



Constructing advanced dielectric elastomer based on copolymer of acrylate and polyurethane with large actuation strain at low electric field

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ABSTRACT

Dielectric elastomers (DEs) are kinds of advanced functional materials, attracting more attention due to that they can be used to fabricate ingenious devices. In this work, a series of advanced DEs based on the copolymer of polyurethane (PU) and acrylate, were chemically prepared through the condensation polymerization between p (BA-HEA), the *n*-butyl acrylate (BA) and hydroxyethyl acrylate (HEA) copolymer and diphenyl methane diisocyanate (MDI), named as p (BA-HEA)@MDI. The absence of urethane group was confirmed by infrared spectroscopy measurements. And the DEs show a decreased elastic modulus and an improved elongation at break with the decreasing content of MDI. For the absence of strong polar urethane group, the dielectric permittivity of new DEs still keeps at a relatively high value. And the p (BA-HEA)@MDI-3 shows the highest electromechanical sensitivity and the foreseeable highest actuation strain (14.4%) at a relatively low electric field (15.2 kV/mm) without any pre-strains, which is almost 2 times of that of VHB 4910 (acrylic adhesive tape, 3M Corporation). The present work provides a new strategy to design high performance dielectric elastomers.

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1. Introduction

Dielectric elastomers (DEs) are a type of electroactive polymers that can provide efficient electrical-mechanical energy transduction [1–8]. As dielectric elastomer, commercial low-cost materials such as acrylates (3M VHB), silicone rubbers, thermoplastic polyurethane (TPU), and natural rubbers have been widely investigated and demonstrated their actuation abilities [9–14]. However, the high electric field (>100 kV/mm) and high pre-strain needed to achieve satisfied actuation are limiting their commercial application [15–17]. An electric field of or below 20 kV/mm is considered as harmless electric field for biomedical devices and human-machine interfaces [8,15,18,19]. However, how to realize a large strain under a low driven electric field is still a great challenge in

developing ideal DEs to fully mimic the human muscles [20,21].

Many theoretical works flourished in order to predict the performance and understand the mechanism of actuation. From the standpoint of material design, literature shown that the strategy to improve the electromechanical actuation performance of DEAs should be enhancing the dielectric permittivity and decreasing the elastic modulus simultaneously while keeping a high breakdown strength [15,22]. As described, the actuated strain (S) can be expressed by Ref. [23]

$$S = \varepsilon_0 \varepsilon_r E^2 / Y \quad (1)$$

where ε_0 is the permittivity of free space, ε_r is the relative dielectric constant of the elastomer, E corresponds to the electric field and Y represents the Young's modulus. The actuation sensitivity is defined as:

$$\beta = \varepsilon_r / Y \quad (2)$$

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and it is often used as a figure of merit to describe a theoretical improvement for actuation abilities.

In this work, we prepared a series acrylate based DEs through the atom transfer radical polymerization (ATRP) method, named *p*(BA-HEA)@MDIs. The ATRP is one of the most frequently used controlled/living polymerization techniques, which allows the preparation of well controlled polymers of narrow molecular weight distribution (a very important parameter for the mechanical performance of polymers) and is easy to realize with very simple reaction equipment under gentle experimental conditions. While, it is rarely found the ATRP is used to prepare dielectric elastomers. In this study, the acrylate copolymer of *n*-butyl acrylate (BA) as soft monomer and 2-hydroxyethyl acrylate (HEA) as functional monomer was prepared via the ATRP method. Then new DEs were fabricated through the reaction between the prepared acrylate copolymer and various amount of diphenylmethane diisocyanate (MDI). The resulting DEs were investigated with respect to dielectric and mechanical properties, and displayed excellent actuated performance at low electric field as actuators.

2. Experimental

2.1. Materials

2-hydroxyethyl acrylate (HEA, 98%) was purchased from Aldrich and used after removal of the inhibitor. *N,N,N',N'',N''*-pentamethyldiethylenetriamine (PMDETA, 99%), Ethyl bromoisobutyrate (EBiB, 99%), and diphenyl methane diisocyanate (MDI, 98%), were purchased from Aldrich. Copper(I) bromide (CuBr, 99%), ascorbic acid (AA, 98%), *n*-butyl acrylate (BA, 98%), and dimethyl formamide (DMF) were purchased from Beijing Chemicals Company, Beijing, China.

2.2. Synthesis of *p*(BA-HEA) copolymers

The *p*(BA-HEA) copolymers were synthesized according to our previous work [24,25]. The EBiB (100 μ L), PMDETA (360 μ L), BA (70 g, 78.6 mL), and HEA (30 g 27.2 mL) were firstly added into a 250 mL flask containing 50 mL of DMF. The reaction system was then degassed by nitrogen atmosphere for 10 min before adding AA (300 mg) and CuBr (9 mg) under a nitrogen atmosphere. The polymerization was conducted for 24 h at 60 °C. Then the reaction was terminated by exposing to air. The reaction solution was diluted by THF and then precipitated in excess deionized water to remove the catalyst complex and reactant residues. Finally, the product was dried in a drying oven at 80 °C for 72 h. The final product is soft and viscous yellow solid.

2.3. Preparation of *p*(BA-HEA)@MDI elastomer films

The elastomer films were prepared through a solution casting method. In detail, the product and the MDI were thoroughly dissolved in DMF respectively, based on the predetermined molar ratio of OH groups from *p*(BA-HEA) macromolecule and NCO groups (4:1, 8:1 and 12:1). And then both solutions were mixed together before poured into a glassy dish and eliminating bubbles in vacuo for 10 min at room temperature. Then the glassy dishes were transferred into a drying oven to finish the solidification at 60 °C for 4 h and eliminate the solvent at 80 °C for 12 h. Finally, the films were peeled off carefully. The elastomer films were named as *p*(BA-HEA)@MDI-1, *p*(BA-HEA)@MDI-2 and *p*(BA-HEA)@MDI-3 respectively.

2.4. Characterization

¹H NMR spectra were measured on a Bruker ARX 400 MHz spectrometer using CDCl₃ as solvent and tetramethylsilane (Me₄Si) as internal standard. GPC measurements were performed on a Waters GPC system, in which THF was used as the eluent at a low flow rate of 1.0 mL/min at 30 °C and monodispersed polystyrene standards were used to generate the calibration curve. Infrared spectra of elastomer films were measured by using a Nicolet 5600 IR spectrometer in the 600–4000 cm⁻¹ wavenumber range.

To investigate the basic mechanical properties of the samples, the elastomer sample films were punched into rectangular shape samples with dimensions of 50 mm × 10 mm and then loaded into a tensile tester (Instron 3365) subjecting to a constant strain rate of 50 mm/min until rupture occurred at room temperature. All films were subjected to triplicate testing to ensure accuracy of the data. The dynamic mechanical properties were measured with a TA Q800 DMA. The sample films with around 260 μ m thickness were punched into rectangular shape samples with dimensions of 25 mm × 10 mm. Dynamic temperature ramp test was done at 5 °C/min, 1 Hz frequency and 1% strain.

Dielectric properties of the films were measured by using an impedance analyzer (Agilent 4294A) over the frequency from 100 Hz to 1 MHz. The homemade electrode made according to the previous literature [16], using silver-coated copper powder as conductors had been coated prior to measurement on specimens with rounded shape. The films were slightly compressed during the dielectric constant measurements to ensure good contact between the film and the electrodes. The electrical breakdown strength of the elastomers was investigated in a heat-transfer fluid bath at room temperature under a direct current field. Two spherical copper electrodes were pressed against the elastomer film from both sides. Then the voltage was applied by using a CS2674A high-voltage amplifier until electrical failure occurred.

Electroactuation performance was studied using a diaphragm actuator setup under prestrain-free experimental conditions. Two concentric circular homemade electrodes were coated onto opposite sides of the elastomer film while the film was held in a rigid test frame with two flat rings with a circular opening and connected to an outside voltage source via copper tape. After the actuation DC voltage was applied, due to synergistic effect of the limitation of the frame edge and the effect of gravity, the material was actuated downwards perpendicular to its surface and the displacements of elastomer films were monitored by a laser sensor (Panasonic HL-G1), which focused on the center of the actuator and connected to a personal computer. The area strain can be calculated from geometric relations based on the diameter and depth of the scallop.

3. Results and discussion

3.1. Synthesis and characterization of *p*(BA-HEA) and *p*(BA-HEA)@MDIs

The molecular weight and distribution of *p*(BA-HEA) were measured using GPC. In detail, the number-average molecular weight of *p*(BA-HEA) was 7.3×10^4 g/mol with polydispersity (PDI) value of 1.31. Furthermore, the composition of *p*(BA-HEA) was determined by the ¹H NMR spectra (see Fig. 1). By referencing the previous studies [25–27], the peaks at $\delta = (3.79–4.19)$ were assigned to the protons of -CH₂- from side chains of BA and HEA. The peaks at $\delta = (0.92–0.95)$ were assigned to the protons of methyl of BA. According to the average integrals of peaks at $\delta = (0.92–0.95)$ and $\delta = (3.79–4.19)$, the molar ratio of BA and HEA of *p*(BA-HEA) could be calculated. Then the weight ratios of *p*BA of *p*(BA-HEA) copolymers could be calculated according to the

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