Electromechanical Strain Response of $PDMS/TiO_2/SiO_2$ at Variable Electric Fields

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This paper refers to the electromechanical strain response of $PDMS/TiO_2/SiO_2$ composites as an electroactive polymer actuator. Lightweight electro-active polymers (EAPs) are ideal for control surface since they offer high strain rate, fast response, high elastic energy density and ease of manufacture [1,2]. We investigate the frequency response and vibrational modes of rectangular films with aluminum electrodes using Scanning Laser Doppler Vibrometry (SLDV) technique described elsewhere [3,4]. The results showed a similar behaviour with those of acrylic and silicone films coated with carbon-based compliant electrodes from literature. Field-induced vibrational strains decrease with frequency due to the increase in stiffness of sample and change of dielectric permittivity. These materials are possible candidates in polymer actuators for micro-pump and micro-valve applications in MEMS.

Keywords: electroactive polymers, dielectric elastomers, non-destructive technique, frequency response, vibrational modes

Within the general category of deformable polymer that change shape or size, the many feasible actuator technologies include electrostrictive polymers, piezoelectric polymers, shape memory polymers, electrochemically actuated conducting polymers, and polymer-based air-gap electrostatic devices [5]. Electro-active polymers (EAPs) are "smart materials" that exhibit large displacement and change their mechanical behaviour in response to electric stimulation. They have been emerging as substitutes for existing conventional actuators such as electromagnetic motors, piezo actuators or shape memory alloy actuators. Some electroactive polymers have been shown to have very large electric field-induced strains and their characteristics make them promising materials for many electromechanical actuator, transducer and active vibration damping applications [1,2,5] but also in many modern electronics and electric systems [6].

EAP materials can be divided into two major categories based on their activation mechanism: electronic ("dry") and ionic ("wet"). Electronic EAP are driven by electrostatic forces (Columbic forces) and include materials that are squeezed by the attraction force between opposite charges on two electrodes. They include electrostrictive, electrostatic, piezoelectric and ferroelectric materials that can hold deformation under dc voltage. In contrast, ionic EAP materials (gels, polymer–metal composites, conductive polymers, and carbon nanotubes) are driven by diffusion of ions and they require an electrolyte for the actuation mechanism. They need low voltage (~ 5 V) to induce actuation compare to electronic EAP that require high activation field (>100V/μm) close to the breakdown level [5].

During the last decade, dielectric elastomers (DE) also called electrostatically stricted polymers (ESSP), as a subclass of EAPs, showed great actuation performance such as area strains (up to 300%), high elastic energy density (3,4 J/cm^3), high speed of response of the order of milliseconds [1,2]. Figure1 shows the working principle of the basic unit of a dielectric elastomer actuator (DEA).

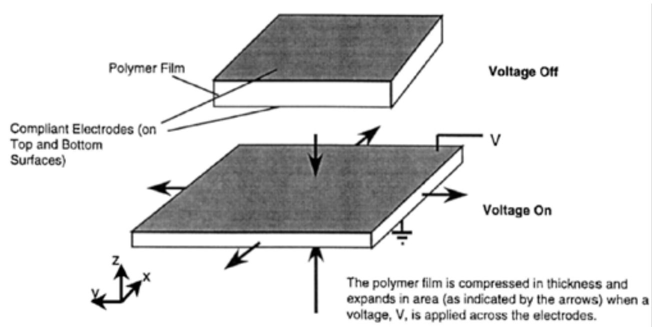

Fig. 1 Working principle of the dielectric elastomer actuator [2]

The working principle of a dielectric EAP is based on the electrostatic forces (Columbic forces) between two electrodes. DEA is basically a flexible capacitor with a dielectric soft material sandwiched between two compliant electrodes. When an electrical voltage is applied between the electrodes, the electrostatic forces from the charges on the electrodes squeeze the incompressible elastomer film (Maxwell stresses) and expand in area. According to the derivation of Pelrine [7], the equivalent electrode pressure p_e mainly depends on the applied voltage U and the thickness of the dielectric film d:

$$p_e = \varepsilon_0 \varepsilon_r \left(\frac{U}{d}\right)^2$$

where ε_o is the free-space permittivity and ε_r is the relative permittivity of elastomer film.

The electromechanical response of DEAs is based on both electrostrictive and electrostatic (Maxwell) effects. The measured experimental strain is then:

$$S = S_E + S_M = ME^2$$

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where S_E is the contribution of pure electrostriction, S_M is the electrostatic induced strain (Maxwell effect) and M is apparent electrostrictive coefficient.

In general, dielectric elastomer actuators contain two distinguishable regions, known as the active and non-active region (fig. 2). The active region denotes the area covered with electrode that is under the influence of the electric field. The non-active region corresponds to the elastomer film not covered with electrode and is prerequisite to avoid electrical shortage on the boundary of the DEA. However, the non-active region should be minimized to improve the actuation performance since it restrict the deformation of the active elastomer region.

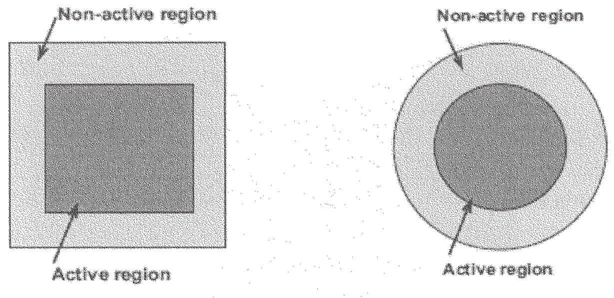


Fig. 2 Active and non-active region

Incompressible, soft elastomers with high dielectric breakdown strengths and high dielectric constants qualify as films for DE actuators. Generally, silicone and acrylic elastomers are most commonly used in actuator and sensor application. However, acrylic have good frequency response and show thermal instability unlike silicones that have lower viscoelasticity effect, are stable on a wide range of temperatures (-100^0 - +250^{0}C) and at UV-radiations.

So far, many configurations of actuators have been proposed such as the planar, tube, roll extender, diaphragm and bender actuator [1,2]. To increase the generated force of the actuator, a multilayered dielectric film system with alternating compliant electrode layers is needed, even the passive mechanical stiffness is increased. The spring roll actuator is a very promising design for achieving large actuation forces with dielectric elastomers. With rolled actuators, elongations of up to 26% and forces of up to 15 N were achieved [8].

In this paper we investigate the surface displacements of a single dielectric elastomer sheet covered with aluminum electrodes. The resonance frequencies are measured and the vibrational modes are determined on a frequency range of up to 200 Hz at high activation voltages with Scanning Laser Doppler Vibrometry (SLDV).

Experimental part

Methods and materials

Due to their interesting properties such as elasticity, insulating ability, thermal stability over a large temperature range, high hydrophobicity and easy processing, silicones are used in various industries such as biomedical, electronics, automotive and footwear. Polydimethylsiloxane (PDMS) is a silicone polymer with low stiffness widely used in MEMS technology where he plays a structural role as protective layers, encapsulating elements, valves and diaphragms [9]. The Young's modulus of bulk PDMS usually fall within 12 kPa to 2.5MPa depending on the processing conditions. The incorporation of different inorganic components into siloxane-based structures is carried out in order to improve the mechanical, thermal, electrical and optical properties and the dimensional stability or to obtain new properties derived from the hybrid nature of the material. Silica (SiO_2), titania (TiO_2), carbon black, $BaTiO_3$ and pyrite powder are some filler used [10, 11].

PDMS/TiO_2/SiO_2 composites were prepared according to a solvent-free sol-gel procedure by a group of researchers from "PetruPoni" Intitute of Macromolecular Chemistry [12]. Composites based on polydimethylsiloxane reinforced with silica and titania were prepared [12] by mixing polydimethylsiloxane with proper oxides precursors, tetraethylorthosilicate (TEOS) purchased from Fluka (Steinheim, Germany) and tetrabutylorthotitanate (TBT) purchased from Merck-Schuchardt (Hohenbrunn, Germany), in different ratios, in the presence of dibuthyltindilaurate (DBTDL) received from Merck-Schuchardt (Hohenbrunn, Germany) as a catalyst. The composite films have a thickness ranging from 0.47 to 0.99 mm. The Young modulus of composites films was calculated as the slope of the linear part of experimental stress-strain curves obtained from uniaxial tension tests [13]. For this study, the films were cut in rectangular form of about 40 mm x 40 mm. Aluminum foil of about 30 mm x 30 mm were glued on both face of the specimen films. Electromechanical parameters of PDMS/TiO_2/SiO_2 composites are listed in table 1.

Transverse, longitudinal as well as surface strain of deformable polymers is being examined using optical, capacitive or mechanical test methods involving sophisticated equipment (e.g. optical laser interferometer [14], laser Doppler vibrometers [3,4,15,16], Hall effect isotonic displacement transducers [17] or vision systems with digital image processing). All those test methods offer both advantages and disadvantages, however, so far no

Table 1
ELECTROMECHANICAL PARAMETERS OF PDMS/TiO_2/SiO_2 COMPOSITES

Sample	Young's moduli (MPa)	Relative Permittivity at 1 kHz	TBT (wt.%)
P1	0,805	3,48	-
T5	0,364	3,65	2,4
T7,5	0,28	3,72	4
T10	0,321	3,64	5
T15	0,38	3,75	7
T20	0,411	4,1	9
TM	0,09	-	5

common and complete methodology has been worked out for monitoring of mechanical displacements in electronic EAP materials, especially those relaying on high-voltage (HV) activation.

We have previously reported the investigation of the electromechanical responses of $PDMS/TiO_2/SiO_2$ composites subject to high dc voltage stimuli, using a displacement sensor technique [18]. The thickness contractions showed a quadratic dependence at electric fields of up to 6 V/μm.

Dynamic displacements, especially in the case of soft, free-standing and HV-excited specimens, are difficult to monitor with sufficient precision, repeatability and easiness of operation. Moreover, such monitoring requires being relatively wide band, covering quasi-static, dc-related displacements as well as dynamic movements, associated with ac excitation up to several kHz. Non-contact optical methods, using laser interferometers, are reliable, precise and extensively used.

In this study we used a laser-based technique for surface displacements of rectangular films in free-boundary conditions subject to high sinusoidal peak-peak voltages, of up to 6 kV_{pp}.

Laser Doppler vibrometry (LDV) was developed to measure and analyse the mechanical vibrations, that have a relevance in a very large number of industrial applications, from process monitoring to on line diagnostics and quality control. LDV techniques have proved to be suited for many applications, allowing to overcome problems related to vibration measurements, such as intrusivity, frequency response, and to implement measurements under harsh conditions (high temperature surface and noisy environments) or when hard-to-reach, small, or weak structures are analyzed. Vibrometry is a very sensitive optical technique capable of measuring sub-nanometer or even sub-picometer displacements from near DC to several MHz [19].

Laser Doppler vibrometer is basically an interferometer (Michelson or Mach-Zehnder) which measures the instantaneous velocity of a target through the measurement of the Doppler shift of scattered laser beam coming from the vibrating object. The operating principle of LDV is illustrated in figure 3. The laser beam is focused on the vibrating surface of sample, which diffuses the light with a frequency shift proportional to the velocity along the laser axis. Usually, the laser beam is split in two, with one beam acting as reference beam, while the other beam (measuring beam) is directed to the vibrating object. The two beams are finally mixed at the detector where the signal is demodulated. Frequency demodulation supplies the instantaneous velocity while the phase demodulation provides the displacement of the sample. There are

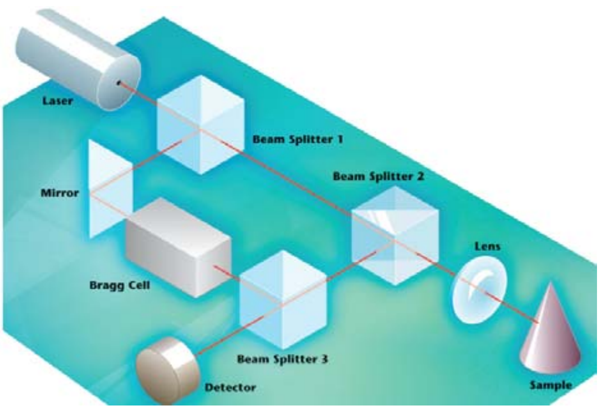


Fig.3 Optical schematic of laser Doppler vibrometer (Mach-Zehnder)

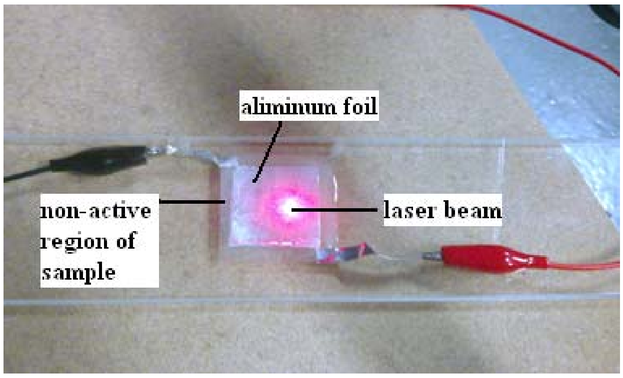


Fig. 4. Sample with laser beam

different types of laser vibrometers such as single point vibrometer for out-of-plane measurements, differential vibrometer and in-plane vibrometer that use two measuring beams, 3D vibrometer, rotational vibrometer, scanning and 3D scanning vibrometer [19].

Scanning Laser Doppler Vibrometer (SLDV) is a combination between a single point vibrometer and a scanning system that moves the laser beam across the surface of the target object.

Many researches in the field of dielectric EAP applied the vibrometry technique to study the dynamic electro-mechanical response of dielectric elastomer [15,16].

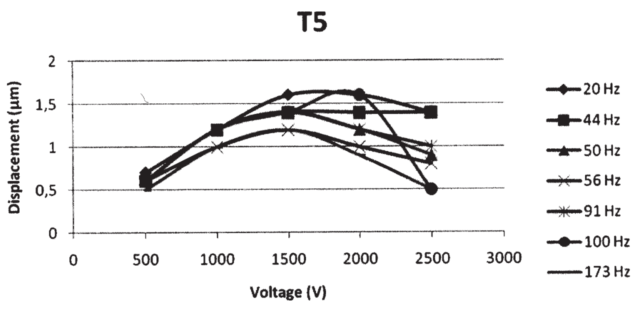
Results and discussions

The experimental setup used in our measurements was described in details in previous papers [3,4]. As already reported, the measurements were made in ES2T laboratory from CRITT M2A, France. The laser vibrometer PSV 200 from Polytec allowed us to measure the resonance frequency and vibration modes of rectangular samples covered with aluminum foils at HV-activation fields. Figure 4 shows the sample and the laser beam that moves on the vibrating surface. Vibrational modes were recorded on the active region of the samples.

The SLDV results showed that all samples have almost the same resonance frequencies and are more present in the first part of the frequency range. For all resonance frequencies the complex vibrational modes were investigateat different HV-excited specimens. Some examples of surface displacement modes were presented in [3,4]. Similar investigations werereported by Fox [15] and Dearing [16] on DE membranes with carbon-based compliant electrodes stretched on a rigid frame.

Figure 5 ilustrate the strain-voltage curves at various resonance frequencies obtained in this study.

From figure 5 can be observe that surface displacements decrease and the stiffness of samples increases at high frequencies due to the variation of dielectric permittivity and Young modulus. The amplitude of deformations is comparable with those obtained by Dearing [16] on silicone dielectric elastomer membranes with compliant electrodes.



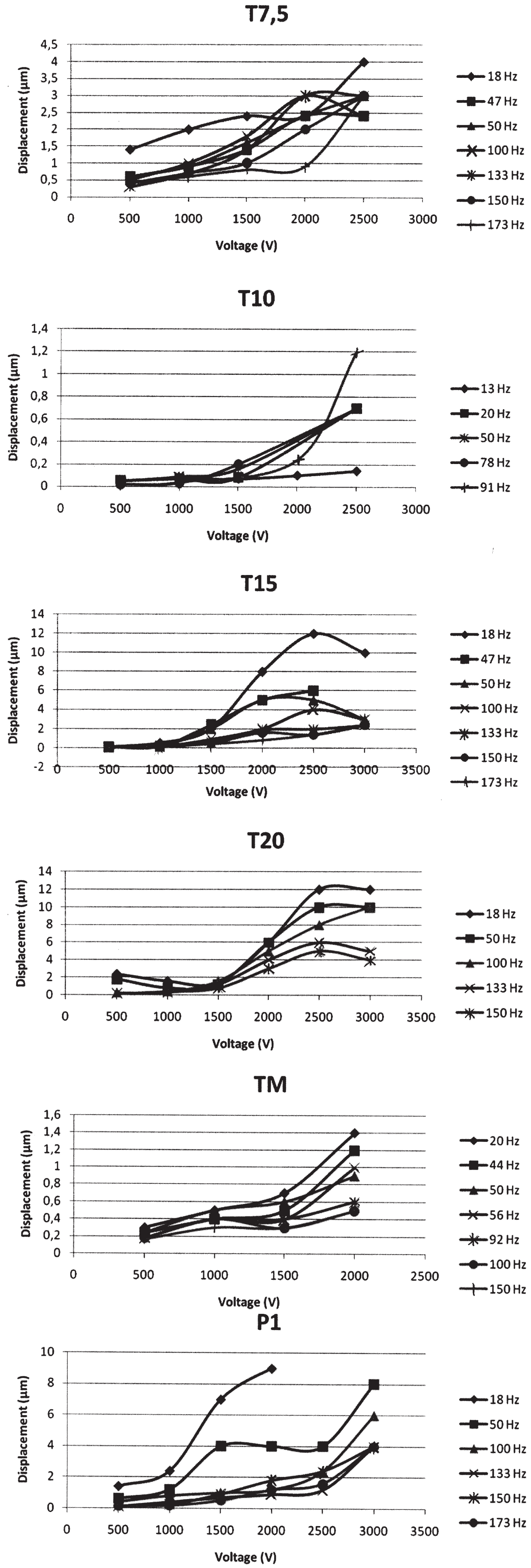


Fig. 5. Strain versus voltage at different resonance frequencies

Displacements increase with voltage and maximum values are obtained at voltages ranging from 1500 to 2500 V.

Also, it can be observed that the sample T20 which have the highest dielectric permittivity (ε =4.1) and TiO_2content (9 wt.%) showed the best performance and a dynamic actuation sensitivity of 2.4µm/kV_{pp} at resonance frequency of 18 Hz.

Conclusions

The Scanning Laser Doppler Vibrometryhis a non-destructive method for dynamic caracterisation of dielectric EAP. It was applied to investigate the resonance frequencies and vibrational modes of rectangular film samples in free-boundary conditions stimulated with high sinusoidal voltages. This laser-based technique has a good accuracy and allowed measuring the dynamic frequency response of dielectric elastomer materials in a frequency range of up to 200 Hz.

The results obtained on PDMS/TiO_2/SiO_2 composites films showed relatively good actuation performance considering that the aluminum electrodes are not very compliant compared to carbon-based electrodes. Furthermore, using compliant electrodes and reducing the thickness of samples will allow us to develop diaphragm actuators with good actuation capability as possible candidates for micro-vales or micro-pumps applications in MEMS technology.

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