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PROPERTIES OF POLYURETHANE THIN FILMS

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On the basis of the electrostatic field induced strain, dielectric and stress-strain measurements, the electromechanical properties of a polyurethane elastomer film based polyester were investigated. Up to about 7 MV/m, the induced strain showed a quadratic dependence on the applied electric field strength, which is consistent with an electrostrictive response. A strain of about 7.5 %, higher than those previously reported in the literature, was obtained. A remarkable apparent electrostrictive coefficient of about $7x10^{-16}$ m^2/V^2 was found. The response time of the induced strain was found less than 700 ms, which means that the deformation of film is quite fast. On the basis of the measured values of Young's modulus and dielectric constant, the contribution of the Maxwell effect of 3.07 % was deduced. The investigated polyurethane shows some important characteristics needed in applications as actuators, sensors and transducers.

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1. Introduction

Since the report by Zhenyi and co-authors [1] in 1994 on the electromechanical properties of a commercial polyurethane elastomer, much attention has been paid on this type of polyurethane [2-11] due to their some remarkable characteristics such as large strain, high specific energy and small response time. Moreover, these elastomers are lightweight, pliable, low manufacturing cost, inexpensive and easily moulded into any desirable shapes. These make the polyurethane elastomers very attractive for a wide range of electromechanical applications such as sensors, actuators and transducers [6,8,11]. The electromechanical properties of this material have not been completely investigated and the fundamental mechanisms responsible for electrostriction are not yet well understood. Besides, in polymeric elastomers, including polyurethanes, the contribution of the Maxwell effect to the strain response could be significant. In order to solve these problems, besides commercial polyurethanes, new polyurethane elastomers must be synthesised and characterised. The goals of our paper are (1) to determine the electromechanical properties of a polyurethane elastomer based polyester and (2) to measure its dielectric and mechanical properties in order to establish the contributions of the effects responsible for electrically actuation in the respective polymer.

2. Experimental

A polyurethane elastomer based on poly(ethylene adipate), 4,4'-methylene bis(phenylisocyanate) and 1,4-butanediol with the molar ratio 1:6:5 was synthesised and used. Polymer films with a thickness of 0.055 mm was prepared by casting dilute polymer solution on glass plate and then dried for 24 h at 60° C under air atmosphere. The electric field induced strain in thickness direction of film was measured under ambient conditions using a modified Michelson interferometer and a He-Ne laser as light source. In order to hold the film in horizontal position, the vertical movable mirror from the standard Michelson interferometer was replaced by a $\pi/4$ inclined mirror. The polymer films were placed between two opposing metallic disks acting as electrodes. On the upper thin disk, a lightweight mirror was placed. A

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very thin layer of hydrocarbon fluid was applied between the metal disks electrodes and film. This was done in order to minimise the electrode constraint by reduction the frictional force on the film as it expends laterally during the electric field induced decrease in thickness. In the same time the fluid layer reduces any measurement error produced by air trapped between the electrodes and sample and holds the film flat and perpendicular to the laser light. Any thickness change of the film under applied electric field produces a change of phase between the two recombined lights: one reflected from the fixed vertical mirror and the other one reflected from the mirror placed horizontally on the upper electrode. A photometer converted the changeable interference fringes into an electric signal, which is than displayed on a store oscilloscope. Finely from the electric signal the thickness deformation of the polymer film was calculated. Dielectric constant and dielectric loss measurements were carried out on films with vacuum evaporated silver electrodes by a BM 507 TESLA impedance meter in the frequency range from 50 Hz to 500 kHz under ambient conditions.

The Young's modulus was determined from the stress-strain curve recorded in air at room temperature with a cross-head speed of 5.8 mm/ min. using a tensile testing equipment presented elsewhere [12]. The dumbbell-type specimen was 10 mm wide at the two ends and 2 mm wide and 10 mm long for the neck.

3. Results

Fig. 1 shows the induced strain as a function of the applied electric field. The strains were measured one minute after the application of the electric field. The electric field was brought to zero after each measurement. It was observed that the strains were not entirely recovered over the period considered here as the electric field decreases to zero.

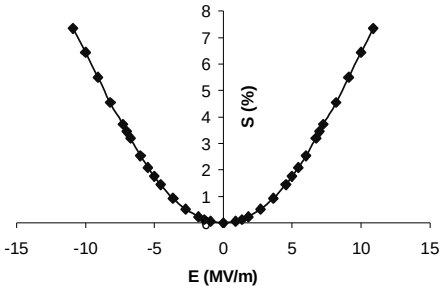

Fig. 1. Dependence of strain on electric field.

Compressions in the thickness direction of the film were observed irrespective of the applied electric field sign. Also a symmetrical strain profile was found against inversion of the electric power polarity. At lower electric field strengths, up to about 7 MV/m, the induced strain shows a quadratic dependence, whereas at higher values rather a linear one exists. For the highest applied field (~ 11 MV/m) remarkable strain of about 7.5 % was found. This value is larger than those previously reported on polyurethane elastomer films with more or less restraining electrodes: ~3 % for ~20 MV/m [1] and 1.34 % for 0.4 MV/m [2].

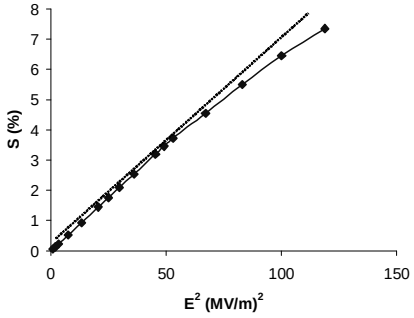

Fig. 2. Dependence of strain on square electric field.

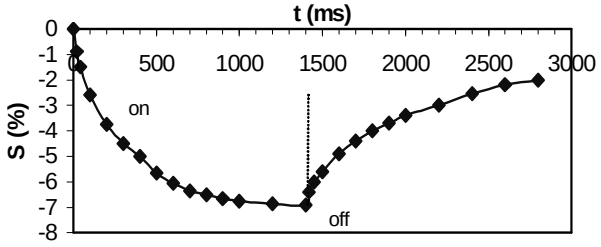

Fig. 3. Time response of contraction and relaxation processes.

The electric field dependence of the strain may be more clearly observed in the Fig. 2, where the strain is plotted versus the square of the electric field. From the slope of the straight line corresponding to the lower electric fields, an apparent electrostrictive coefficient of 7.07×10^{-16} m^2/V^2 was determined. This result is about 10 times larger than the largest electrostrictive coefficient reported on the less constrained polyurethane films [1].

The time dependence of the induced strain during the contraction and relaxation processes are shown in Fig. 3. The contracting response is quite quick (~700 ms) and shows a steady strain. The strain is more slowly when the electric field is off.

The dielectric behaviour was characterised over the frequency range from 50 Hz to 400 kHz and the typical results are presented in Fig. 4. An important frequency dispersion of the relative dielectric constant from ~8.8 to ~5.0, over the whole frequency range, was found. These results reflect the existence of a large variation in the local environment and length of the chain segments involved in the polarization process.

The stress-strain curve shows a typical profile for a polymeric elastomer (Fig. 5). From the initial part of the curve, where the Hooke's low is supposed to be valid, the Young's modulus was determined and a value of 3.6 MPa was obtained.

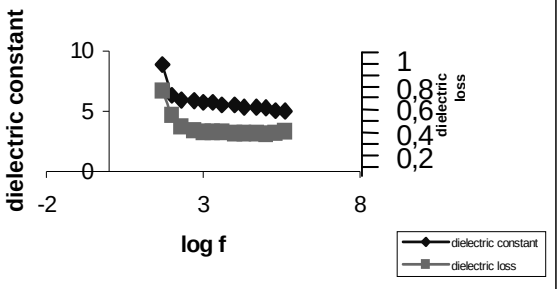

Fig. 4. Dielectric constant and dielectric loss as a function of frequency.

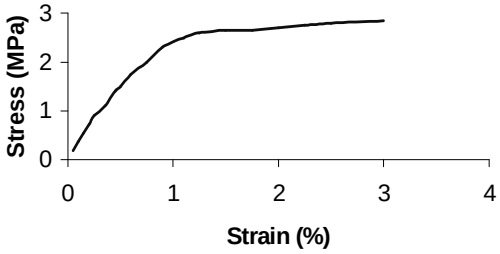

Fig. 5. Stress – strain curve.

4. Discussion

In an isotropic material, such as polyurethane elastomer, the strain induced by an electric field can be caused by two effects [1,3,5,11]: the electrostrictive effect that expresses the dependence of strains on the dielectric properties and the Maxwell effect which is mainly due to the interaction between the free charges on the electrodes (Coulomb interaction). Strains resulting from both effects are proportional to the square of the applied electric field.

If S denotes the strain and E is the applied electric field, the electrostrictive effect can be expressed [3,11] as

$$S_E = Q\varepsilon_0{}^2(\varepsilon_r - 1)^2 E^2 = ME^2 \tag{1}$$

where ε_r is the relative dielectric constant, ε_0 is the vacuum dielectric permitivity, Q is the electrostrictive coefficient of the polymer and M is a parameter often used [1,3,11] to describe the experimental dependence of strain response of the material on the applied electric field.

On the other hand, the thickness change of an unconstrained film due to the Maxwell effect is obtained as [13]

$$S_M = -\frac{1}{2}\frac{\varepsilon_0\varepsilon_r}{Y}E^2(1+2\mu) \tag{2}$$

where Y is the Young's modulus and μ is the Poisson's ratio.

Our results show a quadratic dependence of the strain on the applied static electric field up to about 7 MV/m. It suggests that one or both of the above effects could be involved. For higher electric fields the strain depends rather linearly on the electric field suggesting that the polymer has non-linear dielectric properties.

In order to identify the contributions of the mechanisms above, the dielectric constant and the Young's modulus were determined. Because the most elastomeric polymers, including polyurethanes, are

essentially incompressible in volume, the Poisson's ratio can be considered close to 0.5 value [6,11,13]. Then the equation (2) can be written as

$$S_M = -\frac{\varepsilon_0 \varepsilon_r}{Y} E^2 \tag{3}$$

In our case, where the electrodes are metallic disks, the mechanical constraints originate both in the constraining effect of the electrodes (caused by the frictional force between electrodes and film) and the opposing lateral stresses generated by the inactive film from the outside of the disks. During the lateral extension of the film, this inactive part of the elastic film acts as a spring during its contraction.

When the electrodes are thin layers of vacuum evaporated metals [1], the mechanical constraints come mainly from the metallic layer itself (Young's modulus of the metallic layer is higher than that of the polymer film). In order to determine the closest value of the real induced strain, both constraining effects have to be minimised. For this reason, we applied a thin hydrocarbon liquid layer between the metal disks electrodes and film and used a film with its total area much larger than the area of the electrodes. The absence of the air between electrodes and film, due to the presence of the liquid layer, does also improve the thickness strain deformation.

Under these experimental conditions it is reasonable to use the relation (3) for calculation the Maxwell effect contribution to the global induced strain.

For the dielectric constant of 8.8 corresponding to the lowest frequency (Fig. 3) and the Young's modulus of 3.6 MPa, the percentage contribution of the Maxwell effect was found equals to 3.07 %. After subtraction of the Maxwell strain from the measured induced strain, the pure electrostrictive strain can be obtained.

The induced strains and electrostrictive coefficient were found larger than those reported in the literature on the similar polyurethane elastomers [1,2]. The type of the electrodes-film ensemble used for the electric field induced strain measurements has an essential role. Chemical and physical structures of the polymer film could also be important. Futher studies are being carried out to determine the structure-electromechanical properties relationship.

5. Conclusions

The investigated polyurethane elastomer shows remarkable electrostatic field induced strain and apparent electrostrictive coefficient, higher than those reported in the literature on similar polyurethanes.

These results can be attributed both to the preparation mode of the electrodes-film ensemble and physical and chemical structure of the polyurethane film.

The pure electrostrictive effect is more important than the Maxwell effect.

The obtained electromechanical characteristics recommend this polymer as a potential material in applications as actuators, sensors and transducers.

References

[1] M. Zhenyi, J. I. Scheinbeinm, J. W. Lee, B. A. Newman, J. Polym. Sci., Part B **32**, 2721 (1994).
[2] T. Hirai, H. Sadatoh, T. Ueda, T. Toshiaki, Y. Kurita, M. Hirai, S. Hayashi, Angew. Makromol. Chem. **240**, 221 (1994).
[3] Q. M. Zhang, J. Su, C. H. Kim, R. Ting, R. Capps, J. Appl. Phys. **81**, 2770 (1997).
[4] J. Su, Q. M. Zhang, R. Y. Ting, Appl. Phys. Lett. **71**, 386 (1997).
[5] J. Su, Q. M. Zhang, C. H. Kim, R. Y. Ting, R. Capps, J. Appl. Polym. Sci. **65**, 1363 (1997).
[6] R. E. Pelrine, R. D. Kornbluh, J. P. Joseph, Sensors and Actuators **64**, 77 (1998).
[7] J. Su, Q. Zhang, P. Wang, A. G. MacDiard, K. J. Wynne, Polym. Adv. Technol. **9**, 317 (1998).
[8] R. Kornbluh, R. Pelrine, V. Shastri, 1st NASA/JPL Workshop on Biomorphic Explorers, 1998.
[9] R. Liu, Q. Zhang, L. E. Cross, J. Appl. Polym. Sci. **73**, 2603 (1999).
[10] R. Pelrine, R. Kornbluh, Q. Pei, J. Joseph, Science, 287 (2000).
[11] Y. Bar-Cohen, Electroactive Polymer (EAP) Actuators as Artificial Muscles. Reality, Potential and Challenges, SPIE Press, Washington (2001).
[12] P. Coman, M. Gaspar, I. Diaconu, Romanian Reports in Phys. **45**, 575 (1993).
[13] I. Krakovsky, T. Pomijn, A. Posthuma de Boer, J. Appl. Phys. **85**, 628 (1999).