



A new low moduli dielectric elastomer nano-structured composite with high permittivity exhibiting large actuation strain induced by low electric field

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ABSTRACT

Dielectric elastomers (DEs) are able to deform significantly in response under an external electric field. DE actuators have promising applications in many emerging fields such as biomimetic robots, haptic devices, tunable lens, and loudspeakers. Practical applications of DE devices are limited by the extremely high electric field and the high pre-stretch. Herein, we have designed and fabricated a new type of soft DE composites of partially reduced graphene oxide (RGO)/polystyrene-*b*-poly (n-butyl acrylate)-*b*-polystyrene triblock copolymer (SBAS). An extremely soft SBAS was tailor-made as the elastic matrix. A facile colloidal blending method, i.e. simply mixing the SBAS latex with GO aqueous dispersion, was used to fabricate the nano-structured composite with a segregated network of RGO nano-sheets. The design strategy led to the DE composite film of 1.5 wt% RGO/SBAS owning low modulus (0.51 MPa), high relative permittivity (~11) and relatively high dielectric breakdown strength (33 kV/mm). The DE composite film exhibited 21.3% maximum area actuation strain at relatively low electric field strength 33 kV/mm without pre-stretch.

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1. Introduction

Dielectric elastomers (DEs) are able to deform significantly in response to an external electric field in a reversible way. Thus, dielectric elastomer actuators (DEAs) can be used as an electro-mechanical transducer [1,2]. Compared with the traditional electro-mechanical transducer, DEAs are featured with softness, large actuation strain and low density. These features promise the application of DEAs in a wide range of emerging technology fields like biomimetic robots [3], haptic devices [4], tunable lens [5], lighter-than-air vehicles and loudspeakers [2].

Many elastomers have been evaluated as DEs including silicones, acrylics, and polyurethanes. A milestone study suggested that pre-stretching VHB 4910 acrylic elastomer and silicone rubber was able to dramatically increase the area actuation strain up to 158% and 93%, respectively [1]. However, the pre-stretched DEs

have the problems of stress relaxation and mechanical fatigue, which would significantly decrease the life time of a DE device. Also, the need for a rigid frame fastening DE films significantly limits the configuration of a pre-stretched DE device. So, it is desirable for new DEs showing high actuation strain without pre-stretch [6–8].

The actuation mechanism of DEAs has been well accepted. By applying a high electric field, the DEAs fabricated by a thin film of DEs sandwiched by two compliant electrodes would be deformed by the Maxwell stress, leading to a compression in the thickness direction and expansion in the planar direction. According to Pelrine et al. [1], the Maxwell stress exerting on the DE films can be expressed as

$$P = \epsilon_0 \epsilon E^2 \quad (1)$$

where P is the Maxwell stress along the thickness direction, ϵ_0 is the permittivity of free space (8.85×10^{-12} F/m), ϵ is the relative permittivity of a DE, and E is the electric field (V/m). At sufficiently small strains (<20%), the thickness strain S_z caused by the Maxwell stress can be formulated as [1,9].

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$$S_z = -P/Y = -\epsilon_0 \epsilon E^2 / Y \quad (2)$$

where Y is the elastic modulus. According to Eq. (2), it is clear that low modulus, high electric field, and high permittivity could give an elastomer large actuation strain.

Since the relative permittivity of typical DEs is low (2.8–6) [2], extensive research efforts have focused on the preparation of DE composites to significantly increase the permittivity and thus decrease the actuating voltage. The high permittivity inorganic fillers such as TiO_2 [10], BaTiO_3 [11], lead magnesium niobate–lead titanate (PMN–PT) [12] and conductive fillers such as carbon nanotubes (CNTs) [13], graphene sheets [14], and partially thermally reduced graphene oxide (RGO) [15] have been used as functional fillers to increase the permittivity. However, those DE composites with increased permittivity often suffered from much increased elastic modulus, higher dielectric loss, and decreased dielectric breakdown strength. As a result, the maximum actuation strain was only increased a little and even decreased in certain situations [16,17]. Simultaneously decreasing the modulus and increasing the permittivity of DEs by adding polar solvent or plasticizer has been shown to be an effective way to improve the actuation strains [18,19]. However, decreasing modulus often led to decrease in mechanical strength. The solvent loss and leakage under stress would bring about the problems of long-term performance stability [8,19].

In the current contribution, we fabricate a novel soft DE nano-structured composite with high permittivity and high dielectric breakdown strength, exhibiting high actuation strain without pre-stretch at relatively low electric field. We design and synthesize the elastomer latex of polystyrene-*b*-poly (n-butyl acrylate)-*b*-polystyrene (SBAS) triblock copolymer. The SBAS latex could not be accessed from a conventional emulsion polymerization. The recent progress in reversible addition/fragment transfer (RAFT) emulsion polymerization [20] allows us to tailor-make SBAS copolymers with high elasticity and low modulus. The micro-phase separation of the triblock copolymer would lead to the formation of polystyrene (PSt) nano-domains well-dispersed in poly(n-butyl acrylate) (PnBA) matrix [21]. The rigid PSt domains act as physical crosslinking points, leading to well-defined crosslinked structures to prevent the chains from slipping under stress. The block of PnBA gives the copolymer a relatively high permittivity due to the existence of polar ester groups. At the same time, the bulky side groups of butyl ester make PnBA matrix have a low entanglement density. So, SBAS could be a very soft elastomer when the copolymer composition of PSt was designed to be low enough [20,22]. Then, the graphene oxide (GO)/SBAS composite films are fabricated by simply drying the mixture of the as-prepared SBAS latex and the GO aqueous dispersion at room temperature. It is expected that the GO sheets would uniformly cover the surface of SBAS particles due to the strong π - π stacking interactions of the multiple aromatic regions between GO and PSt during film formation [17,23]. It is also expected that a segregated network of the GO nano-sheets would form at a low percolation threshold, leading to a nano-structured composite with low modulus. Finally, the in situ thermal reduction of the GO/SBAS composites is conducted to further increase the permittivity of the RGO/SBAS composite film. It turns out that we can achieve a RGO/SBAS composite with low modulus, high relative permittivity, and high dielectric breakdown strength. It is further demonstrated that a composite film without pre-stretch could exhibit 21.3% area actuation strains driven by a relatively low electric field.

2. Experimental

2.1. Materials

The GO aqueous dispersion (10 mg/mL) was supplied by Tan Gu Shang Xi Material Technology Co., LTD (China). Styrene (St) and acrylic acid (AA) were purified by vacuum distillation. n-Butyl acrylate (nBA) was washed with sodium hydroxide aqueous solution (10 wt%) to remove the inhibitor. Potassium persulfate (KPS, >99%), hydrochloric acid (HCl, 37 wt%), and sodium hydroxide (NaOH, >96%) were used without further purification. The oligo (acrylic acid-*b*-styrene) trithiocarbonate macroRAFT agent (AA₂₀-*b*-St₅-RAFT agent) was synthesized according to the procedure described elsewhere [24]. ¹H NMR spectrum revealed that a macroRAFT agent molecule was composed of 20 AA and 5 St units. GPC suggested that the relative molecular weight of AA₂₀-*b*-St₅-RAFT agent was 2309 g/mol.

2.2. Synthesis of SBAS copolymer latex via RAFT emulsion polymerization

SBAS was synthesized in RAFT emulsion polymerization by sequential adding styrene, n-butyl acrylate, and styrene, according to the procedure previously described [20]. The detailed procedure can be found in the Supporting Information.

2.3. Fabrication of SBAS composite films

First, a given amount of the GO aqueous dispersions was diluted by deionized water and then subjected to ultrasonication for 2 h, obtaining a uniform GO dispersion. Then, the uniform GO dispersion was mixed with the as-prepared SBAS latex under stirring for 24 h. Finally, the mixture dispersion of GO/SBAS was casted in a polytetrafluoroethylene dish and dried at 25 °C. The resulted GO/SBAS composite film was then further dried at 75 °C for 24 h under vacuum. The thermal reduction of the GO/SBAS composite film was conducted at 180 °C for 4 h under normal pressure to obtain RGO/SBAS composite film.

2.4. Characterization

The molecular weight and PDI (Mw/Mn) were measured by GPC Waters 1525/2414 (Waters, USA). Monomer conversion was measured by gravimetry. The composition of SBAS was determined by ¹H NMR using tetrahydrofuran-*d*₈ as a solvent on a DMX 500 MHz spectrometer (Bruker, Germany). TGA were conducted on a TA Q500 (TA Instruments, USA) under N₂ gas atmosphere. XPS experiments were conducted on an Escalab250Xi X-ray photoelectron spectrometer (Thermo Fisher Scientific Company, USA) to analyze the chemical compositions of GO and RGO. The particle sizes and zeta potential of particles and GO dispersion were measured by a ZETASIZER Nano series (Malvern Instruments Limited, UK) at 25 °C. UV–vis absorption spectra were conducted on a UV-2450 spectrophotometer (Shimadzu, Japan). The morphology of the SBAS composites was characterized by transmission electron microscopy on a JEM1200-EX (JEOL, Japan).

The mechanical properties of pure SBAS and the RGO/SBAS composites were measured by a Zwick/Roell Z020 (Zwick, Germany) universal material tester with a loading rate of 20 mm/min at 20 °C. The testing samples were cut into the dumbbell shape (35 mm × 7 mm × 0.3 mm). For each sample, five specimens were tested.

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