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Influence of fiber characteristics on directed electroactuation of anisotropic dielectric electroactive polymers with tunability

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ABSTRACT

Dielectric elastomers constitute a technologically important class of stimuli-responsive polymers due primarily to their unique ability to achieve large strains (>300 area%) upon exposure to an external electric field. In most reported cases, actuation strains are measured as dielectric elastomers constrained to a circular test configuration essentially waste energy by undergoing isotropic, rather than directional, electroactuation. Recent independent studies have demonstrated, however, that the addition of relatively stiff fibers to a soft dielectric elastomer matrix promotes more energy-efficient anisotropic mechanical behavior and electroactuation response. In this work, we investigate the effects of fiber strain and mechanical properties on electroactuation in anisotropic dielectric electroactive polymers with tunability (ADEPT) fabricated from an acrylic dielectric elastomer. Increases in fiber loading level and stiffness are observed to enhance both mechanical and electroactuation properties to different extents, and we introduce an electroactuation anisotropic enhancement factor to quantify the ratio of electroactuation to mechanical anisotropy. This factor is determined to vary linearly with fiber concentration for nearly all the different ADEPT composites examined in this study.

1. Introduction

As the search for stimuli-responsive materials intensifies to meet the growing demands of microrobotic, microfluidic and haptic devices, fundamental and technological interest in electroactive polymers continues to increase [1–8]. Electroactive polymers (EaPs), macromolecules that change size and/or shape upon exposure to an applied electric potential or electric field, are divided into ionic and electronic classifications. Only those belonging to the latter category undergo electroactuation due to the presence of an external electric field and are relevant to the present study. Examples of such EaPs include ferroelectric polymers, such as poly(vinylidene fluoride) and its copolymers [9–11], which experience a change in lattice dimensions upon electroactuation, and dielectric elastomers (D-EaPs) [12,13], which come closest to emulating the electromechanical behavior of mammalian skeletal muscle [14–16]. In general, D-EaPs are inexpensive and scalable soft materials that primarily deform due to field-induced charge separation, which generates a compressive (Maxwell) normal stress as a result of electrostatic attraction. Since electroactuation of D-EaPs is isochoric, compression of the homogeneous elastomer in the transverse (z) direction is accompanied by isotropic expansion in the lateral (x-y) plane (*cf.* Fig. 1). This expansion is frequently augmented by repulsive charge buildup along each of the electrodes. For an ideal D-EaP, the Maxwell stress (σ_z) is given by $\varepsilon\varepsilon_0(\Phi/d)^2$, where ε is the dielectric constant of the elastomer, ε_0 is the permittivity of free space, Φ denotes the electric potential and d is the elastomer thickness (so that Φ/d yields the magnitude of the electric field) [1,17]. A wide range of different covalently-crosslinked elastomers varying in chemistry from natural and fluorinated rubber to silicones and acrylics have been considered as D-EaPs [12,13]. Physically-crosslinked systems composed of selectively-swollen thermoplastic elastomers [12,14–16,18] have also been successfully employed for this purpose.

While numerous studies continue to explore emerging technologies for D-EaPs [8], others have sought to improve the electroactuation efficacy of various D-EaPs. One of the most common ways in which this can be achieved is through the use of uniaxial or

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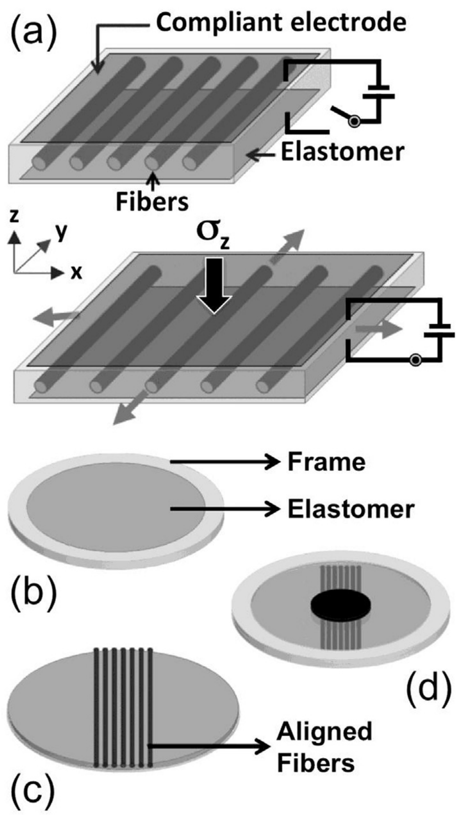

Fig. 1. In (a), schematic illustration of the ADEPT design paradigm wherein oriented HDC fibers are incorporated into the matrix of an elastomer that undergoes electroactuation due to the presence of compliant electrodes when subjected to an external electric field. Here, σ_z denotes the normal Maxwell stress that compresses the elastomer in the *z*-direction and expands the elastomer in the lateral *x*- and *y*-directions. In (b-d), the fabrication process is depicted, starting with the production of equibiaxially prestrained elastomer films (b) without and (c) with fibers that could be uniaxially strained prior to assembly. The composite displaying the active area for electroactuation is portrayed in (d).

biaxial mechanical prestrain, which serves two purposes. Prestrain reduces d and permits use of lower Φ to induce electroactuation at a material-specific electric field. It also increases molecular stretching and thus promotes an increase in the electromechanical modulus [19] and a corresponding change in mechanical stress-strain behavior [20]. A significant drawback of this strategy is the need for a cumbersome and rigid frame to maintain retention of prestrain during application. Addition of a prestrain frame furthermore compromises the desirable lightweight attribute of D-EaPs, and the level of induced prestrain can degrade over time due to stress relaxation of the D-EaP. Pei and co-workers [21,22] have overcome this disadvantage by incorporating a second network-forming polymer into a mechanically prestrained acrylic elastomer and then removing the prestrain frame. By retaining its original prestrain, the resultant bi-network D-EaP can exhibit actuation strains in excess of 400 area%. More recently, Vatankhah-Varnosfaderani et al. [20] report that bottlebrush silicone elastomers, consisting of chemically-crosslinked chains possessing side chains that can be independently varied in length and density, are molecularly prestrained and can attain actuation strains up to 360 area% at $\Phi/d = 13$ kV/mm without mechanical prestrain. In all of these instances, electroactuation conducted within the confines of a circular test configuration or an expandable diaphragm remains isotropic, in which case energy is expended in all directions normal to σ_z. For many potential applications, however, D-EaP electroactuation should be conducted along a particular direction to ensure efficiency at low overhead.

Directional, or anisotropic, electroactuation can be realized through the precise arrangement of fibers within a D-EaP matrix prior to electrical stimulation. Bolzmacher et al. [23] have found that polyamide (PA) fibers incorporated into a silicone D-EaP behave as trusses that can hold mechanical prestrain and generate as much as 35% linear strain. Similarly unidirectional constraint of D-EaPs with either PA or carbon fibers oriented in the orthogonal direction is responsible for 25% or 28% linear actuation strain, respectively, at the onset of dielectric breakdown [24]. In comparable cylindrical actuators, the maximum actuation strain recorded [25] is 36%. We have previously demonstrated [26] that the addition of unstrained, high-dielectric-constant (HDC) polyurethane (PU) fibers to an acrylic elastomer equibiaxially prestrained to 200% not only increases ε of the composite but also promotes composition-dependent anisotropic electroactuation along the fiber normal at substantially elevated elecromechanical efficiency. Results previously obtained from our anisotropic dielectric electroactive polymers with tunability (ADEPT) design clearly indicate that judicious selection of a relatively stiff HDC additive capable of introducing electromechanical anisotropy yields D-EaP composites that are economically viable and that exhibit tremendous promise for electroactuation that is both directional and composition-tailorable. The overarching objective of the present study is to discern if the performance of ADEPT composites can be further and systematically improved by modifying either fabrication or composition. With this intention in mind, we explore the use of an equibiaxially prestrained acrylic elastomer containing PU fibers subjected to different levels of uniaxial strain prior to incorporation, as well as PA and silicon carbide (SiC) fibers, to establish the roles of fiber stiffness and/or ε on the extent of anisotropic electroactuation.

2. Experimental

The acrylic elastomer investigated in this study was a commercial adhesive (VHB-4905; 3M Co. Minneapolis, MN, USA) commonly employed as a D-EaP (its properties are detailed elsewhere [1,12,13]), and the PU fibers (RadiciSpandex Corp. Gastonia, NC, USA) were multifilament dry-spun fibers composed of 3–8 filaments, each measuring 40-50 μm in diameter. The elastic moduli of these fibers varied with strain: 9-10 MPa at 0%, 15-20 MPa at 100% and 30-54 MPa at 200%. A PA fiber measuring 76 μm in diameter with an elastic modulus of ~2 GPa was obtained from Jarden Applied Materials (Columbia, SC, USA), whereas a ceramic (Nicalon™ SiC) fiber measuring 14 μm in diameter with an elastic modulus of ~270 GPa was supplied by COI Ceramics, Inc. (Magna, UT, USA). As depicted in Fig. 1, each ADEPT composite was prepared by first equibiaxially prestraining two elastomer films to 200% and then fixing each on a circular rigid frame. Equally spaced fibers were uniaxially aligned on one of the films, while the other film was carefully brought in contact with the fiber-containing film to generate a laminate, which was subsequently placed under vacuum at ambient temperature for 12 h to ensure maximum interfacial consolidation. Dielectric properties of the resulting composites at 25 °C were measured along the fiber normal on specimens placed between two parallel-plate electrodes in an Instek LCR-8101G meter operated at frequencies ranging from 10^{-1}–10^{3} kHz. Orthogonal tensile moduli of the composites were measured from uniaxial tensile tests conducted on a MTS-30G load frame operated at a constant crosshead speed of 4.4 mm/min. The actuation response of each ADEPT composite was evaluated in a routine circular test configuration. An active area (*cf.* Fig. 1) was fabricated at the center of each test specimen through the application of