Stretch dependence of the electrical breakdown strength and dielectric constant of dielectric elastomers

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Abstract. Dielectric elastomers are used for electromechanical energy conversion in actuators and in harvesting mechanical energy from renewable sources. The electrical breakdown strength determines the limit of a dielectric elastomer for its use in actuators and energy harvesters. We report two experimental configurations for the measurement of the stretch dependence of the electrical breakdown strength of dielectric elastomers, and compare the electrical breakdown field for compliant and rigid electrodes on the elastomer. We show that the electrode configuration strongly influences the electrical breakdown field strength. Further we compare the stretch dependent dielectric function and breakdown of the acrylic elastomer VHB 4910™ from 3M™, and ZRUELAST™ 1040™ natural rubber (ZRUNEK rubber technology). While the dielectric permittivity of VHB decreases with increasing stretch ratio, the dielectric constant of rubber is insensitive to stretch. Our results suggest natural rubber as a versatile material for dielectric elastomer energy harvesting.

1. Introduction

Dielectric elastomers proved useful for electromechanical energy conversion [1–3], the latest research frontier is the use of dielectric elastomers in dielectric elastomer generators (DEGs) for the harvesting of mechanical energy, especially from renewable energy sources. In DEGs dielectric elastomers are employed as deformable capacitors by coating the elastomer with compliant electrodes. The performance of DEGs is limited by failure mechanisms such as material rupture, loss of tension, electromechanical instability and electrical breakdown [4–8]. Dielectric breakdown is the ultimate failure mode of any dielectric material - for soft materials the measurement of the breakdown strength is still challenging. Herein we describe two experimental procedures for measuring the breakdown strength of dielectric elastomers, and show that the electrode configuration used strongly influences the breakdown voltages.

The acrylic elastomer VHB (3M) has not only been used in actuators but also in the first DEGs [9]. Since then it has been extensively investigated as model material for both small-scale [10–15] and large-scale energy scavenging [10,16]. However, VHB is far from being an optimal material for DEGs due to its high viscoelastic losses. Natural rubber is the most familiar elastomer; it is cheap, abundant and can be sustainably sourced. In this work we will compare the dielectric function and breakdown strength of VHB 4910 and ZRUELAST natural rubber, and suggest the use of natural rubber for dielectric elastomer generators.

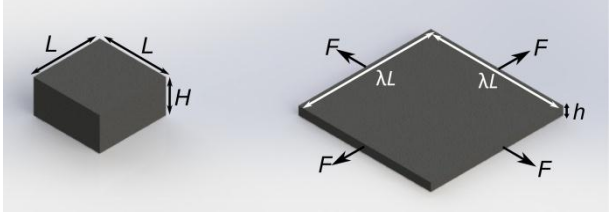


Figure 1: The undeformed membrane with length *L* and thickness *H* (left) is subject to an equal-biaxial force *F* in its plane (right). The membrane's length expands by a factor of λ and its area by a factor of λ^2. The thickness is reduced from *H* to *h* by $1/\lambda^2$.

2. Experimental techniques

Figure 1 shows the concept of a deformable dielectric elastomer capacitor. When the elastomer is stretched biaxially from the initial state with length L to λL in both in-plane directions, it reduces its thickness from H to $h = H * \lambda^{-2}$, due to the incompressibility of the elastomer. Thereby the capacity of the elastomer membrane changes accordingly to λ^4, giving rise to huge capacitance changes when stretched even moderately. Both, in the actuator and generator mode, dielectric elastomers are limited by electrical breakdown.

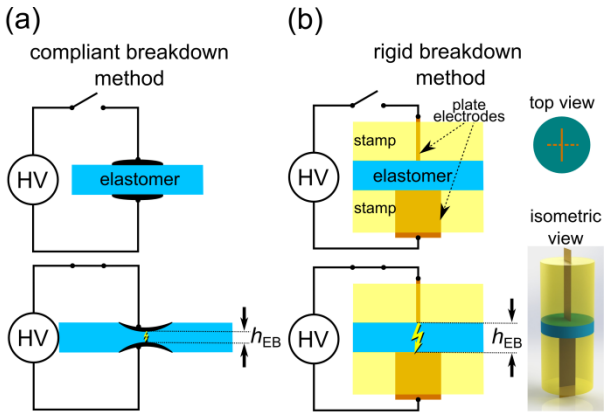


Figure 2: (a) Experimental set-up for breakdown measurements with compliant electrodes. The stretched elastomer membrane is sandwiched between compliant carbon grease electrodes (top). When a high voltage (HV) is applied the membrane reduces its thickness till breakdown occurs (bottom). (b) Experimental set-up with a mechanically clamped elastomer membrane. The elastomer is clamped between two stamp electrodes, which are arranged perpendicular to each other (top and isometric view). The mechanical clamping between the stamps prevents the elastomer from deforming electromechanically under high voltage.

The dielectric strength of soft materials is still under debate. Reported values differ significantly, most likely caused by different experimental conditions employed for breakdown measurements. In this work we use the two experimental arrangements depicted in figure 2 for dielectric breakdown measurements. In the set-up shown in figure 2a compliant electrodes are used. When a voltage is applied in such a configuration the elastomer thins down and in consequence the electric field increases. This positive feedback may cause electromechanical instability and finally breakdown, known as Stark Garton breakdown mechanism in literature [17,18]. In order to suppress the actuation of the elastomer during the breakdown measurement, we clamp the elastomer between two stamp electrodes, as shown in figure 2b. To minimize the probability of an electrical breakdown due to material imperfections it is favorable to utilize small electrode areas. When the elastomer is rigidly clamped between two stamps, the compression of the elastomer is governed by the bulk modulus, rather than the elastic modulus. Since the bulk modulus is some orders of magnitude larger than the elastic modulus, compression of the elastomer is effectively suppressed.

The compliant carbon black electrodes in figure 2a with a diameter of 5 mm diameter allow actuation of the membrane in the electric field. The electrodes in the clamped elastomer configuration of figure 2b consist of brass plates with dimensions of 40 mm x 8 mm x 50 µm embedded in epoxy resin. The stamps are placed on both sides of the sample with the brass electrodes oriented perpendicular to each other, limiting the potential breakdown to the small intersecting area of the electrodes. For both methods the electric field in the membrane is increased with a voltage ramp at a rate of 50 V/s. The applied voltage is increased until electrical breakdown occurs or when a current larger than 0.2 mA is recorded. The highest voltage value is taken as the breakdown voltage.

3. Experimental results

The VHB and ZRUELAST membranes are biaxially stretched with a homebuilt radial 12-arm stretcher. The strain λ is fixed by gluing polyamide rings onto the stretched membranes. Successively the thickness of the samples is measured using a micrometer screw (Mitutoyo). Since VHB is very soft and sticky, it is sandwiched between two glass slides with known thickness and the overall thickness is measured. By subtracting the glass' thickness the thickness of elastomer membrane is obtained with a sufficiently high accuracy of ±5 μm. Due to the stiffness and non-stickiness of ZRUELAST, the thickness of the stretched natural rubber membranes is directly measured with the micrometer screw.

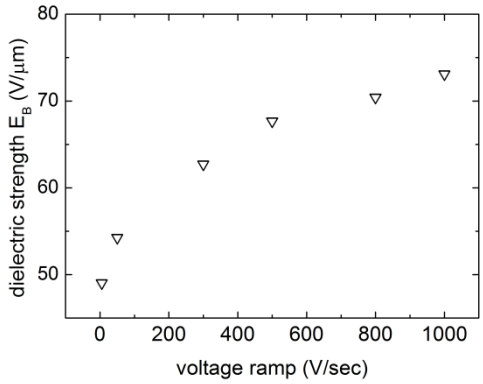
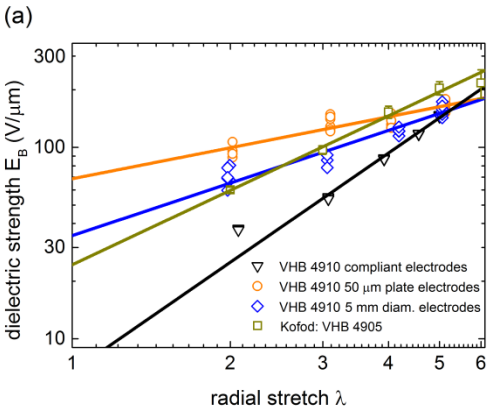


Figure 3: Breakdown field strength versus voltage ramp, showing an increase of the breakdown field strength with increasing voltage sweep rate.

For the breakdown measurements we have increased the voltage supplied to the sample at a constant rate from 5 to 1000 V/s. Figure 3 shows the evolution of the breakdown field strength versus the voltage ramp rate. Measurements were performed with the arrangement shown in figure 2a on VHB samples with a stretch rate of λ=*3*. It is interesting to note that the breakdown field strength increases with increasing voltage ramp rate. In order to allow a comparison of the experiments discussed below, we have chosen a voltage ramp rate of 50 V/s in all further breakdown measurements.

Figure 4a shows the data obtained for VHB 4910 as a function of the radial stretch ratio. As suggested in [6] we fit the experimental data with the phenomenological relation $E_{EB}(\lambda) = E_{EB}(1) * \lambda^R$, where $E_{EB}(1)$ is the electrical breakdown field when the membrane is not stretched (λ=1), R is the degree of sensitivity of the electrical breakdown field toward the stretch ratio λ. R=0 implies that the electrical breakdown field is independent of stretch.



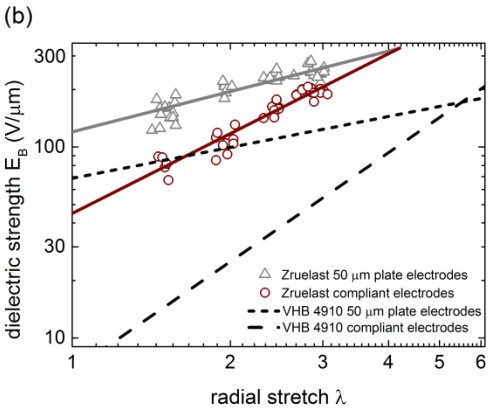


Figure 4: Experimental values for the dielectric strength E_{EB} of VHB 4910 (a) and ZRUELAST natural rubber (b) using compliant and rigid electrode configurations. The data are fitted to $E_{EB}(\lambda) = E_{EB}(1) * \lambda^R$.

Independent of the experimental conditions the dielectric strength of VHB increases with the stretch ratio. The breakdown fields E_{EB} obtained with compliant electrodes (black triangles) are lower than those with rigid electrodes in clamped conditions (orange squares). At high stretch levels the increasing stiffening of the elastomer suppresses actuation, the values converge for both electrode configurations. We believe that the breakdown strength measured with rigid electrodes under clamped conditions reveals the true material limit, while the lower values with compliant electrodes may be caused by the Stark Garton electromechanical instability breakdown mechanism. To show the importance of electrode area on breakdown measurements we have also used stamps with a larger electrode area. These data are represented by blue diamonds, the fit line is blue. The stamps consist of electrodes with 5 mm diameter pressed into an insulating plastic ring with 10 mm diameter. The plastic ring was used to prevent actuation in a similar way to the experimental scheme of figure 2b. The latter electrode configuration is similar to the one used by Kofod and coworkers in Ref. [19]. The breakdown results of Ref. [19] are included in figure 4a as green squares and the corresponding fit line is green. The difference between the breakdown results obtained in this work and in [19] may arise from the use of different voltage ramps as well as from the different initial thickness of VHB 4905, which is half of the VHB 4910 used in this work.
The influence of the size of the electrode area is significant. As shown in figure 4a, larger electrode areas result in lower breakdown strength, presumably caused by material imperfections.

Figure 4b illustrates the measured breakdown values for ZRUELAST natural rubber using compliant electrodes and rigid electrodes under mechanical clamped conditions. Similar to VHB, the experimentally measured values with compliant electrodes are significantly lower than that with rigid ones. Similar to VHB the breakdown values converge when the stretch ratio increases, due to the stiffening of the rubber membrane. It is interesting to note the larger breakdown fields of natural rubber in comparison to those of VHB, suggesting the use of natural rubber for dielectric elastomer energy harvesting. Table 1 summarizes the stretch dependence of the breakdown field strength of VHB and rubber for the two experimental techniques.

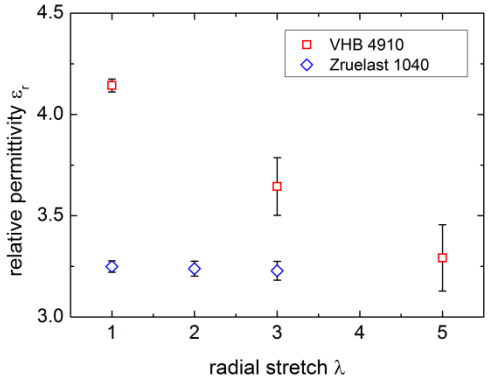


Figure 5: Relative dielectric permittivity @ 1 kHz of VHB and rubber versus stretch ratio. While VHB shows a stretch dependent permittivity, the permittivity of rubber is insensitive to stretch.

The relative dielectric permittivity ε of the two elastomers was measured with a NOVOCONTROL broadband dielectric spectrometer at different stretches. The stretched samples were prepared as described above, by fixing the stretch with a polyamide ring glued to the membrane. For the measurements silver paste electrodes with 14 mm diameter were used. As shown in figure 5, VHB shows a clear stretch dependence of the dielectric permittivity, as also noted by others [20,21]. Reasons for a stretch dependent dielectric permittivity are dipoles oriented along the main chain, strain-crystallization, and generally a stretch dependent glass transition temperature. It is interesting to note that rubber behaves different than VHB. We have observed no significant stretch dependence of the dielectric permittivity of rubber. Table 2 lists the dielectric permittivity and loss tangent measured at a frequency of 1 Hz (a typical frequency used in actuation and energy harvesting with dielectric elastomers) and 1 kHz. The stretch independent dielectric function and the low loss tangent also at a frequency of 1 Hz suggest the use of rubber in dielectric elastomer energy harvesting.

4. Conclusion

The measurements reveal a stretch dependence of the dielectric breakdown of the acrylic elastomer VHB and natural rubber ZRUELAST. Breakdown values depend on the electrode configuration, being lower when compliant electrodes are used instead of rigid electrodes and mechanical clamping of the membranes. Our measurements further show the influence of electrode size on the breakdown values; the breakdown strength decreases with increasing area, probably due to material imperfections. Natural rubber is shown to have a dielectric permittivity insensitive to stretch in contrast to the stretch strain dependent permittivity of VHB. The comparison of ZRUELAST and VHB 4910 strongly suggests the use of natural rubber for dielectric elastomer energy harvesting.

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Table 1: Breakdown strength, dielectric function and loss tangent of VHB 4910 and of ZRUELAST natural rubber.

	E_{EB} (V/µm) @ different λ			
Material / Method	λ=2	λ=3	λ=4	λ=5
VHB 4910				

rigid electrodes	100	124	145	163
compliant electrodes	25	54	94	143
ZRUELAST 1040				
rigid electrodes	195	260	--	--
compliant electrodes	118	207	--	--

Table 2: Dielectric function and loss tangent of VHB 4910 and ZRUELAST 1040 natural rubber.

	Stretch dependent dielectric function and loss tangent			
Material	λ=1	λ=2	λ=3	λ=5
VHB 4910				
ε (@ 1Hz)/tan(δ)	4,24 / --	--	$3{,}83 / 8 \cdot 10^{-3}$	$3{,}44 / 1 \cdot 10^{-2}$
ε (@ 1kHz)/tan(δ)	$4{,}12 / 2 \cdot 10^{-2}$	--	$3{,}64 / 3 \cdot 10^{-2}$	$3{,}29 / 2 \cdot 10^{-2}$
ZRUELAST 1040				
ε (@ 1Hz)/tan(δ)	$3{,}36 / 1 \cdot 10^{-2}$	$3{,}39 / 3 \cdot 10^{-2}$	$3{,}35 / 2 \cdot 10^{-2}$	--
ε (@ 1kHz)/tan(δ)	$3{,}25 / 6 \cdot 10^{-3}$	$3{,}24 / 1 \cdot 10^{-3}$	$3{,}23 / 1 \cdot 10^{-3}$	--

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