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Potentialities of flexoelectric effect in soft polymer films for electromechanical applications

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Abstract. Among the transduction mechanisms of interest for sensing and/or actuation applications at nano/micro scale, the piezoelectric effect has been widely exploited owing to the solid state nature of piezoelectrics, the large ability of specific classes of materials for the mechanical-to-electrical energy conversion and easy integration. However, every piezoelectric (also generally ferroelectric) presents well-known intrinsic drawbacks such as required poling step and related aging. In contrast, uniquely flexoelectric materials do not suffer from these disadvantages because flexoelectricity, a universal effect in all dielectric solids defined as the electrical polarization induced by a strain gradient, does not imply preliminary electric field-induced macroscopic polarization. Besides, strain gradient may be easily obtained by bending plate or cantilever-shaped structure and in this case it is nothing but the local curvature of the flexible system. Thus, as strain gradient (curvature) inversely scales with both elastic stiffness and thickness, this study will focus on the evaluation of the potentialities of flexoelectric effect in soft polymer films for electromechanical applications, with an emphasis on the thickness influence. In this way, analytical results combined to experimentally obtained effective flexoelectric coefficients for some typical polymer classes may provide guidelines for the development of soft and low frequency flexoelectric mechanical transducers.

1. Introduction

Although it was theoretically described in the late eighties by Tagantsev [1], flexoelectricity, which is a linear electromechanical coupling, has recently become a hot topic in the materials science community, in the last 5 years. The main reason of this revival is the possibility to exploit flexoelectric effect in micro/nano electromechanical systems (MEMS/NEMS), being currently developed for both sensing and actuation applications along with the ongoing advances in nanotechnology. As a matter of fact, *direct* flexoelectricity, which is a universal effect in all solid dielectrics corresponds to the appearance of an electric polarization induced by a strain gradient, defined for instance in the thickness direction by $P_{3flex} = \mu_{1133} \frac{\partial S_{11}}{\partial x_3}$ (1), where μ_{1133} is the transverse flexoelectric coefficient, S_{11} is the axial strain (1-direction is the axial direction of the clamped cantilever-shaped sample in figure 1) and x_3 is the position coordinate along thickness direction. Thus, although flexoelectric coefficients are presently two or three orders of magnitude lower than piezoelectric ones (e_{31}), yielding the electrical polarization $P_{3piezo} = e_{31} S_{11}$ (2), strain gradients may reach huge values at small scales, up to 10^6 m^{-1} , for instance in epitaxial thin films. Clearly, the strain gradients in thin films are significantly larger than the corresponding strain values ($\sim 1\%$) and the strain gradients in the bulk form of the material, so P_{3flex} may exceed P_{3piezo} in specific cases. Bibliography on



flexoelectricity in elastic dielectrics reveals that in the last ten years many studies have been devoted to both theoretical and experimental works on *oxides* (bulk and thin film forms), and two major trends have been demonstrated and validated. First, flexoelectric coefficients scale with the dielectric constant (K). So, high- K materials should be privileged for obtaining the best flexoelectrics. For instance, perovskite materials like Barium Strontium Titanate (BST) ceramic [2] exhibits the largest flexoelectric coefficients, in the range 10^{-6} - 10^{-5} C/m, associated with a large K (~ 13000). Second, the flexocoupling coefficient F , defined as the ratio of μ coefficient over the dielectric constant K : $F = \mu/\varepsilon_0 K$ (3), where ε_0 is the vacuum permittivity, should be quasi constant and lie between 1 V and 10 V, independently on the thickness [3]. Compared with oxide films, few studies have been devoted to the flexoelectric effect in organic materials but it was recently published that flexoelectric coefficients in thermoplastic and thermosetting polymer thick (~ 500 μm) films of PVDF (polyvinylidene fluoride in its non-piezoelectric form) or polyethylene (PE) are about 1-10 nC/m [4]. Even if these μ coefficients are three orders lower than those of BST bulk ceramics, the flexibility (i.e. low Young's modulus Y) and lightweight of soft polymers render them interesting for sensing and actuating applications. Thus, in the present study, we report experimental results on the thickness dependence of the effective transverse flexoelectric and flexocoupling coefficients of some soft polymer films and analytical results to assess if their use may be envisaged as transducer materials in specific applications such as mechanical energy harvesting or curvature sensing.

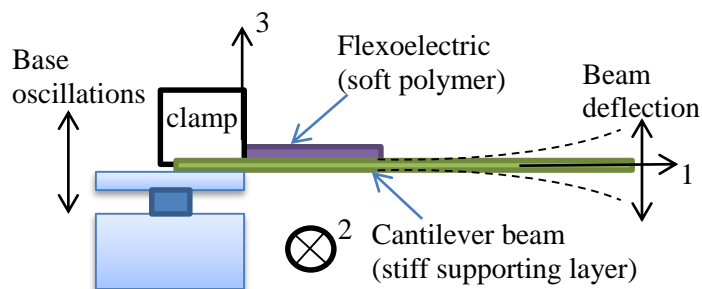


Figure 1. Experimental set-up for flexoelectric characterisation with a cantilever-shaped bilayer (flexoelectric/passive steel substrate) deflected by base oscillations

2. Experimental procedure

The studied polymers have been selected for their relative softness ($Y < 1\text{GPa}$) and for their different global polarity: a polyurethane (PU) grade, which is a semi-crystalline thermoplastic elastomer with a block copolymer morphology. Non-piezoelectric but polar (α -form) PVDF and also a semi-conducting polythiophene (PT)-based blend have also been tested. The two former polymers are considered to be insulating while the latter (PT blend) exhibits a semi-conducting behavior. The ~ 200 μm -thick PU polymer films have been prepared by dissolving the commercially available granules in N, N-dimethylformamide (DMF) and the solutions have been homogeneously poured onto a stainless steel substrate using a film applicator and then dried at 40°C for 20 h and subsequently annealed at 130°C for 3 h. PU and PVDF films with thicknesses lower than 5 μm have been prepared by spin coating onto the same substrate. Then, the deposited films were annealed on a hot plate at 95°C for 5 min to evaporate excess of DMF. PT-blend films have been deposited onto stainless steel substrate by drop casting method using a commercial aqueous PT solution and dried at 60°C for 2h. The resulting thicknesses lie between 3 and 8 μm . The stiff (200 μm thick and $Y = 209$ GPa) stainless steel substrate serves both as a bottom electrode and also as a supporting layer to ensure a pure bending mode of the cantilevered beam (figure 1) clamped at one end. In fact, the beam deflection magnitude must be smaller than the dimensions of the cantilever to use the assumptions of the Euler-Bernoulli beam theory. In this case, the local beam curvature used for the determination of the flexoelectric coefficient may be calculated from the measured deflection. Experimental and calculation details are given in our previous work [5]. A top point electrode of Aluminum with 100 nm in thickness was deposited by evaporation on the top surface of the polymer/steel bilayer (unimorph structure) for dielectric and direct flexoelectric measurements. Direct (current output) flexoelectric effect have been measured using a lock-in amplifier tuned to the frequency of the applied curvature (i.e. controlled cantilever bending).

3. Results and discussion

3.1. Flexoelectric and flexocoupling coefficients

Figure 2 shows the *direct* effective flexoelectric coefficients measured at 10 Hz as a function of the thickness, in quasistatic condition since the first resonance frequency of the unimorph cantilevers is close to 165 Hz, imposed by the stiff supporting steel layer.

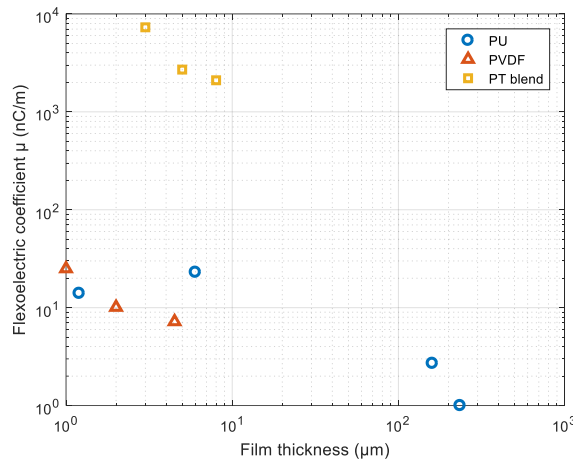


Figure 2. Flexoelectric coefficient μ of the polymer films versus thickness.

The PU and PVDF films exhibit μ coefficients in the 1 nC/m- 30 nC/m range, with a slight thickness dependence. These results are consistent with the μ coefficients previously given in the literature for such insulating polymer films [4]. In comparison, the semi-conducting PT blend films present very large *flexoelectric-like* μ coefficients, because, by definition, only dielectrics are flexoelectric. The largest μ of the PT blend series is 7300 nC/m, with a decrease with increasing thickness, even if further film thicknesses should be tested to confirm the trend. The thickness dependence of the flexocoupling constant F is more obvious (figure 3), but the largest F values are globally obtained for PVDF and PU films, in the 1-10 μ m range. This opposite trend in comparison with that of μ coefficient is due to the huge dielectric constants measured at 100 Hz of the PT series ($K \sim 6000$ -19000), significantly larger than those of PVDF and PU series (7-10). Finally, figures 2 and 3 reveal that very large flexoelectric-like coefficients may be achieved, in agreement with a scaling effect with the dielectric constant, but the F coefficient, which is of significant importance because it reflects the curvature (ζ)-induced flexoelectric field E ($E = F \cdot \zeta$), is not necessarily enhanced by large μ values. Besides, the order of magnitude F coefficient of the studied polymers may reach large values [6], in comparison with the theoretical estimate (1-10 V)[3], experimentally validated in many oxides. These results strongly suggest that the measured response is not only due to bulk effect but also surface contributions like the so-called surface flexoelectricity and possibly surface piezoelectricity [3], depending on the insulating or moderately conducting nature of the polymer.

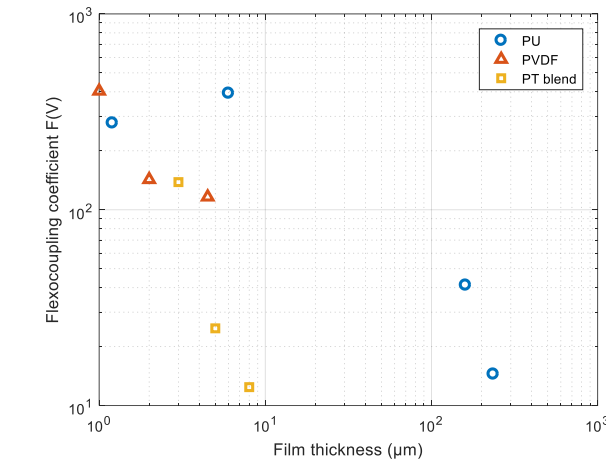


Figure 3. Flexocoupling constant F of the polymer films versus thickness.

3.2. Flexoelectric energy conversion

Figure 1 represents the typical case of base-excited cantilever, which is a widely used structure in the field vibration energy harvesting using piezoelectric material clamped on the stiff beam. In the case of flexoelectric energy conversion, the same unimorph structure may be employed but also more simply a single flexoelectric cantilever without supporting layer since strain gradients (i.e. cantilever curvature) are the consequence of beam bending, whatever the location of the neutral axis. For estimating the potential of the studied polymer films, a Figure of Merit (FoM) for energy harvesting is proposed from a simple electrokinetic model, where the flexoelectric film is modelled by a current source in parallel with its clamped capacitance [7] and connected to a resistive electrical load, in which the dissipated power density equals the harvested one. For a fixed flexocoupling coefficient F , the optimal harvested power density as a function of the curvature ζ is expressed by: $P =$

$\mu^2 \omega \zeta / (\epsilon_0 K)$ (4), where ω is the angular frequency of the beam vibrations. Thus, from the viewpoint of the material, the Figure of Merit is $FoM = \mu^2 / (YK)$ (5), since under a fixed base excitation, ζ increases with decreasing Y .

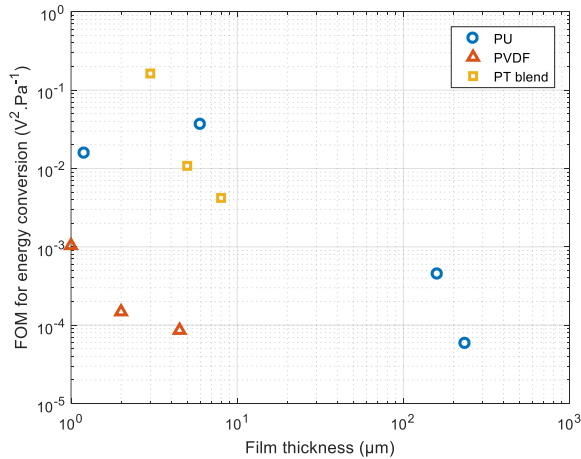


Figure 4. Figure of Merit for flexoelectric energy conversion of the polymer films versus thickness.

Figure 4 gives the FoM of the different polymer films as function of the thickness. The FoM was calculated using the bulk Young's modulus of the polymer, obtained from literature or supplier data. Figure clearly shows that thin (<10 μm) films of PU and PT blend present the best characteristics combination for flexoelectric energy harvesting purpose. The difference between the FoM of PU and PVDF mainly originates from their difference in Young's modulus ($Y_{PU}=28$ MPa and $Y_{PVDF}=1.1$ GPa), because their μ and K coefficients are of the same order of magnitude. Despite their huge dielectric constant values, the large flexoelectric μ coefficient of PT blend films (figure 1) contributes to the large FoM since the elastic modulus $Y_{PT\ blend}=700$ MPa is close to that of PVDF.

4. Conclusion

In this study, quasistatic flexoelectric current measurements have been carried out for three polymer film series in the 1-100 μm range: polyurethane (PU), polyvinylidene fluoride (PVDF) and polythiophene based compound (PT blend). The semi-conducting PT blend films present the largest flexoelectric-like μ coefficients (up to ~ 7000 nC/m), in comparison with the μ coefficients of PU and PVDF in the 10 nC/m range. Two figures of merit have been calculated from measured flexoelectric and dielectric characteristics: the fundamental flexocoupling F coefficient, which is high for PU and PVDF films and the FoM for flexoelectric energy conversion (harvesting), which highlights the potential of both PU and PT blend. These measurements and given trends indicate that the flexoelectric μ coefficient is not the only key factor determining the ability for a polymeric material to be integrated in electromechanical applications. Thus, high μ value may be suitable for curvature sensing, but not necessarily for mechanical energy harvesting. At last, the thickness dependence of the coefficients accompanied with large F values may originate from surface contributions, which need to be experimentally evaluated.

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