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Large electromechanical strain at low electric field of modified polyurethane composites for flexible actuators

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ABSTRACT

An electrostrictive polyurethane (PU) modified with three different polyaniline (PANI) loadings was demonstrated to overcome the limited actuated strains at low electric fields. The neat PU and all the composites were simply fabricated by solution casting. The experimental results show that high actuated strains (10.8%) at low electric fields (4.5 kV/mm) were observed for the modified PU/PANI-ES-HCl composite. In addition, the electrostrictive coefficient of the modified polymer increased by 20-fold compared to the neat PU. The modified PU/PANI-ES-HCl composites presented clearly a homogenous dispersion of filler in the polymeric matrix, and provided increased dielectric constant, high electromechanical response and low Young's modulus. Consequently, this PU/PANI-ES-HCl composite is potentially a useful flexible material for actuators in low electric fields.

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1. Introduction

Nowadays, electroactive polymers (EAPs) are presented as smart materials that have the ability to respond to external voltage stimuli and exhibit large deformations. EAPs, such as a dielectric elastomer, have the significant advantages of large electromechanical strain, high energy density, high coupling efficiency, fast response speed, light weight, low cost, and they are easy to produce even when large areas and modified shapes are required [1–4]. Among the most common electronic EAPs are electrostrictive polymers, which have been utilized in electromechanical applications of functional actuators, acoustics, vibration control and robotics [5,6]. Recently, electrostrictive polymers have shown capability to convert mechanical energy into electrical energy in energy harvesting applications [7,8]. However, the main drawback of electrostrictive polymers and dielectric elastomers is the very high electric field (> 100 kV/mm) required to produce large induced strains of the order 10% [9,10].

According to prior literature, increasing the electrostrictive coefficient (M_{33}) can provide high electromechanical responses. The M_{33} coefficient is proportional to the dielectric constant and inversely proportional to the Young's modulus of the material [11,12]. Therefore, in order to improve the capability of electromechanical response, we need to increase the dielectric constant and maintain low Young's modulus, which leads to a significant

reduction in the applied electric field. Among the various methods to increase the dielectric permittivity, the development of polymeric composites filled with conductive loading is most promising, providing an easy way to tune the electrical properties by choosing a suitable loading level. One of the most conductive polymers is polyaniline (PANI), which has the advantages of high conductivity, ease of polymerization, environmental stability, and compatibility for dispersion in a variety of polymer matrices [13,14].

In this letter, we present the electrostriction of polyurethane (PU) modified by charge injection through loading with three different kinds of additive PANI, i.e. PU filled with PANI emeraldine base is denoted as PU/PANI-EB, while that filled with emeraldine salt powder is denoted as PU/PANI-ES, and that filled with emeraldine salt using additional hydrochloric acid is denoted as PU/PANI-ES-HCl composite. Moreover, the influences of these additives on the dielectric and mechanical properties are also presented. Experimental results indicated that the modified PU/PANI-ES-HCl composite exhibits strongly enhanced electromechanical properties, when driven by a low electric field.

2. Experimental

2.1. Materials and preparation of composite films

Polyurethane Estane 58,888-NAT 021 was commercially obtained from Lubrizol Corporation. The PU matrix consists 46 wt% of

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hard segments based on MDI (4,4-methylene bis(phenyl isocyanate)), with BDO (1,4-butanediol) and PTMO (poly(tetramethylene oxide)) as the soft segments. Polyaniline emeraldine base (PANI-EB, 556459) used as a filler was supplied by Sigma-Aldrich. Firstly, dried PU granules were dissolved in 1-Methyl-2-pyrrolidone (M79603, Sigma Aldrich) at 20 wt% of PU, under continuous stirring at 80 °C for approximately 45 min to gain a homogeneous mixture. An ultrasonic probe (Hielscher UP400S) was used to disperse the PANI particles into NMP solvent for 20 min. Then the PANI solution was incorporated into the PU matrix with continuous stirring at 80 °C for 1 h, to obtain a homogeneous mixture. After that hydrochloric acid (HCl, fuming 37% acid) was poured into the PU/PANI mixture and stirred at room temperature for 10 min. The final mixture was cast onto a glass surface and cured at 60 °C for 24 h to remove most of the solvent. Then the dried film was peeled off from the glass plate and annealed at 125 °C for 3 h. The final thickness of these films was about $80 \pm 5 \mu\text{m}$.

2.2. Characterization

Scanning Electron Microscope (SEM, FEI Quanta 400) was applied to observe the shape and size distributions of the dispersed particle phase. Molecular level information and concentrations of the reaction groups in PU-PANI were observed by FTIR (Bruker EQUINOX 55) over the wavenumber region $4000\text{--}800 \text{ cm}^{-1}$. The dielectric constant was determined by using an LCR meter (IM 3533 HIOKI) in the frequency sweep mode. Elastic moduli of all the samples were evaluated from the slope at 5% strain in the stress-strain curve, obtained using a strain gauge apparatus (BFG50N, Mecmesin) at the strain rate of 5 mm min^{-1} at 25 °C. We measured the longitudinal strains at low frequency and low electric field (1 Hz, $E \leq 4.5 \text{ MV/m}$) with a photonic displacement sensor (MTI-2100). The film samples were placed on a horizontal brass disk, and a second brass disk was positioned on top of the film. Then the required bipolar voltage was applied with the help of a function generator (Siglent SDG1010) through a high-voltage supply (Trek model 610E). Finally, the longitudinal strain of the sample was calculated from the electric signal, which was measured using a lock-in amplifier (SR510 Stanford).

3. Results and discussion

SEM micrographs of the PU/PANI-EB and the PU/PANI-ES-HCl polymer composites are shown in Fig. 1. It is seen that the PANI

particles in the PU/PANI-ES-HCl composite appear randomly positioned, with good homogenous filler dispersion. However, the PU/PANI-EB shows microagglomerates of PANI (about $1 \mu\text{m}$).

FTIR spectra of pure PU and the composites with different kinds of PANI filler are shown in Fig. 2. For pure PU a (N–H) stretching peak at 3327 cm^{-1} , (C=O) stretching peak at 1728 cm^{-1} and (C–N) stretching peak at 1367 cm^{-1} are observed. However, the IR spectrum of the PU/PANI-ES-HCl polymer has the (N–H) stretching peak at 3321 cm^{-1} , (C=O) stretching peak at 1728 cm^{-1} and (C–N) stretching peak at 1363 cm^{-1} . When these two spectra are compared, major shifts are observed in the (N–H) stretching due to hydrogen bonding. The interaction between carbonyl groups (C=O) in PU and amine functional groups (N–H) in PANI component is a significant factor to examine in the following spectra. The strong absorption peaks around $3320\text{--}3330 \text{ cm}^{-1}$ are ascribed to the stretching vibrations of N–H group in the urethane segments. When adding PANI particles into the PU host, shift to higher frequency of the amine group peak was observed. Moreover, the C–N stretching vibration peaks, which are attributed to PU-PANI interactions [15], shift towards lower frequencies, except for PU/PANI-EB. It is known that stronger intermolecular forces lead to higher frequency of absorption. Therefore, the interactions between PU matrix and PANI-ES fillers obviously increase in PU/PANI-ES-HCl, affecting elastic modulus of this composite. Moreover, Jiang et al. proposed that the aromatic group and the hydrogen bonds are of high polarizability, which enhances the dielectric constant [16].

The effects of various fillers and frequencies on the dielectric constant of neat PU and the composites are presented in Fig. 3(a). For all the samples, the dielectric constant was found to decrease with the frequency. In addition, it is also noted that the dielectric constant increased with PANI loading regardless of the frequency. Especially at extra low frequencies, the composite of PU/PANI-ES-HCl evidently has a higher dielectric constant than the pure PU, or the composites PU/PANI-EB and PU/PANI-ES. The value of dielectric constant of PU/PANI-ES-HCl composites at 1 Hz increased to 142.6, which is about 18.3 and 6.2 times larger than those of pure PU and PU/PANI-ES composites, respectively. This can be explained by Maxwell-Wagner polarization of heterogeneous materials. This effect accounts for the dielectric property effects of the interfacial polarization between PU matrix and PANI filler. Several papers explained that the introduction of conductive filler increases the number of capacitors in the polymer composite, proportionally increasing the quantity of charge stored on either surface of the sample, and increasing the dielectric constant [17,18]. Our previous papers proposed that the gains in dielectric

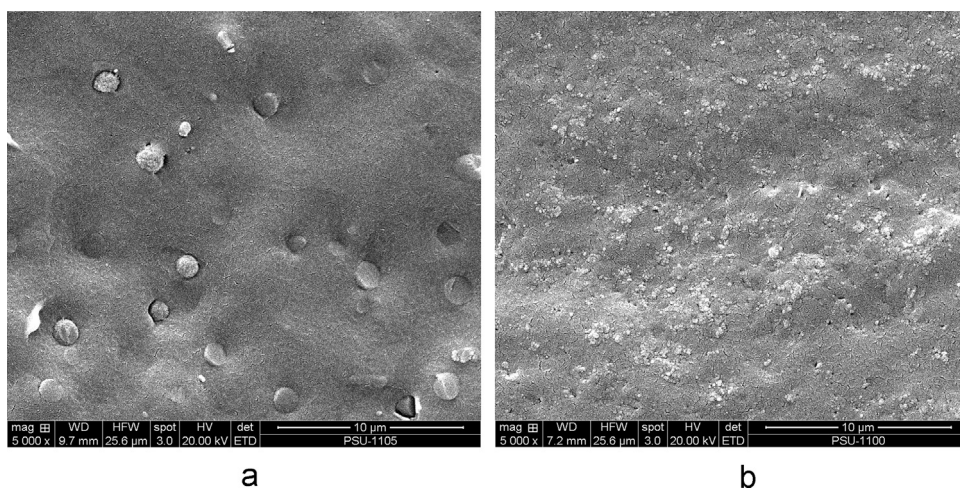


Fig. 1. Morphology of fractured surfaces of the PU composites with (a) PANI-EB and (b) PANI-ES-HCl loading.

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