Applied Physics Letters

The thickness and stretch dependence of the electrical breakdown strength of an acrylic dielectric elastomer

Cite as: Appl. Phys. Lett. **101**, 122905 (2012); https://doi.org/10.1063/1.4754549
Submitted: 19 June 2012 • Accepted: 07 September 2012 • Published Online: 21 September 2012

Jiangshui Huang, Samuel Shian, Roger M. Diebold, et al.

View Online Export Citation

ARTICLES YOU MAY BE INTERESTED IN

A review on dielectric elastomer actuators, technology, applications, and challenges
Journal of Applied Physics **104**, 071101 (2008); https://doi.org/10.1063/1.2981642

The dielectric breakdown limit of silicone dielectric elastomer actuators
Applied Physics Letters **104**, 052905 (2014); https://doi.org/10.1063/1.4863816

Method to analyze electromechanical stability of dielectric elastomers
Applied Physics Letters **91**, 061921 (2007); https://doi.org/10.1063/1.2768641

AIP Publishing

© 2012 American Institute of Physics.

The thickness and stretch dependence of the electrical breakdown strength of an acrylic dielectric elastomer

Jiangshui Huang,[1] Samuel Shian,[1] Roger M. Diebold,[1,2] Zhigang Suo,[1,a)] and David R. Clarke[1,a)]
[1]*Harvard School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA*
[2]*Materials Department, University of California, Santa Barbara, Santa Barbara, California 93106, USA*

(Received 19 June 2012; accepted 7 September 2012; published online 21 September 2012)

The performance of dielectric elastomer actuators is limited by electrical breakdown. Attempts to measure this are confounded by the voltage-induced thinning of the elastomer. A test configuration is introduced that avoids this problem: A thin sheet of elastomer is stretched, crossed-wire electrodes are attached, and then embedded in a stiff polymer. The applied electric field at breakdown, E_B, is found to depend on both the deformed thickness, h, and the stretch applied, λ. For the acrylic elastomer investigated, the breakdown field scales as $E_B = 51\, h^{-0.25}\, \lambda^{0.63}$. The test configuration allows multiple individual tests to be made on the same sheet of elastomer. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4754549]

Thin dielectrics are widely used to provide electrical insulation for a wide variety of electronic and electrical devices, often integrated into the device structures themselves. The most notable examples of this are the nanometer-scale silica dielectric layer in CMOS devices and the silica-carbon-oxy-fluoride glasses used to insulate the metal interconnect structures distributing power and signals in silicon devices. Thicker dielectrics, such as sleeves and sheaths, are used to insulate wires and electrical cables; for instance, high-density polyethylene is usually the material of choice for high voltage electric power cables. Although these and other insulating dielectric materials are very different and the mechanisms of failure may not be the same, it has been known for many years that there is a general empirical relationship between the breakdown field, E_B, and the thickness, h, of the dielectric, $E_B \propto h^{-n}$.[1–9] This relationship fits data for amorphous solids, such as silica glass and polymers, semi-crystalline polymers as well as a wide range of crystalline ceramics, including polycrystalline and single crystal alumina[10] although the values of n are not unique. The majority of insulating solids are stiff and do not deform appreciably in response to the Coulomb attraction force between oppositely charged electrodes when an electric field is applied across them. This absence of deformation is not the case for soft elastomers and indeed their change in thickness with an applied voltage has been exploited to create electric field-induced actuators.[11–22] Very large actuation strains can be achieved using soft elastomer dielectrics but they must first be biaxially stretched. It is now understood that pre-straining is necessary to forestall an electromechanical instability mode associated with dielectric thinning identified by Stark and Garton.[23] They showed that as the voltage across an elastomer is increased, it thins and, in doing so, the electric field increases and that this coupled positive feedback process continues until the local breakdown field is attained. Prestretching stiffens the dielectric and may even remove the electromechanical instability.[14] Prestretching also increases the breakdown field.[24–26] For instance, for the most widely studied dielectric elastomer, the acrylic elastomer produced by 3M corporation, VHB, it has been reported that the breakdown electric field increases from 18 to 218 MV/m when equi-biaxially stretched up to 6 times.[25] Since the maximum actuation strains for soft dielectric actuators are limited by electrical breakdown, the effect of electrical breakdown on thickness is of particular practical importance. Furthermore, from a practical perspective, the thinning in response to an applied voltage poses a challenge to how to measure the electrical breakdown field.

The traditional method of making electrical breakdown measurements is to use large area electrodes, much larger than the dielectric thickness, and ramp up the voltage until breakdown occurs.[2,3] This is the method that has previously been used to evaluate the breakdown field of elastomers.[24–26] The other method consists of a flat electrode on one side of the dielectric and a curved electrode on the other, with a curvature typically large compared to the dielectric thickness.[27] In both test geometries, the electrodes are generally free to displace towards one another when the voltage is increased unless the electrodes are very stiff. Indeed, in some cases such as the curved electrode configuration test, the curved electrode is pressed into contact with the dielectric to ensure good electrical contact.

In this contribution, we describe an alternative testing geometry that prevents displacements of the electrodes as the voltage is increased and use it to determine breakdown data for the commonly investigated dielectric acrylic elastomer, VHB. Based on preliminary experiments, our experiments are designed to separate the effects of thickness and pre-stretch. Elastomer sheets of two starting thicknesses were investigated, one 0.5 mm in thickness (VHB 4905) and the other 1.0 mm thick (VHB 4910) (both sheets have the same polymer composition; the last two numbers in the notation refer to the sheet thickness). A bi-layer, also 1.0 mm thick, was produced by pressing two sheets of VHB 4905 together. Stretch is defined as the ratio of the length after

a)Authors to whom correspondence should be addressed. Electronic addresses: suo@seas.harvard.edu and clarke@seas.harvard.edu.

 © 2012 American Institute of Physics

stretching to the original length of the elastomer. The stretches were all equi-biaxial and are denoted by λ (Fig. 1). The elastomer is taken to be incompressible, so that its deformed thickness is related to its stretch by the relationship $h = H/\lambda^2$, where h is the deformed thickness of the sheet and H is the initial thickness.

Our electrical test arrangement is shown in Figure 1. It consists of an elastomer sheet with an array of parallel cylindrical wires on either side, all embedded by casting a stiff polymer around them. There are two main features of the test configuration: the elastomer is prevented from thinning and changing dimensions by the stiff polymer block in which the elastomer is embedded, and the wires encapsulated in the stiff polymer on either side of the elastomer are arranged at right angles to one another. A further feature of the test arrangement is that the elastomer sheet can be pre-stretched biaxially prior to the placement of the wire electrodes and subsequent embedding of the elastomer. The polymer blocks on either side of the elastomer sheet were made from a two component polyurethane mixture (Smooth Cast 300, Smooth-On) that, once cross-linking had initiated, was cast around the stretched elastomer to form a stiff plastic layer about 3 mm in thickness after fully curing. The cast polymer is not only much thicker than the VHB elastomer but also orders of magnitude stiffer (Young's modulus = 1.0 GPa versus 1.8 MPa of the VHB according to 3M datasheet). After allowing the polymer to fully cure, it was removed from the mold and electrical connection was made to the individual wires. The electrodes consisted of high purity (99.99%) copper wires, with a diameter of 18 μm (Semiconductor Packaging Materials). The array of parallel, crossed wires facilitates making a series of breakdown measurements all at the same elastomer thickness and stretch, and on the same sample. The arrangement allows for a large number of individual tests to be made, and the average breakdown voltages for each sheet thickness and stretch were determined from no fewer than 24 tests on each sample.

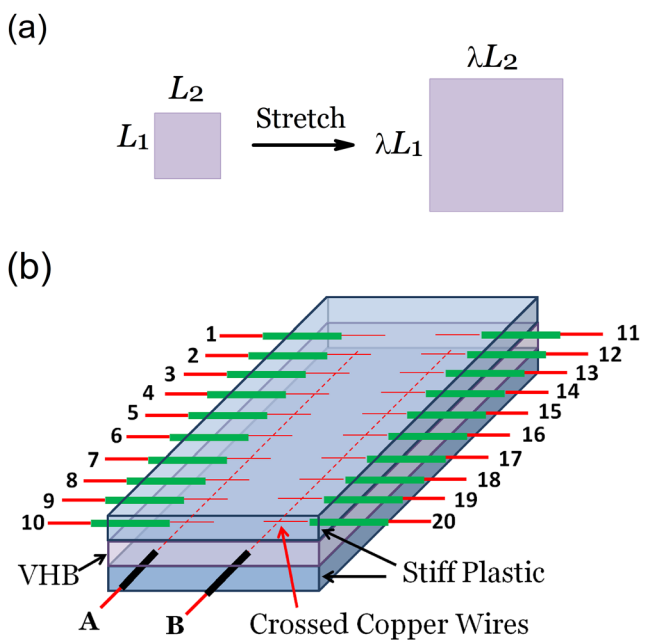

FIG. 1. (a) An elastomer sheet is stretched biaxially by an amount denoted as the stretch ratio λ. (b) Electrical test arrangement consists of a pre-stretched elastomer sheet with an array of parallel cylindrical wires on either side, all embedded by casting a stiff polymer around them. The polymer blocks on either side of the elastomer sheet were made from a two component polyurethane mixture that, once cross-linking had initiated, was cast around the stretched elastomer to form a stiff plastic layer about 3 mm in thickness after fully curing. The electrodes consisted of high purity (99.99%) copper wires with a diameter of 18μm encapsulated in the stiff polymer on either side of the elastomer are arranged at right angles to one another. Electrical connection was made to the individual wires, allowing for a large number of individual tests to be made on the same sheet. For example, 20 tests can be made in the configuration shown in the schematic.

All the measurements were made at room temperature and the test consisted of increasing the applied voltage (Gamma High Voltage Research, model number ES40P-5W-DAM) at a fixed ramp rate of 50 V/s until electric breakdown occurred at a voltage, Φ_B. The breakdown criterion was when the current exceeded the 125 μA pre-set of the voltage source. This was usually accompanied by a spark, apparent visually in many instances, and a "popping" sound. The breakdown paths were conducting afterwards suggesting that thermal breakdown and subsequent carburization occurred before the voltage supply automatically cut-off. The values of the breakdown fields reported here are the engineering breakdown fields, namely the voltage divided by the deformed dielectric thickness

$$E_B = \Phi_B \lambda^2 / H. \quad (1)$$

The test configuration was chosen for a number of reasons. It suppresses deformation of the elastomer under voltage and localizes the electric field more effectively than small pads, decreasing the volume of the dielectric subject to high fields. It also allows a large number of test sites to be addressed without the necessity of precisely having to align small pads and make separate connections to them.

Our measurements show that the average breakdown voltage decreases monotonically with increasing stretch (Fig. 2(a)), consistent with previous measurements of the same elastomer,[24–26] but our numerical values are almost twice as large. We believe that the larger breakdown fields are a direct consequence of the smaller volume of material under electric field in our cross-cylinder test configuration. The existence of a size effect is consistent with previous studies of the breakdown field of several materials, including one on elastomers.[28,29] Amongst the clearest studies of electrode size effects are the recent study of hafnia and silica films which also show that the smaller the contact area, the larger the breakdown field.[30] We also observe a thickness dependence of the electric breakdown field (Fig. 2(b)) but this is less marked than that previously reported. By comparing the breakdown fields in Figure 2(b) for the elastomer sheets of different initial thicknesses, it was found that the breakdown electric fields do not collapse onto a single curve. From this, we conclude that the breakdown field depends not only on the thickness but also on the stretch. This conclusion is substantiated by comparing the electric breakdown field as a function of stretch for the two elastomers of different thickness and a bilayer VHB4905 as is shown in Figure 2(c).

Empirically, we can write that the breakdown field depends on both the deformed thickness and the stretch: $E_B = C h^{-x} \lambda^y$, where C is a constant and x and y are

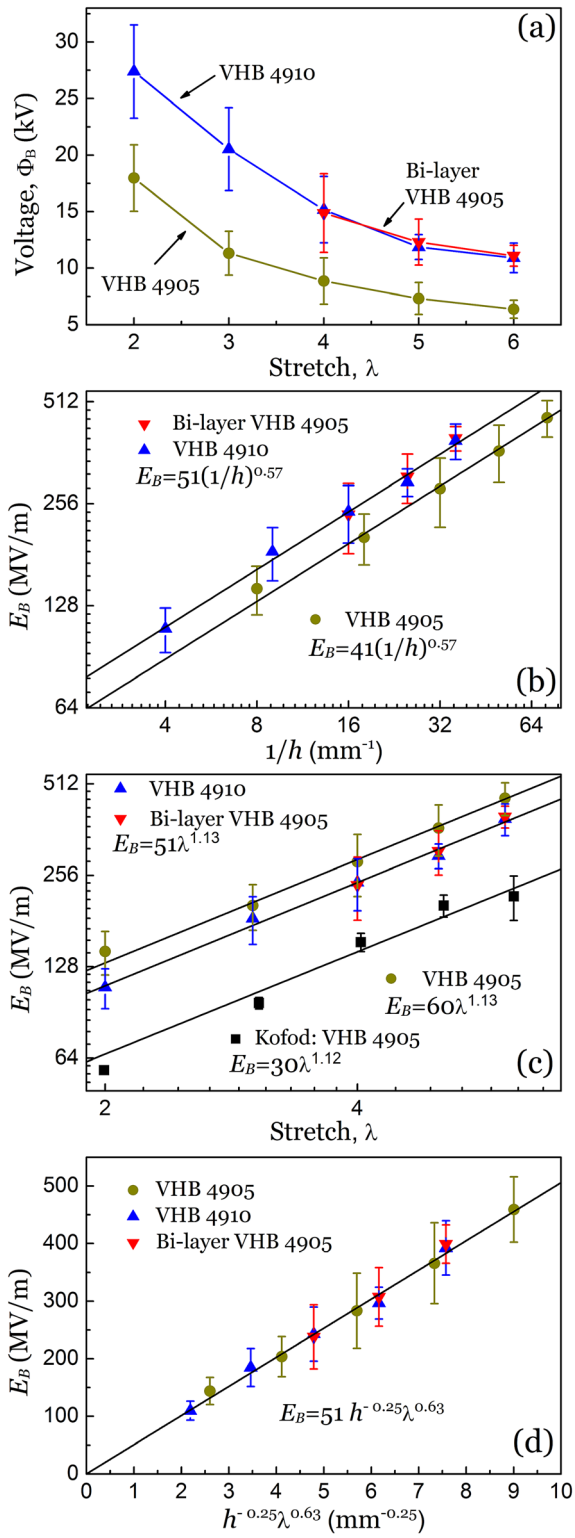

FIG. 2. (a) For each initial dielectric thickness, the breakdown voltages decrease monotonically with increase of stretch. (b) Breakdown electric field as a function of thickness for VHB 4905, two-layer VHB 4905, and VHB 4910 stretched to the values indicated. The lines through the data correspond to a power law dependence on (1/*h*) with an exponent of 0.57. Data for VHB 4910 and two-layer VHB 4905 are fitted to same line. The vertical displacement between the two lines indicates the dielectric strength also depends on the stretch. (c) Breakdown electric field as a function of stretch. The lines through the data correspond to a power law dependence on λ with an exponent of 1.13. Again the difference between the two lines indicates the dielectric strength should depend on the thickness. Data from the work of Kofod[25] are also included and an exponent of 1.12 is obtained by fitting. (d) Experimentally determined scaling between the breakdown electric field and the parameter $(h^{-0.25}\lambda^{0.63})$.

exponents to be determined. All the breakdown data for the elastomer with different thicknesses and values of pre-stretch collapse onto a single scaling as shown in Figure 2(d), corresponding to $E_B = 51\,h^{-0.25\pm0.01}\,\lambda^{0.63\pm0.03}$, where the field is in units of MV/m and the thickness is in mm. In terms of the initial thickness of the dielectric, the breakdown field can alternatively be expressed as $E_B = 51H^{-0.25}\,\lambda^{1.13}$. The stretch exponent, 1.13, is very similar to that, 1.12, based on fitting to Kofod's data[25] (Fig. 2(c)) although the pre-factor is somewhat larger, suggesting that nevertheless the same physical processes are involved.

The observed dependence of the breakdown field on the elastomer pre-stretch suggests a number of possibilities. One is that the dielectric constant, ε, of the elastomer varies with deformation and depends on the pre-stretch. Unfortunately, there is conflicting information in the literature as to whether the dielectric constant or the refractive of the VHB materials depends on pre-stretching. Kofod *et al.* reported that the dielectric constant only slightly dropped with increase of the pre-stretch up to 6, but there is another report that the dielectric constant decreases from its nominal value of 4.7 to 2.6 at a stretch of 5.[25,31] A much smaller reduction, up to 15%, has been reported when a pre-strain is applied.[32,33] As it is expected that breakdown occurs when a critical value of the electric field inside the dielectric, E_B^{int}, is attained and that is related to the applied field, $E_{applied}$, by, $E_B^{\text{int}} = E_{applied}/\varepsilon$, decreasing the dielectric constant by stretching would decrease the applied breakdown field rather than increase it as it is observed. Another possibility is that in stretching the elastomer, the vibrational amplitudes of the individual molecules are reduced and consequently the activation volume is decreased through which mobile charges can migrate under field. Such an activated state description applies to the breakdown of many amorphous polymers, such as high-density polyethylene and polypropylene.[3] However, this form of thermally activated breakdown is usually applicable to breakdown under sustained field conditions rather than short ramping conditions. Finally, we are not in a position to evaluate a third possible breakdown model associated with shape changes due to cavities, such as air bubbles, under field although we believe that our test configuration avoided the entrapment of air bubbles. As long ago as 1964, Garton and Krasucki[4,34] showed that electrical breakdown in an insulating viscous liquid could occur when an air bubble is entrained. Under the influence of the field, the bubble changes shape, elongating in the direction of the field, until breakdown occurs. Essentially, the same mechanism has recently been invoked in soft elastomers containing bubbles by Wang *et al.*[27] However, knowing this work, we took great care to avoid the entrapment of any bubbles. Furthermore, the probability of having a small bubble in each test volume in the crossed cylinder geometry seems highly unlikely. One would also expect that if this was a viable mechanism, bubbles at the interface between the two 0.5 mm sheets of VHB 4905 would have grown and the breakdown fields would be consistently smaller but this is not the case.

In summary, by using a constrained test geometry that prevents thinning in response to an applied geometry and possible electromechanical instability, we have demonstrated that the electric breakdown field of an elastomer sheet

commonly used in dielectric actuators depends on both thickness and biaxial pre-stretch. The breakdown field is found to scale according to simple power law $E_B = 51\ h^{-0.25}\ \lambda^{0.63}$ over the range of thickness and mechanical stretching studied. More detailed studies are necessary to investigate the scaling over a greater range of thicknesses and stretches as well as how stretch affects physical properties including dielectric constant and optical properties. Measurements of the sustained voltage, time, and temperature dependence are also needed to assess voltage creep.

Our research was supported by both the Harvard MRSEC program of the National Science Foundation under Award Number DMR-0820484 and by the Gordon McKay endowment.

[1]A. E. W. Austen and S. Whitehead, Proc. R. Soc. London, Ser A **176**, 33–50 (1940).
[2]S. Whitehead, *Dielectric Breakdown of Solids* (Clarendon, Oxford, 1953).
[3]L. A. Dissado and J. C. Fothergill, *Electrical Degradation and Breakdown in Polymers* (Peter Peregrinus Ltd., London, United Kingdom, 1992).
[4]K. C. Kao, *Dielectric Phenomena in Solids* (Elsevier Academic, San Diego, California, USA, 2004).
[5]F. Forlani and N. Minnaja, Phys. Status Solidi **4**, 311 (1964).
[6]V. K. Agarwal and V. K. Srivastava, Thin Solid Films **8**, 377 (1971).
[7]V. K. Agarwal and V. K. Srivastava, Solid State Commun. **12**, 829 (1973).
[8]B. Helgee and P. Bjellheim, IEEE Trans. Electr. Insul. **26**, 1147 (1991).
[9]H. K. Kim and F. G. Shi, IEEE Trans. Dielectr. Electr. Insul. **8**, 248 (2001).
[10]M. Yoshimura and H. K. Bowen, J. Am. Ceram. Soc. **64**, 404 (1981)
[11]R. Pelrine, R. Kornbluh, Q. Pei, and J. Joseph, Science **287**, 836 (2000).
[12]J. Huang, T. Li, C. C. Foo, J. Zhu, D. R. Clarke, and Z. G. Suo, Appl. Phys. Lett. **100**, 041911 (2012).
[13]J. Huang, T. Lu, J. Zhu, D. R. Clarke, and Z. G. Suo, Appl. Phys. Lett. **100**, 211901 (2012).
[14]T. Lu, J. Huang, C. Jordi, G. Kovacs, R. Huang, D. R. Clarke, and Z. Suo, Soft Matter **8**, 6167–6173 (2012)
[15]G. Kovacs, L. During, S. Michel, and G. Terrasi, Sens. Actuators, A **155**, 299 (2009).
[16]C. Keplinger, M. Kaltenbrunner, N. Arnold, and S. Bauer, Proc. Natl. Acad. Sci. U.S.A. **107**, 4505 (2010).
[17]F. Carpi, G. Frediani, S. Turco, and D. De Rossi, Adv. Funct. Mater. **21**, 4152 (2011).
[18]I. A. Anderson, T. C. H. Tse, T. Inamura, B. M. O'Brien, T. McKay, and T. Gisby, Appl. Phys. Lett. **98**, 123704 (2011).
[19]B. M. O'Brien, T. G. McKay, T. A. Gisby, and I. A. Anderson, Appl. Phys. Lett. **100**, 074108 (2012).
[20]A. T. Conn and J. Rossiter, Smart Mater. Struct. **21**, 035012 (2012).
[21]A. Cheng, "ViviTouchTM offers a new sensory dimension to mobile gaming," WW-EAP Newsletter **13**, 2 (2011).
[22]P. Chouinard and J. Plante, IEEE/ASME Trans. Mechatron. **17**, 857 (2012).
[23]K. H. Stark and C. G. Garton, Nature **176**, 1225 (1955)
[24]J. S. Plante and S. Dubowsky, J. Solids Struct. **43**, 7727 (2006).
[25]G. Kofod, P. Sommer-Larsen, R. Kornbluh, and R. Pelrine, J. Intell. Mater. Syst. Struct. **14**, 787 (2003).
[26]C. Jordi, A. Schmidt, G. Kovas, S. Michel, and P. Ermanni, Smart Mater. Struct. **20**, 075003 (2011).
[27]Q. Wang, M. Tahir, L. Zhang, and X. Zhao, Soft Matter **7**, 6583 (2011).
[28]S. Cygan and J. R. Laghari, IEEE Trans. Electr. Insul. **EI-22**, 835 (1987).
[29]S. Diaham, S. Zelmat, M. L. Locatelli, S. Dinulescu, M. Decup, and T. Lebey, IEEE Trans. Dielectr. Electr. Insul. **17**, 18 (2010).
[30]C. Sire, S. Blonkowski, M. J. Gordon, and T. Baron, Appl. Phys. Lett. **91**, 242905 (2007).
[31]M. Wissler and E. Mazzab, Sens. Actuators, A **138**, 384 (2007).
[32]T. G. Mckay, E. Calius, and I. A. Anderson, Proc. SPIE **7287**, 72870P (2009).
[33]C. Jean-Mistral, A. Sylvestre, S. Basrour, and J.-J. Chaillout, Smart Mater. Struct. **19**, 075019 (2010).
[34]C. G. Garton and Z. Krasucki, Proc. R. Soc. London, Ser. A **280**, 211 (1964).