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Electromechanical response of silicone dielectric elastomers

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Abstract. This paper presents an experimental technique to investigate the electromechanical properties of silicone dielectric elastomers actuated with high DC electric fields. A non-contact measurement technique is used to capture and monitor the thickness strain (contraction) of a circular film placed between two metallic disks electrodes. Two active fillers such as silica (10, 15 and 30 wt%) and barium titanate (5 and 15 wt%) were incorporated in order to increase the actuation performance. Thickness strain was measured at HV stimuli up to 4.5 kV and showed a quadratic dependence against applied electric field indicating that the induced strain is triggered by the Maxwell effect and/or electrostriction phenomenon as reported in literature. The actuation process evidences a rapid contraction upon HV activation and a slowly relaxation when the electrodes are short-circuit due to visco-elastic nature of elastomers. A maximum of 1.22 % thickness strain was obtained at low actuating field intensity (1.5 V/μm) comparable with those reported in literature for similar dielectric elastomer materials.

1. Introduction

Over the last two decades, the field of electrically controllable polymers displaying significant size or shape change has emerged. These responsive materials, referred to as electroactive polymers (EAPs), can be used as actuators, sensors and energy harvester in various fields such as micro-electromechanical systems (MEMS), in bio-mimetic robots and smart prosthetic as artificial muscle, in haptic and microfluidic devices, electrically tunable diffraction grating [1-8].

EAP actuators transform electrical energy directly into mechanical work and produce large strain and stress. When an electric field is applied on an EAP material it can undergoes bending, transverse, longitudinal as well as surface strain that can be examined using various equipments and techniques. Transverse or thickness strains are much smaller than area strains and usually are difficult to monitor with sufficient precision [9, 10]. Acrylics, polyurethanes and silicones are the most widely used as dielectric layers in class of dielectric elastomers (DE) due to some remarkable electromechanical properties such as large electric field induced strain (area strain up to 380%), high specific elastic energy and fast speed response [11-15].

Silicone elastomers have good mechanical properties (low modulus and high elongations), good stability over a wide range of temperature, humidity and frequency but the main disadvantage is the low dielectric permittivity (in range of 2.5 ÷ 3.0), which requires increased activation voltage to obtain reasonable actuation effects. Dielectric fillers such as metallic oxides, graphite, pyrite, multi-walled nanotubes (MWNTs), ceramics (e.g., barium titanate, titanium oxides, lead zirconate, etc.) and conductive polymer nanoparticles are used to enhance electrical properties of silicone elastomers [16-22]. Prestraining the elastomer film was found to increase the actuator performance, but this has effect on Young's modulus and dielectric constant [23, 24].

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In this paper we investigated the electromechanical properties of some silicone elastomers with two active fillers: silica as a reinforcing agent and barium titanate ($BaTiO_3$) as dielectric permittivity enhancer [18]. Circular samples were placed between two metallic disks and actuated with high voltage (HV) stimuli. The displacement (thickness strain) was measured using a non-contact technique at RT conditions.

2. Experimental part

Non-contact optical methods, using laser interferometers, are reliable and extensively used but they involve the use of sophisticated equipment and require samples with well optically defined surface or mirroring attachments mounted on specimen holders.

In this paper we used a non-contact measurement system designed to monitor the gap between the probe tip and a metal target (figure 1). Typically, the probes are used in machinery systems to monitor the end thrust or radial vibration of a rotating shaft. The probe tip contains an encapsulated coil assembly which, when connected to the driver unit, produces a high frequency oscillation that induces eddy currents in the target material. The eddy currents induced in the target, and hence the oscillation amplitude, are proportional to the probe gap. The linear range of MTN/EP080 is 0 ÷ 2 mm and the sensitivity is 8 V/mm. The probe position should be chosen to minimize the effect of surface roughness, changes in diameter or eccentricity, which will affect the accuracy of the measurement. The target area, perpendicular to the probe tip, must have a diameter of at least 3 times the probe diameter with no discontinuities, such as spigots or keyways, impinging.

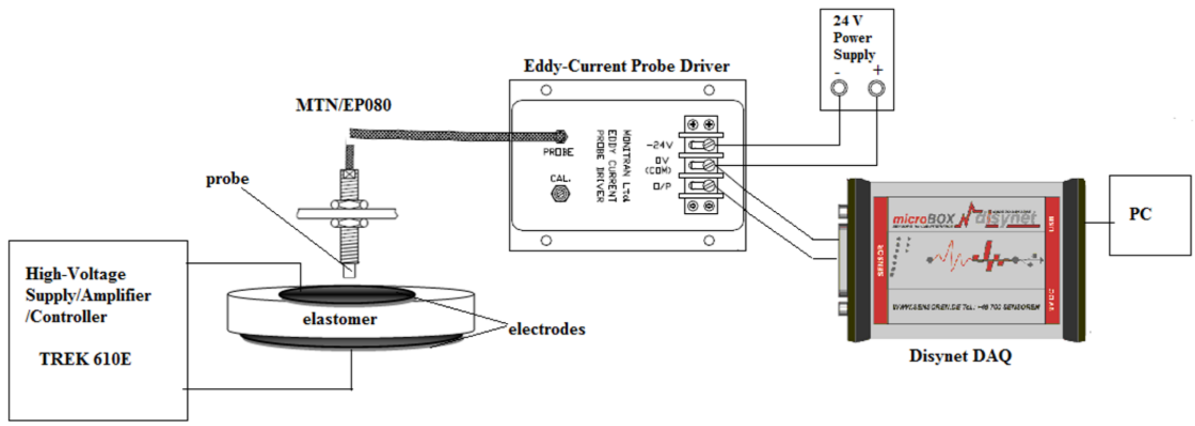

Figure 1. Non-contact measurement system for monitoring the thickness strain of dielectric elastomers.

First, a calibration procedure was adopted for MTN/EP080 using the Tribometer UMT-2 from CETR (figure 2a). The relationship between displacement and the output voltage from ECPD is found to be linear as shown in figure 2b. The calibration process is such that the sensor probe is fixed on the carriage stage of the tribometer and moved by a know distance (Δ) away from the metallic target in an increment of 0.1 mm and the resultant increase in voltage (V) is measured by a digital voltmeter.

The electromechanical measurements were performed on six silicone elastomers with different amount of active fillers like silica and $BaTiO_3$ (table 1) [18]. Similar to other researches [10, 25, 26], circular samples were placed between two metallic disks acting as electrodes. The lower electrode had a diameter of 23.5 mm and 1.4 mm in thickness while the upper electrode dimensions are 18 mm and 0.34 mm, respectively. A very thin layer of hydrocarbon liquid was applied between the metal disk electrodes and the elastomeric samples [25, 26]. This was done in order to minimize the electrode constraint by diminishing of frictional force on the film [27] as it expands laterally during the electric field activation, which induces a decrease in thickness. In the same time the liquid layer reduces any

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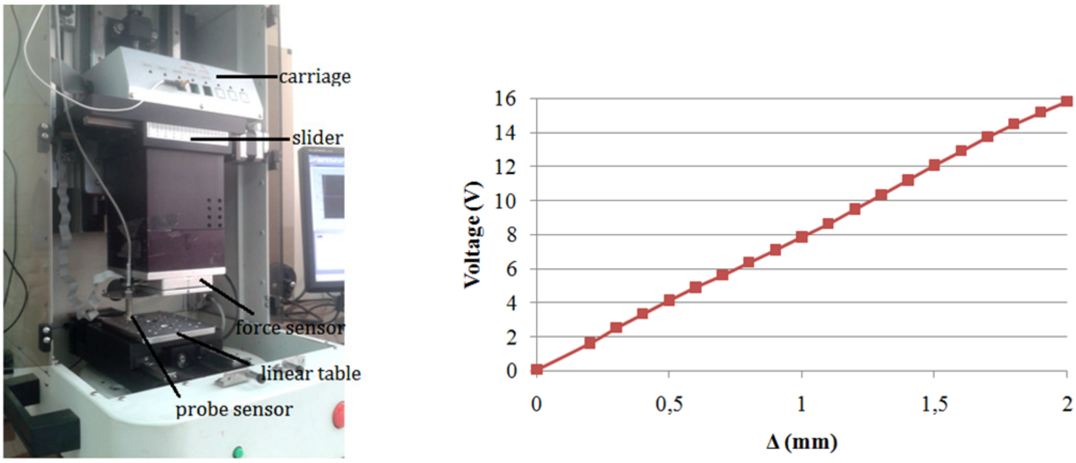
measurement error produced by air trapped between the electrodes and sample, and holds the film flat and perpendicular to the probe.



(a) (b)

Figure 2. Calibration procedure of displacement sensor: (a) sensor probe fixed on carriage stage to be moved with fine increments; (b) calibration curve.

Dielectric elastomer samples were excited by step-increasing HV stimuli (produced by Trek 610E supply-amplifier) ranging from 0.2 to 4.5 kV in 0.5 kV steps lasting 60 s each. The resulting thickness strains were captured by sensor probe MTN/EP080 and recorder on a PC via a single-ended channel of a Data Acquisition Board from Disynet. The compressive stress produced by the weight of the upper electrode (0.6667 g) is about 1.12 kPa that is much smaller than the Young modulus of specimens and it may be assumed that the material conforms to the Hook law and thus such small stress results in very small initial displacement (compression) of DE sample. The initial thinning of the test specimen is thus much smaller than the displacements registered when the DE specimen is actuated and may be neglected. Moreover, mechanical initial compressive stress much higher than 1.4 kPa is also commonly imposed on EAP specimens when the samples are initially highly pre-stressed [11, 23] or when the sample fixture or electrodes limit free movement of the HV-actuated EAP specimen.

3. Results and discussions

When an electric field is applied to isotropic dielectric sample, the induced strain is a superposition of pure quadratic electrostriction and strain caused by electrodes attraction (Coulomb forces). The pure electrostriction is the direct coupling between electric polarization and mechanical strain response, while the Maxwell effect is mainly due to attractive force between the opposite charges on the electrodes as:

$$S_E=-Q\varepsilon_0^2(\varepsilon_r-1)^2E^2 \quad (1)$$

$$S_M=-\frac{\varepsilon_0\varepsilon_r}{2\,Y}E^2 \quad (2)$$

where Q is the pure electrostrictive coefficient, ε_0 is the dielectric permittivity of free space, ε_r is the relative dielectric permittivity of sample, E is the electric field and Y is the Young's modulus. The measured experimental strain S (total strain) should be

$$S=S_E+S_M \quad (3)$$

The percentage of the Maxwell contribution to the global induced measured strain can be evaluated from the relation:

$$\frac{S_M}{S_E}=\frac{\frac{\varepsilon_0\varepsilon_r}{2\,Y}}{M} \tag{4}$$

Dielectric elastomer samples stimulated with high voltages of up to 4.5 kV showed a fast thickness strain (contraction) but when the HV stimulus was detached and the test specimen was shorted it returned to its initial thickness (relaxation) slowly due to the visco-elastic nature of elastomers. In some case, the visco-elasticity was also responsible for gradual and delayed reaction to step HV excitation, manifested as "rounding" of the steps in figure 3.

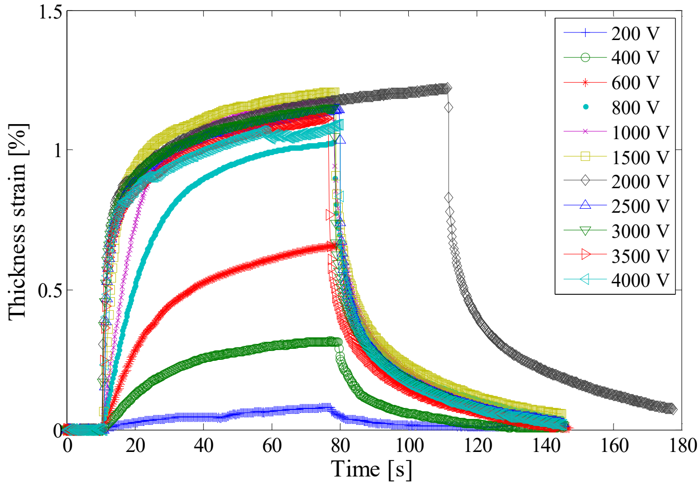


Figure 3. Thickness strain in time for S10B15 sample.

The total thickness strains of all the test specimens are plotted in figure 4 against the electric field applied. It can be observed that up to an electric field value, which depends on the film thickness, the induced strain presents a quadratic dependence, whereas at higher values the strain tended to reach saturation [28]. Such nonlinear curves have been registered in all dielectric elastomer materials by other researchers using various measurement techniques [9, 25, 26]. The deviation from the quadratic dependence is better observed in figure 5, where the thickness strains are plotted versus the square of actuating field intensity. From the slope of the straight lines the apparent electrostrictive coefficients were calculated (table 1). Also, figure 5 displays linearity at relatively low actuating fields.

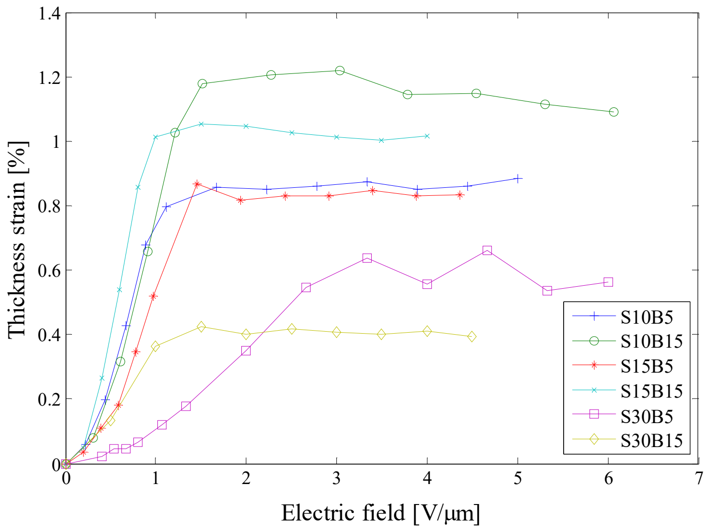


Figure 4. Dependence of thickness strain on electric field.

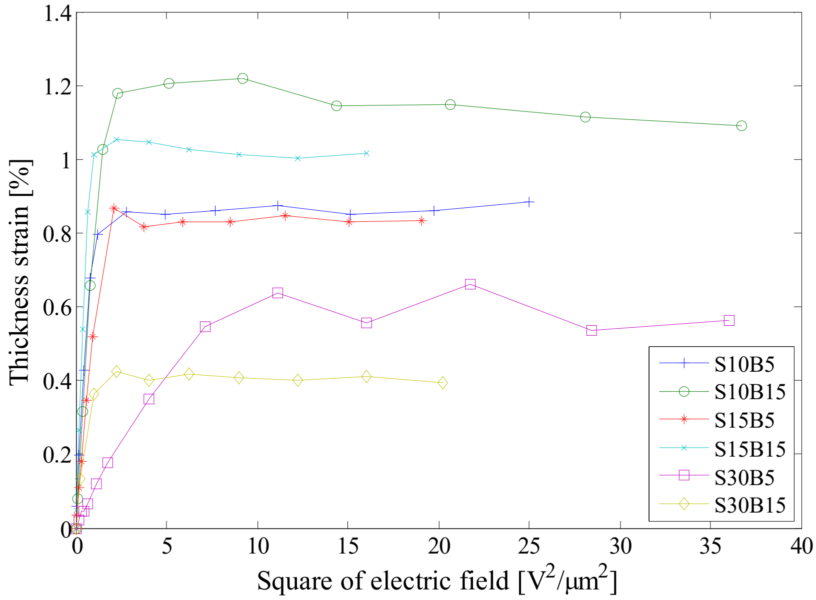


Figure 5. Relative thickness change as function of the square of applied DC actuating field.

The Maxwell contribution deduced for test specimens have very low values that could be mainly attributed to the high value of the apparent electrostrictive coefficient and to rigid electrodes that can constraint the elastomer to expand in area. Therefore, it is recommended for a further work to use compliant electrode [29 - 32].

For low strain (less than 20%), where the Hooke's law is supposed to be valid, the effective compressive pressure *p* and mechanical energy density *e* can be approximated by the expressions:

$$p=SY \tag{5}$$

$$e=\frac{1}{2}YS^2 \tag{6}$$

Table 1. Mechanical, dielectric and electromechanical parameters of tested specimens.

Sample	Silica (wt%)	$BaTiO_3$ (wt%)	Thickness (mm)	Y (MPa)	ε (at 10 Hz)	Maximum thickness strain (%)	M 10^{-13} (m^2/V^2)	S_M/S (%)	Actuation pressure (MPa)	Elastic energy density e 10^{-3} (J/cm^3)	Electromechanical sensitivity β=ε/Y (MPa^{-1})
S10B5	10	5	0,9	0,81	3,67	0,88	9	0,0022	0,0071	0,31	4,53
S10B15	10	15	0,66	0,89	3,95	1,22	7	0,0028	0,0108	0,66	4,43
S15B5	15	5	1,03	1,91	3,66	0,86	6	0,0014	0,0165	0,71	1,91
S15B15	15	15	1	2,1	4,09	1,05	10	0,0008	0,0221	1,16	1,94
S30B5	30	5	0,75	5,85	3,89	0,65	0,8	0,0036	0,0386	1,27	0,66
S30B15	30	15	1	8,25	4,26	0,42	4	0,0005	0,0351	0,74	0,51

The highest thickness strain of 1.22% was obtained for S10B15 sample. As we expected, the thickness contraction decrease as the sample becomes stiffer and increase with barium titanate content.

This fact can be observed in figure 6 were thickness strain is plotted against Young's modulus and barium titanate content.

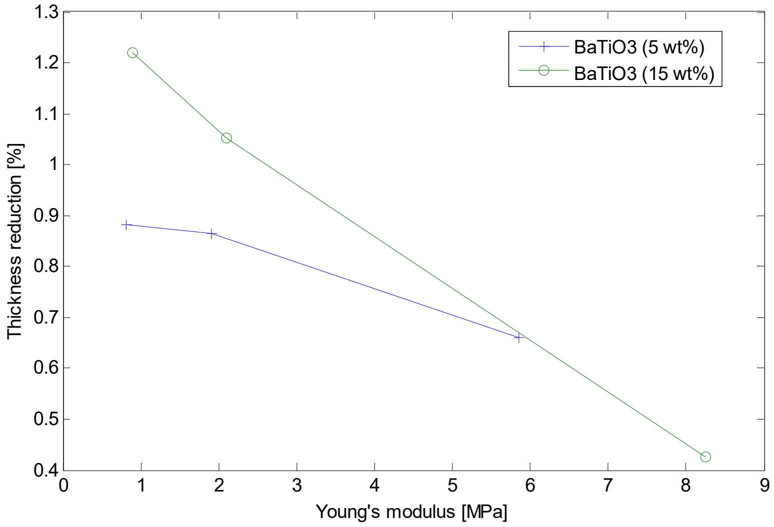


Figure 6. Thickness strain versus Young's modulus.

4. Conclusions

In this work the authors investigated the electromechanical response of some silicone dielectric elastomers. Silicone composites films with silica as a reinforcing agent and barium titanate as dielectric permittivity enhancer were placed between two metallic disks electrodes and stimulated with HV steps up to 4.5 kV. The thickness strain (contraction) was measured using a non-contact measurement technique based on an eddy current probe sensor that captured the gap between the probe tip and a metal target. A static calibration was performed in order to establish the relation between displacement and output voltage of the probe sensor. Six circular sample recipes were activated and deactivated with HV step stimuli lasting 60 s each. Thickness strains showed a quadratic dependence against applied electric field and a maximum of 1.22 % thickness strain for S10B15 was obtained at low actuating field intensity. Also, other electromechanical parameters such as apparent electrostrictive coefficient, Maxwell contribution, electromechanical sensitivity and elastic energy density were calculated. We think that the low values of Maxwell contribution could be mainly attributed to the high value of the apparent electrostrictive coefficient and to rigid electrodes that can constraint the elastomer to expand in area. Therefore, Maxwell contribution can be increased by using compliant electrodes [29-32]. The thickness strain increase with barium titanate content that is a dielectric permittivity enhancer and decrease with Young's modulus due to silica content, as we predicted. In conclusion, the tested silicone elastomers have the potential to be used as active layer in dielectric elastomer actuator (DEA) technology and other electromechanical applications.

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