



Enhanced strain response and energy harvesting capabilities of electrostrictive polyurethane composites filled with conducting polyaniline



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ABSTRACT

The aim of this work was to study electrostrictive behavior and vibration energy harvesting performance of conductive polyaniline (PANI)/polyurethane (PU) composites at low percolation threshold. Polyurethane composites with various PANI contents (0–2 wt%) were fabricated by solution casting. AFM, TGA and DSC techniques were used to investigate the topography, intermolecular interactions and dispersion effects in these composites. Moreover, dielectric and mechanical properties were determined to characterize electrostriction and energy conversion behaviors. The experimental results indicated that the PANI filler in PU matrix improved the electromechanical coupling, which is related to the interfacial charges between these phases. When an external electric field was applied to the electrodes, the generated current increased with the PANI content. Theoretical power analysis supported the experimental results, demonstrating that 2 wt% PANI in PU gave the maximum output power in a low electric field at 20 Hz mechanical frequency. Consequently, conductive PANI dispersed into electrostrictive PU can provide high power density and good efficiency of electromechanical conversion.

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1. Introduction

Electroactive polymers or EAPs are a new emerging category of materials. They include piezoelectric polymers and electrostrictive polymers. Piezoelectricity gives a linear relationship between the strain and the electric field [1], whereas electrostriction generally gives a quadratic dependence of the strain on the electric field [2]. In recent years, the EAPs have been popular in research on energy conversion, their benefits including high flexibility, low cost, and compatibility with any desired shape. A piezoelectric polymer has a high energy density created by a process called poling that leads to a polar phase. Typically, poling is performed under high electric field strength and at an elevated temperature, to facilitate domain alignment [3]. However, this fabrication process is difficult in practice. No such poling treatment is needed for electrostrictive polymers. Furthermore, these materials are conformable, lightweight, and durable [1,2]. The processing needed to fabricate these

polymers is simple and can be manipulated to control and optimize mechanical and electrical properties.

Among the various available electrostrictive polymers, polyurethanes (PUs) are of great interest for the large strains that electric fields induce in them [4]. Since electrostrictive polyurethane can achieve both large mechanical deformation and is electrically polarized, it can convert between electrical and mechanical energy forms, and can be used in actuators. A potential application in the reverse direction is energy harvesting, where this polymer could convert mechanical vibrations into electrical energy [5].

Several articles propose that the energy conversion capability of an electrostrictive polymer depends on its dielectric permittivity. An effective way to manipulate the dielectric permittivity is to add metal powders into the PU matrix [5–7]. Cottinet et al. [5] found that with the addition of 1% carbon nanopowder a PU matrix has the power density of 1 nW/cm³ under a 5 V/μm bias electric field, while the power density of a PU composite with 3% copper nanopowder is 1.78 nW/cm³ [7]. However, these composites have reduced flexibility and the fillers tend to form agglomerates. On the other hand, another electrostrictive polymer such as P(VDF-TrFE-CFE) has a very large power density (7200 nW/cm³) under a bias

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electric field of 5 V/ μm [8]. Unfortunately, this terpolymer has very high modulus, requires complicated preparation, and is also more expensive than an electrostrictive PU matrix.

Therefore, polymer composites with conductive polymer filler as the disperse phase are of great interest with potential to combine high conversion efficiency and good strain capability, and to avoid filler agglomeration effects [9]. In most conductive polymer composites in electrical engineering applications, polyaniline (PANI) has been used to obtain high permittivity due to its relatively high conductivity, ease of polymerization, environmental stability [9–11], and compatibility for dispersion into a variety of polymer matrices. Therefore, to improve the energy harvesting performance of the electrostrictive PU polymer, we chose conductive PANI as the dispersed polymeric filler.

In this work, we prepared PU/PANI composites with various PANI contents by solution casting. To maintain low mechanical modulus, these composites were filled only with low PANI contents (0–2 wt%), far away from the percolation threshold. The topography, intermolecular interactions and phase structure of each composite were assessed by using AFM, TGA and DSC techniques. The dielectric permittivity and Young's modulus were determined, and assessed as functions of filler content. The electrostrictive behavior that depends on the electric field and the figure of merit were assessed for all the composites. In addition, the induced current as a function of dc electric field was measured with various PANI contents. In particular, the energy harvesting potential of these electrostrictive composites was assessed and compared to prior published cases.

2. Materials and methods

2.1. Materials preparation

Thermoplastic polyurethane (PU, Estane 58888-NAT 021) was purchased from Lubrizol Corporation. The PU is based on MDI (4,4' methylene bis(phenyl isocyanate)), BDO (1,4-butanediol) as the hard segments (HS), and PTMO (poly(tetramethylene oxide)) as the soft segments (SS). The HS content is about 46 wt%. Long chain polyaniline (emeraldine salt) grafted to lignin was purchased from Sigma–Aldrich. The particle size of this PANI is about 2–3 μm . The 1-Methyl-2-pyrrolidone (NMP, 99% purity, M79603) was purchased from Sigma–Aldrich.

The composites were prepared as follows. Firstly, 20 wt% of dried PU granules were dissolved in an NMP solvent. The solution was heated to 80 °C for 45 min, to obtain a homogeneous solution. An ultrasonic probe (Hielscher UP400S) was used for 20 min to disperse the PANI filler into NMP solvent. Then the PANI solution was poured into the PU solution and stirred at 80 °C for 1 h by mechanical agitation. The obtained homogeneous mixture of PU and PANI was cast onto a glass plate, and cured at 60 °C for 24 h to remove volatile solvent residues. Finally, the film was peeled off the glass plate and annealed at 125 °C for 3 h. The thickness of these films was 80 \pm 5 μm .

2.2. Characterization methods

2.2.1. Surface topography

The topography, indicative of the micro-phase separation, was assessed by Atomic Force Microscopy (AFM, Nanosurf easyscan2, Switzerland), using Gwyddion software. N-type silicon probes (ACLC, Applied NanoStructures Inc., USA) with a manufacturer specified spring constant of 20–95 N/m were used in tapping mode. The resonance frequency of these 225 μm long cantilevers was within 145–230 kHz, and the tip curvature radius was less than 10 nm. The detector side of the silicon cantilever was coated with

aluminum. The sample was fixed on the sample stage with double-sided adhesive tape, over an about 25 mm² area, and the device was operated at room temperature.

2.2.2. Thermal analysis

The thermal stability of the composites was evaluated with the Thermal Gravimetric Analyzer (TGA, Perkin Elmer TGA7, USA), by monitoring weight changes that occur as a specimen is heated. Each approximately 5 mg sample was heated at a rate of 10 °C/min from 100 to 600 °C under nitrogen atmosphere.

A Differential Scanning Calorimeter (DSC, Perkin Elmer DSC7, USA) was used to determine the glass transition temperature (T_g) and the melting temperature (T_m) from endothermic peaks. Each sample (5–10 mg) was hermetically sealed in an aluminum crucible, and then the DSC cell was kept in an atmosphere equilibrated with liquid nitrogen. Each heating and cooling cycle was performed at a 10 °C/min scan rate covering the range from –150 to 180 °C.

2.2.3. Electrical properties

The dielectric constant (ϵ_r) and the electrical conductivity (σ) were evaluated over the frequency range 10⁰–10⁵ Hz by an LCR meter (IM 3533 HIOKI, Japan), used in frequency sweep testing mode at room temperature. A voltage of 1 V was applied to the sample, which was placed between two brass-plated electrodes (21.38 mm diameter) then the parallel capacitance and conductance readings were used to calculate the dielectric constant and the electrical conductivity, respectively.

2.2.4. Mechanical properties

The elastic modulus of each composite was evaluated in a strain gauge setup, with the help of a force gauge (BFG50N, Mecmesin, UK). The sample strips were 30 mm long, 5 mm wide, and approximately 80 μm thick, and were stretched in the length direction to 5% strain maximum at a 5 mm min^{–1} elongation rate. The fixed end of the sample was clamped to a force sensor, while the other end was clamped to a linear motor moving stage (RC ROBO-Cylinder Model RCP 2CR-SA6, 220 mm-long) that was controlled with a function generator. The elongation vs. force curves were determined for all samples and their Young's moduli (Y) were calculated.

2.2.5. Electrostrictive behavior

The electrostrictive behavior was assessed by measuring the electric field induced strain (S) with a photonic displacement apparatus (MTI-2100 Fotonic sensor, sensitivity 0.0058 mm/mV) at a low frequency and a low electric field strength ($f = 1$ Hz, $E \leq 6$ V/ μm). The sample was placed between two brass-plated electrodes (20 mm in diameter). The electric field (E) was applied to the sample in the thickness direction with the help of a high-voltage lock-in amplifier (Trek model 610E). The electric field induced strain was measured in the thickness direction.

2.2.6. Energy conversion ability

The energy conversion was evaluated from an energy harvesting setup illustrated in Fig. 1. Each rectangular shaped sample was sputter coated with gold electrodes (20 \times 40 mm²). The lower end of the sample strip was fixed to a loudspeaker, while the upper end was clamped to a rigid stand. The loudspeaker acted as a mechanical shaker providing sinusoidal 20 Hz excitation in the transverse sample direction. A laser displacement sensor (KEYENCE IA030, Japan) was employed to observe the displacement of the sample. A dc bias electric field was applied to the sample, originating from a function generator and amplified by a high-voltage amplifier, through the gold coatings on the sample surface. The polymer sample and the bias voltage source were connected

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