487 (2007).

¹¹X. H. Zhao, W. Hong, and Z. G. Suo, *Phys. Rev. B* **76**, 134113 (2007).

¹²K. H. Stark and C. G. Garton, *Nature* **176**, 1225 (1955).

¹³X. H. Zhao and Z. G. Suo, *Appl. Phys. Lett.* **91**, 061921 (2007).

¹⁴X. H. Zhao and Z. G. Suo, *Appl. Phys. Lett.* **95**, 031904 (2009).

¹⁵J. S. Plante and S. Dubowsky, *Int. J. Solids Struct.* **43**, 7727 (2006).

¹⁶Q. M. Wang, L. Zhang, and X. H. Zhao, *Phys. Rev. Lett.* **106**, 118301 (2011).

¹⁷J. Blok and D. G. LeGrand, *J. Appl. Phys.* **40**, 288 (1969).

¹⁸Q. Wang, M. Tahir, L. Zhang, and X. Zhao, *Soft Matter* **7**, 6583 (2011).

¹⁹X. Zhao, S. J. A. Koh, and Z. Suo, *Int. J. App. Mech.* **3**, 203 (2011).

²⁰Date sheets of VHB tapes, Sylgard 527, and Ecoflex.

²¹See supplementary material at <http://dx.doi.org/10.1063/1.3655910> for calculating the breakdown electric field.

²²X. Zhou, B. Chu, B. Neese, M. Lin, and Q. M. Zhang, *IEEE Trans. Dielectr. Electr. Insul.* **14**, 1133 (2007).

**Supplementary material for “Mechanical Constraints Enhance Electrical Energy Densities
of Soft Dielectrics”**

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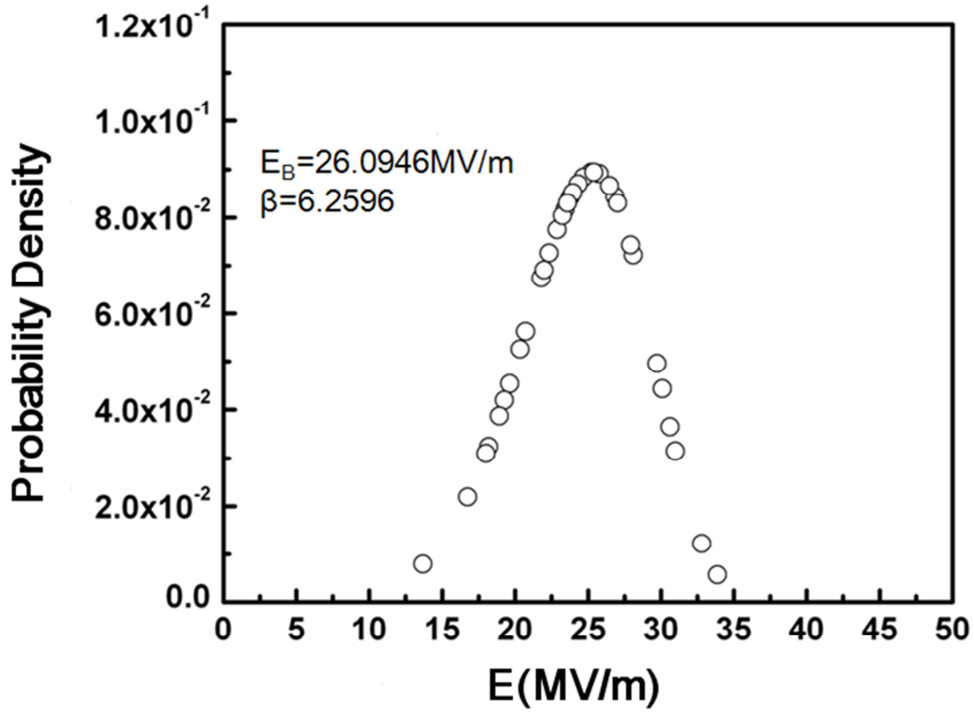


Figure S1. The characteristic breakdown fields of the dielectric films were evaluated by the two-parameter Weibull analysis,

$$P_f = 1 - \exp\left[-\left(E / E_B\right)^\beta\right] \quad (\text{s1})$$

where P_f is the probability of the film to breakdown below an electric field E , E_B the characteristic breakdown field of the film, and β a shape parameter. We plot the probability density as a function of the measured breakdown field for two-side constraint Ecoflex with a defect. A characteristic breakdown field can be fitted to be 25.38MV/m.