Comparative study of electromechanical response in some dielectric elastomers

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Dielectric elastomers, a subclass of electroactive polymers (EAP) materials, are very promising for actuator and sensor applications due to their interesting properties such as high energy density, high strain levels from 10 to 380 % and fast response in order of milliseconds. Polymer films were sandwich between two rigid electrodes and subjected to high DC electric fields. We investigate the transverse strain responses of some dielectric elastomer actuators (DEA) using an eddy current displacement sensor. Electric-field-induced thickness strains in order of micrometers were measured at different high DC step voltage and a quadratic dependence was observed. A comparison has been made between three categories of elastomers. The preliminary results show that performance of the actuators not only depends on the material properties but it is also a function of the actuator structure and electrode materials. We also concluded that these materials may be candidates for actuator materials in micro-actuation mechatronic systems.

1. Introduction

Many kinds of actuators – such as electrical motors, pneumatic or hydraulic cylinders, shape memory alloy (SMA) actuators and magnetic solenoids – have been widely used for engineering disciplines. Pneumatic actuators show the best performance in term of speed and power density, but they cannot be easily miniaturised and an independent source of compressed air is necessary for each of them (Kingsley et al., 2003). SMA actuators have a good power density too, but they show a very low efficiency and a relatively low contraction/ relaxation speed; moreover, they are subjected to degradation along time and they are slow (O'Halloran and O'Malley, 2004).

In the last decade, electroactive polymers have attracted much attention owing to their interesting electromechanical properties. Generally, EAP materials can generate strains that are as high as two orders of magnitude greater than the striction-limited, rigid, and fragile piezoelectric ceramics. Further, EAP materials are superior to shape memory alloys (SMA) in higher response speed, lower density, and greater resilience.

In particular, dielectric elastomers, which are incompressible visco-elastic dielectrics, such as polyurethane, silicone and acrylic exhibit high stress/strain density, low elastic stiffness, high dielectric breakdown strength, good efficiency and high speed of response in the order of milliseconds. Their characteristics allows for the production of linear actuators using dielectric elastomer films that appear to act similar to biological muscles, therefore, dielectric EAP actuators are often referred to as "artificial muscles" (Bar-Cohen, 2006).

Both electrostriction and Maxwell stress contribute to the field-induced strain response. Electrostriction is the direct coupling between electric polarization and mechanical strain response, while Maxwell stress is the attractive force between the opposite charges on the electrodes.

1.1 Basic operation principle of DEA

Principle actuation physics of DEA was elaborate and well documented in some publications (Pelrine et al., 1998; Pelrine et al., 2000a). A schematic overview of the basic actuation mechanism is shown in Fig. 1.
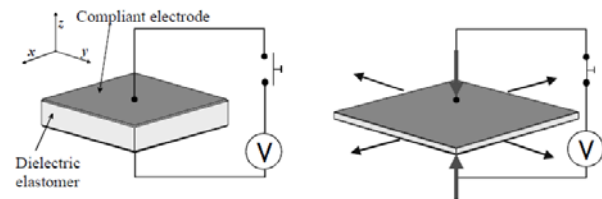

Fig. 1. Operation principle of DEA.

Dielectric polymeric actuators consist of an elastomeric film sandwiched between compliant electrodes to form a capacitor. When a potential is applied across the electrodes, the induced charge causes an electrostatic attraction between the electrodes. The resulting compressive force, or Maxwell stress, leads to a reduction in film thickness, which in turn results in elongation in the plane of the film. According to the derivation of Pelrine

(Pelrine et al., 1998), the equivalent electrode pressure p_e mainly depends on the applied voltage U and the thickness of dielectric film:

$$p_e = \varepsilon_0 \varepsilon_r E^2 = \varepsilon_0 \varepsilon_r (U/z)^2 \qquad (1)$$

where ε_0 is the free-space permittivity ($8,85 * 10^{-12}$ F/m), ε_r is the relative dielectric constant of the elastomer, z is the polymer thickness and E is applied electric field. As soon as the voltage is switched off and the electrodes are short-circuited, the actuator contracts back to its original size and shape.

For small strains with free boundary conditions, the polymer thickness strain, s_z, is given by (Pelrine et al., 2000b):

$$s_z = -p/Y = -\varepsilon_0 \varepsilon_r (U/z)^2 / Y \qquad (2)$$

where Y is the modulus of elasticity.

One of the more useful metrics for comparing actuator materials, independent of size, is the energy density of the material. The *actuator energy density* is the maximum mechanical energy output per cycle and per unit volume of material. The actuator energy density depends on the loading conditions. For small strains with free boundary conditions, the actuator energy density, e_a, of the material can be written as (Pelrine et al., 2000b):

$$e_a = -ps_z = \frac{(\varepsilon_0 \varepsilon_r)^2 E^4}{Y} = \frac{(\varepsilon_0 \varepsilon_r)^2 (U/d)^4}{Y} \qquad (3)$$

Conventionally, the *elastic energy density* is often used [6]:

$$e_e = \frac{1}{2} Y s_z^2 \qquad (4)$$

In order to improve their performance, dielectric elastomers are pre-stretched, typically up to five times their in-plane dimensions. By arranging several actuator layers in parallel, the resulting force can be multiplied and the stiffness of the created material can be increased. When the actuators are combined in series, the arising displacement becomes larger. In order to lower the activation voltage without reducing the actuator's elongation, the film's relative permittivity could be increased. From (1), it is evident that the reduction of the thickness of the film is more effective than the latter effect (Zhang et al., 2005).

Electrode materials should typically have good compliancy and conductivity so that undergo large strain alongside the film, without producing any additional stress and constraint for the actuator (Khodaparast et al., 2007). Various electrodes, including evaporated thin metal film, compliant electrodes such as graphite powder, polymeric elastomers filled with very fine conductive particles (carbon black, silver), electrolyte polymer solutions and conductive polymer (polypyrrole) deposited on polymer electroactive film by in-situ polymerization have been used (Pelrine et al., 1998; Zhenyi et al.,1994; Su et al., 1997a; Su et al., 1997b; Su et al., 1998, Liu et al., 1999; Watanabe et al., 2002). Rigid plate electrodes were also used (Zhenyi et al.,1994; Diaconu et al., 2004; Diaconu et al., 2005).

The aim of this paper is to investigate the transverse strain response of some dielectric polymers subjected to high DC electric fields. We also make a comparison between three different polymer materials in terms of measured thickness strain response.

2. Materials

It was reported the use of a wide range of polymeric materials including polyimides, polyxylene, fluoropolymers, liquid crystal polymers, polydimethylsiloxane (PDMS), etc. in micro-electro-mechanical systems (MEMS) (Huang et al., 2006; Cazacu et al., 2009). Silicones are easily processable (by spin coating, casting, etc.) and permit the realization of good rubber-like dielectrics, owing to the high mobility of the Si-O bonds, which confer high flexibility to main chains. Dielectric breakdown of silicones occurs at fields ranging from 100 to 350 V/μm, enabling thickness strains up to 40-50 % and area strains up to 100 % (Pelrine et al., 2000a).

Dielectric elastomers require high driving electric fields. For this reason, in order to reduce the driving electric fields, polymers with high dielectric constant are necessary (according to equation (1)). With respect to this, new highly dielectric elastomers could be realized by means of a composite approach. By filling an ordinary elastomer (e.g. silicone) with a component having a greater dielectric permittivity, it is possible to obtain a resulting material showing the fruitful combination of the advantageous matrix elasticity and filler permittivity. Using active fillers, the dielectric properties of silicones can be modified.

2.1 Poly(dimethyldiphenylsiloxane)s/silica networks

The synthesized dimethyldiphenylsiloxane copolymers having different contents in diphenylsiloxane units (between 6.7 – 21.6 % moles) were mixed in different ratios with tetraethylorthosilicate (TEOS) as crosslinker in presence of dibutyltindilaurate as catalyst. TEOS was used in large excess, thus in situ formation of silica network occurs concomitantly with crosslinking. Polymer specimens were processed as films from 0.05 to 0.235 mm thick, by pouring the reaction mixture on a substrate, before finalizing crosslinking (Cazacu et al., 2010).

2.2 PDMS/SiO$_2$/TiO$_2$ composites

PDMS/SiO$_2$/TiO$_2$ composites were prepared by adding the pre-established amounts of oxide precursors

(TEOS and TBT) and condensation catalyst (DBTDL) to a polydimethylsiloxane-α,ω-diol, PDMS, (M_v=48000). After vigorous stirring at room temperature and degasation, the mixtures were poured on a Teflon foil. The hydrolysis of the corresponding precursors takes place under the influence of the environmental humidity followed by catalytic condensation with the formation of silicon and titanium oxides networks. The crosslinking of PDMS concomitantly occurs. Thickness of polymer specimens was in range of 0.47 ÷ 0.99 mm (Alexandru et al., 2010; Alexandru et al., 2011).

2.3 Cellulose acetate crosslinked by siloxane derivative

The films based on cellulose acetate crosslinked by siloxane derivative were prepared by adding 1,3-bis(3-glycidyloxypropyl)tetramethyldisiloxane to a 2.5% solution of cellulose acetate in tetrahydrofurane. The two reactants were mixed in various ratios. The pH of the solution was lowered at pH=2 with 3-4 drops of HCl 37% poured into the solution.

The reaction was conducted in acid environment in order to facilitate the opening of the epoxy cycle and its interaction with free or esterified OH groups. The mixture was kept under reflux for 2 hours at a temperature of approximately 65 ºC and then poured on a Teflon substrate. The films formed after solvent evaporation were washed with distilled water in order to eliminate the residual acidity (Stiubianu et al., 2009). Thicknees of films was between 0.028 to 0.062 mm.

3. Method

For experimental investigation of electromechanical response we used a non-contact technique based on a displacement sensor. An eddy-current sensor produces an alternating magnetic field at the probe tip.

When this field is near a conductive material, the field creates (induces) small electrical eddy-currents in the material. These currents generate a magnetic field that opposes the field from the sensor. As the gap between the probe and target gets smaller, the field interaction changes.

The sensor electronics can measure this field interaction and generate an output voltage proportional to the change in the gap. The sensor is used for measurements against electrically conductive and non-ferromagnetic materials.

The experimental setup used for thickness strain measurements is showed in fig. 2 and was presented in detail in a previous paper (Cârlescu et al., 2009).

Fig. 2. Experimental setup for field-induced thickness strain measurements.

Elastomer films were sandwich between two copper rigid electrodes and silicone oil was put on electrode-elastomer interface to reduce the errors and the friction forces and to holds the film flat. The sensor was placed near the upper electrode to measure the displacements of the elastomer films when an electric field was applied between electrodes. High voltage steps were applied by Trek 610E and thickness strains in range of μm were measured as a function of the DC voltage amplitude. All the measurements were made at room conditions.

4. Results and discussion

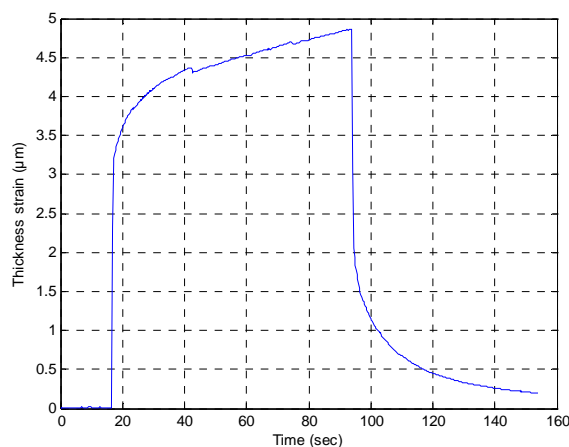Fig. 3 shows an example of thickness strain response in time under a step voltage of 3 kV.

Fig. 3. Time-strain response of specimens.

All films showed compression in thickness direction under applied static electric field. Fig. 4 presents the field-induced thickness strain of poly(dimethyldiphenylsiloxane)s/silica networks.
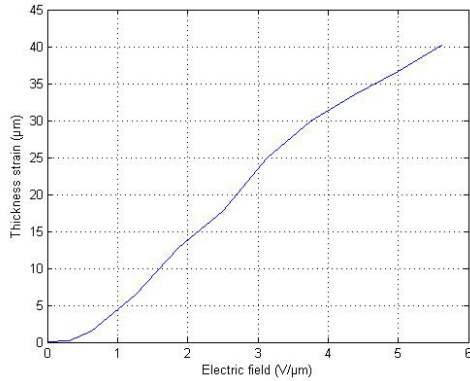
Fig. 4. Field-induced strain response of poly(dimethyl diphenylsiloxane)s/silica networks.

The dielectric breakdown field varies from a category of material to another. For the first category the maximum applied voltage was 900 V, voltage up to 5kV was applied to the second materials and about 1kV was applied for the third category of elastomers.

Figs. 5 and 6 present the field-induced strain responses of PDMS/SiO$_2$/TiO$_2$ composites and cellulose acetate crosslinked by siloxane derivative.

For non-piezoelectric silicone polymers, there are two possible effects that contribute to electric-field induced strain: a Maxwell stress effect and an electrostrictive effect.
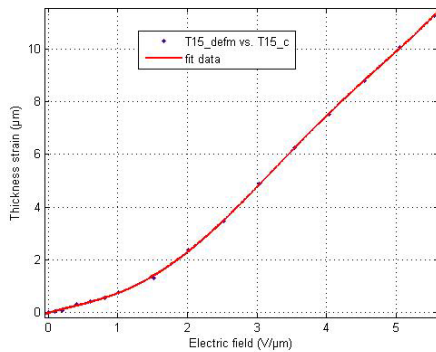
The Maxwell stress arises from the interaction between free charges on both electrodes (Coulomb forces) and it induce a transverse strain, γ, wich has the following relationship with the applied field E (Xia et al., 2003):

$$\gamma = (1 + 2\sigma)\varepsilon_0 \varepsilon_r E^2 / 2Y . \qquad (5)$$

where σ is Poisson's ratio (σ=0.5 for silicones) and Y is the Young's modulus. It can be seen from the Eq.2 that the transverse strain due to the Maxwell stress is directly proportional to the square of the applied field. On the other hand, the electrostrictive effect is due to the coupling between the polarization and the mechanical response in a material. The transverse electrostrictive strain response, γ, due to the applied field is (Zhang et al., 1997):

$$\gamma = Q\varepsilon_0^2 (\varepsilon_r - 1)^2 E^2 . \qquad (6)$$

where Q is the electrostrictive coefficient of the polymer. Thus, the electrostrictive strain is also proportional to the square of the applied electric field. It follows that the total strain due to both effects is proportional to the square of the electric field.

Fig. 7 show the quadratic dependence of thickness strain on electric field applied for poly(dimethyldiphenylsiloxane)s /silica networks.

Fig. 7. Dependence of the strain on square field for poly(dimethyldiphenylsiloxane)s/silica networks.

Fig. 5. Field-induced strain response of PDMS/SiO$_2$/TiO$_2$ composites.

Fig. 8 illustrate that the thickness strain is proportional with square of electric field applied to PDMS/SiO$_2$/TiO$_2$ composites.
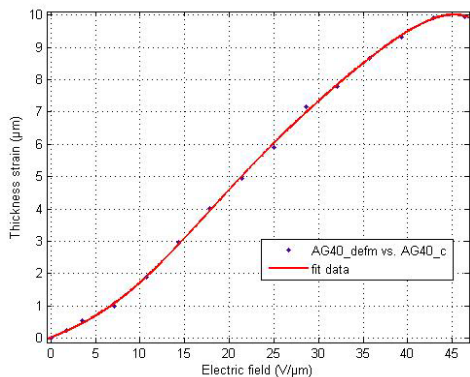
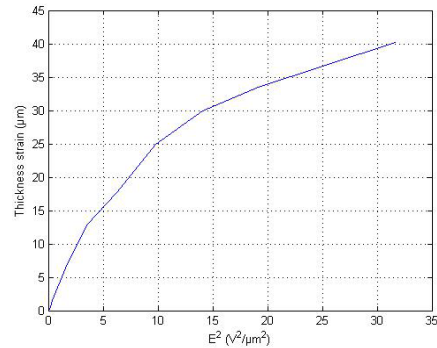*Fig. 6. Field-induced strain response of cellulose acetate crosslinked by siloxane derivative.*
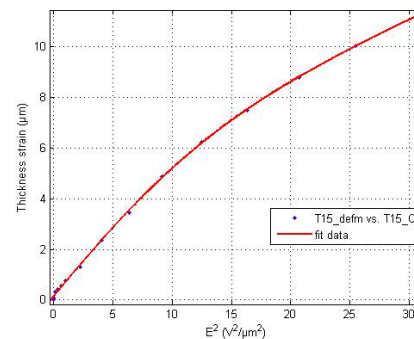

Fig. 8. Dependence of the strain on square field for PDMS/ SiO$_2$/TiO$_2$ composites.

Finally, Fig. 9 presents the same quadratic dependence for cellulose acetate crosslinked by siloxane derivative films.
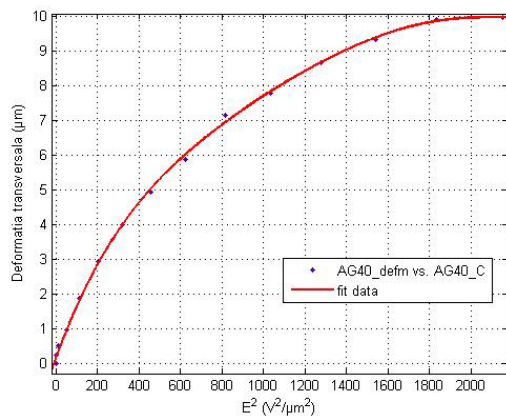

Fig. 9. Dependence of the strain on square field for cellulose acetate crosslinked by siloxane derivative films.

Compressions in thickness direction of the specimens were observed irrespective of the applied electric field sign. Also a symmetrical strain profile was found against inversion of the electric power polarity (Fig. 10).
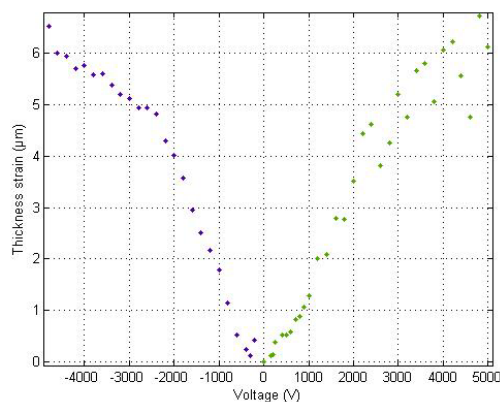

Fig. 10. Thickness strain on positive and negative DC voltage applied.

The levels of field-induced thickness strains are listed in Table 1.

Table 1. Thickness strains of specimens.

Category material	Voltage [V]	Maximum displacement (actuation) [µm]	Actuation sensitivity S [µm/V]
PDMS/silica	900	40	0.04
PDMS/ SiO_2/TiO_2	5500	11	0.02
PDMS/cellulose	1300	10	0.007

The electromechanical response of PDMS/SiO_2/TiO_2 composites in terms of thickness strain was also investigate by Cazacu et al. (2011). They obtained thickness strains in order of nanometres at relative low applied voltage.

6. Conclusions

A series of dielectric elastomers were characterized in terms of electromechanical response. All specimens showed thickness strain response under DC voltage applied in micromechanical actuation tests.

Poly(dimethyldiphenylsiloxane)s/silica networks showed the highest thickness strain response.

These results are promising for actuator and sensor applications if we consider that the electrodes used were rigid plates. A comparison was made between three categories of elastomers.

Further, we consider performing the characterization of these polymer films sandwich between two compliant electrodes and comparing the results with those obtained here. Also, new materials can be synthesized and tested in terms of field-induced strain response at DC and AC activation.

An important work is referring to model the electromechanical response of dielectric elastomers.

The results obtained allow us to recommend these polymer materials for planar or multilayer actuator applications.

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