

EFFECT OF TARGET MATERIAL ON THE LASER INDUCED PRESSURE PULSE MEASUREMENTS FOR THICK P(VDF-TrFE) COPOLYMERS

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Abstract

The laser induced pressure pulse method has been used to study the non-uniform polarization distribution in 0.8 mm thick vinylidene fluoride/trifluoroethylene copolymer (P(VDF-TrFE)) films. Sprayed-on black paint was initially used as the target material and this led to the generation of a bipolar pressure pulse in the sample. With a different target material, we can now produce a monopolar pressure pulse. Measurements have been carried out using the new target material and the piezoelectric coefficient e_{zz} has been estimated.

1. Introduction

Polyvinylidene fluoride (PVDF) and copolymers of vinylidene fluoride and trifluoroethylene P(VDF-TrFE) are widely used as transducers. The advantage of P(VDF-TrFE) is that it can be poled without prior stretching. In most application, the P(VDF-TrFE) films are fairly thin (~10 μm). However, the P(VDF-TrFE) films used in underwater sonar application could be as thick as several hundred μm because of the requirement of lower frequency. If the applied voltage during the poling process is not sufficiently high, the polarization in a P(VDF-TrFE) sample is not uniform, thereby resulting in peculiar resonance characteristics[1-3]. Therefore, it is important to know the polarization distribution in piezoelectric materials.

The laser induced pressure pulse (LIPP) method, also known as the pressure wave propagation (PWP) method, have been used to study to the polarization of piezoelectric materials[4, 5]. Frequently, the pressure pulse entering the sample is bipolar[1, 5, 6], and the reason of this phenomena is still unclear. In this paper, different target materials were used to produce a monopolar pressure pulse. The polarization distributions were determined and the piezoelectric coefficients e_{zz} were calculated.

2. Measurement principle

The experimental setup is shown in Fig. 1. The sample was irradiated by

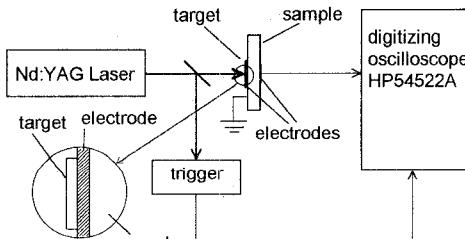


Fig. 1 The set-up for LIPP Measurements

a laser pulse with 10 ns duration and a beam diameter of 8 mm. The diameter of the target was 9 mm whereas the diameter of the sample electrode was about 12 mm. The laser pulse energy was optimised according to the target material used in the measurement. In order to measure the short circuit current, the input resistance of the digitizing oscilloscope was set to 50 Ω .

The principle of LIPP method has been described in detail elsewhere[4, 5]. The absorption of a laser pulse by the target leads to the generation of a pressure pulse, which in turn gives rise to a short-circuit current flowing in the external circuit during its propagation through the sample. From Ref. [7], the short-circuit current for piezoelectric materials if there is no space charge in the sample can be expressed as,

$$I_{cc}(t) = -\frac{S\chi}{d_0} \int_0^{d_0} e_{zz}(z) \frac{\partial p(z,t)}{\partial t} dz \quad (1)$$

where S is the laser beam area, χ is the compressibility of the materials, d_0 is the original thickness of the sample, $p(z, t)$ is the pressure wave profile as a function of time and position, and $e_{zz}(z)$ is the distribution of piezoelectric coefficient.

For a uniformly poled sample, during the penetration of the pressure pulse into the sample ($z = a$), the current measured is directly proportional to the pressure wave near the entrance of sample,

$$I_{cc}(t)|_{z=a} = -\frac{S\chi}{d_0} v e_{zz}(a) p(a,t), \quad (2)$$

where v is the acoustic velocity of pressure pulse.

3. Piezoelectric coefficient e_{zz} estimation

An x-cut quartz with known e_{11} is used to estimate the piezoelectric coefficient e_{zz} of sample[3, 5]. Assume that the laser pulse amplitude and the beam area are approximately constant from pulse to pulse and the same target material is applied to the sample and the x-cut quartz. Considering the conservation of momentum $[M] = S \int p(t) dt \approx S p \tau$ (τ is the width of the pressure pulse) in equation (2) for quartz and the copolymer sample, the piezoelectric coefficient can be expressed as,

$$e_{zz} = \frac{I_0}{I_{0q}} \frac{\rho}{\rho_q} \frac{v}{v_q} \frac{d_0}{d_{0q}} \frac{\tau}{\tau_q} e_{11} \quad (3)$$

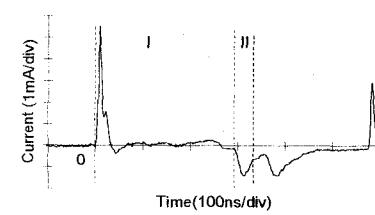
where, I_0 is the peak current amplitude when the pressure pulse enters the sample, and ρ is the density of the sample. The quantities with subscript q denote the corresponding parameters for quartz. In this paper, a 0.8 mm thick x-cut quartz was used for the estimation of e_{zz} .

4. Experimental results

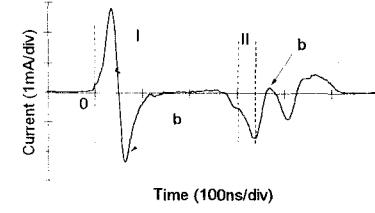
Four P(VDF-TrFE) (80/20) copolymer films, A, B, C and D (about 0.8 mm thick) supplied by North America Atochem were studied in this work. Sample A was melted and hot-pressed at 180°C for 10 minutes. Sample B was as-received and had not been heat-treated before poling. Samples C and D were annealed at 120°C for two hours. Samples A, B, C and D were poled at 108°C, which is below the Curie temperature for first heating ($\uparrow T_c=124^\circ\text{C}$), for two hours with an applied electric field of 26 kV/mm. The electric field was kept on while the samples were cooled to room temperature. Sample D was poled again at 108°C for another two hours (two-step poling)[2]. Table 1 gives a summary of the thermal treatments before poling and the types of electrodes and targets used.

Since sample A was flat we could use a 1.8 mm thick aluminum disk as the target, which was attached to the sample (with gold electrodes) by a couplant (Panametric B2). As Al has a high reflectivity, the pulse energy of the laser was raised to 300 mJ to

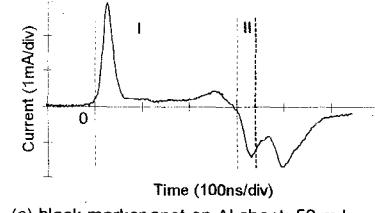
produce a sufficiently strong signal. Fig. 2(a) shows that a monopolar pressure pulse is generated if an Al target is used. Much less energy (50 mJ) is required to generate a strong signal if black paint sprayed on the Al disk is used as the target (Fig. 2(b)). However, a bipolar pressure pulse is produced (the peaks marked b). As shown in Fig. 2(c),



(a) Al target, 300 mJ laser.

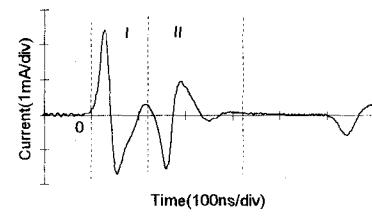


(b) black paint on Al sheet, 50mJ laser.

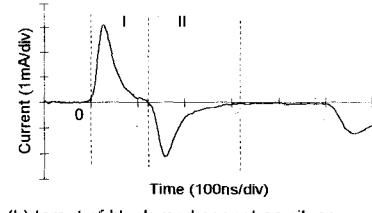


(c) black marker spot on Al sheet, 50 mJ laser.

Fig. 2 Current for sample A



(a) target of black paint on silver paint electrode.



(b) target of black marker spot on silver paint electrode

Fig. 3 Current for sample B

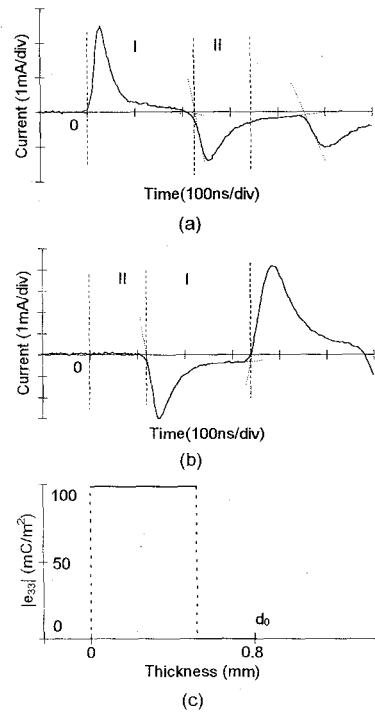


Fig. 4 Current and polarization profile for sample C

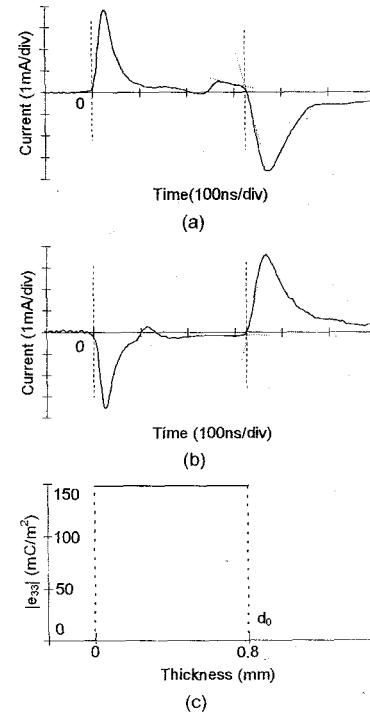


Fig. 5 Current and polarization profile for sample D

if the target is a black spot made by a marker pen on the surface of the Al disk, a monopolar pressure pulse is observed. It can be deduced from Figs. 2(a) to 2(c) that sample A is partially poled through its thickness, with the poled zone I occupying almost 90% of the total thickness and the unpoled zone II occupying the remaining region.

Samples B, C and D are slightly curved after poling, hence the coupling between these samples and the aluminum target are poor. Therefore for sample B, two different black absorbers (black paint and black marker) were in turn painted directly on the silver electrode as targets. When black paint was used as the target, a bipolar pressure pulse was produced (Fig. 3(a)). If black marker spot was used, a monopolar pressure pulse was produced (Fig. 3(b)), but the monopolar pulse was almost as broad as the bipolar pulse. Again, zone I and II represent the poled and unpoled zone, respectively. Sample B is also partially poled with the poled zone occupying about 40% of the thickness.

For samples C and D, a black marker spot is used as the target and a monopolar pressure pulse is observed (Figs. 4(a), 4(b) & 5(a), 5(b)). Figs. 4(a) and 5(a) show the short circuit current when the pressure pulse enters the anode of the sample, which is the side connected to positive voltage during poling. Fig. 4(b) and Fig. 5(b) show the short circuit current when the pressure pulse enters from the cathode side of the sample. The schematic polarization distribution shown in Fig. 4(c) shows that sample C is partially poled with the poled zone I occupying about 60% of the thickness. Sample D, poled using a two step poling method[2], is fully poled (Fig. 5(c)). The piezoelectric coefficient of selected samples are given in Table 1.

Table1: The thermal treatments before poling for all the samples and their characteristics.

Sample	Heat-treatment before poling	Electrode	Poling	Target	Shape of pressure wave	$ e_{33} $ (mC/m ²)
A	melted & hot-pressed at 180°C	evaporated gold	one-step (0.9d)	Al	monopolar	—
				Al+black paint	bipolar	
				Al+black mark	monopolar	
B	as received	silver paint	one-step (0.4d)	black paint	bipolar	—
				black mark	monopolar	
C	annealed at 120°C for 2 hours	silver paint	one-step (0.6d)	black mark	monopolar	108
D	annealed at 120°C for 2 hours	silver paint	two-step (fully poled)	black mark	monopolar	98
						148

5. Discussions

Sample A, B and C are poled under almost identical poling conditions. The poled zones in sample A, B and C have dimensions of about 0.9d, 0.4d and 0.6d, respectively. This shows that thermal treatment of the samples before poling affects the resulting polarization. It is best to melt and hot-press the sample before poling. By using a two-step poling method (sample D), a uniformly poled sample with a high $|e_{zz}|$ value can be obtained.

The occurrence of a bipolar pressure pulse probably arises from several reasons. One possible reason is that the target material may have formed a constraint layer on the surface of the sample[8]. It is also possible that the laser beam has penetrated a certain depth of the target and is absorbed; the pressure pulse that is produced at that position travels towards the target/air interface is reflected. A phase reversal occurs and this gives rise to the negative peak in the observed pressure pulse[9]. By choosing suitable target materials, the bipolar phenomena can be reduced or eliminated.

A comparison of various targets shows that Al target gives the narrowest monopolar pressure pulse and this will lead to a high spatial resolution. One problem is that the samples tend to curve during poling and it is not possible to make good contact if an Al disk is used as the target. To produce a monopolar pressure pulse, we have used a target of black marker spot on silver paint electrode. The shortcoming of this target

material is that the black mark is easily ablated by the laser beam and a fresh layer of the target has to be painted after each shot.

6. Conclusion

The bipolar peak arises from the interaction between the laser beam and the target. By choosing a suitable target material, the bipolar phenomena can be reduced or removed and a monopolar pressure pulse can be generated.

By using the LIPP method, both the magnitude and the position dependence of the piezoelectric coefficient $|e_{zz}|$ can be obtained. If the applied voltage is not high enough during the one step poling process the polarization distribution in P(VDF-TrFE) sample is rather non-uniform. Using the two step poling method, P(VDF-TrFE) samples with uniform polarization can be produced.

ACKNOWLEDGEMENT

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REFERENCES

- [1] Laburthe-Tolra, C. Alquier and J. Lewiner, "Polarization of VDF-TrFE Films at Elevated Temperature", IEEE Trans. Electr. Insul., Vol. 28, pp.344-348, 1993.
- [2] H. L. W. Chan, Z. Zhao, K. W. Kwok and C. L. Choy, "Enhancement of Piezoelectric Activity in P(VDF-TrFE) Copolymer using Two-step Poling", Proc. 8th Intl. Symp. on Electrets (ISE8), Paris, France, 1994, pp.583-588.
- [3] H. L. W. Chan, Z. Zhao, K. W. Kwok, C. L. Choy, C. Aliqué, C. Boué and J. Lewiner, "Polarization of Thick P(VDF-TrFE) Copolymer Films", to be published, J. Appl. Phys., 1996.
- [4] C. Aliqué G. Dreyfus and J. Lewiner, "Stress-Wave Probing of Electric Field Distributions in Dielectrics", Phys. Rev. Lett., Vol. 47, pp.1483-1487, 1981.
- [5] R. Gerhard-Multhaup, G.M. Sessler and J.E. West, "Investigation of Piezoelectricity Distributions in Poly(vinylidene fluoride) by means of Quartz- or Laser-generated Pressure Pulses", J. Appl. Phys., Vol. 55, pp.2759-2775, 1984.
- [6] G. M. Sessler and G. M. Yang, "Spatial Distribution of Trapped Charge in Electron-Beam Charged Polypropylene", 1991 Annual Report of CEIDP, Knoxville, Tennessee, U. S. A., p.108-113, 1991.
- [7] C. Laburthe-Tolra, Doctoral thesis, "Etude des mécanismes de polarisation de polymère polarisation de polymères piézoélectriques, par la méthode de l'onde de pression", University Paris 6, 1991.
- [8] C.B. Scruby and L.E. Drain, "Laser Ultrasonics: Techniques and Applications", Adam Hilger, Bristol, 1990.
- [9] D.A. Hutchins, "Mechanisms of Pulsed Photoacoustic Generation", Can. J. Phys., Vol. 64, pp.1247-1264, 1986.