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# **Organic Electronics**

journal homepage: www.elsevier.com/locate/orgel

# Improving electroactive polymer actuator by tuning ionic liquid concentration



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#### ARTICLE INFO

Article history: Received 2 October 2013 Received in revised form 9 November 2013 Accepted 12 November 2013 Available online 26 November 2013

*Keywords:* Polymer actuator Ionic liquid Actuator fabrication

# ABSTRACT

We have fabricated actuators from a blend of fluoropolymer (FP) with ionic liquid (IL). Here a combination of graphene, graphite, and silver nanoparticles is used to raise the electrode conductivity. As the electrode composition is fixed, we found that the actuator displacement increases with decreasing amount of ionic liquid in the polymer gel electrolyte. A maximum strain of 0.48% was observed from peak-to-peak displacement for an actuator with IL/FP = 0.3 in the polymer gel electrolyte. The simulation results indicate that lowering IL concentration leads to a more compact ion distribution in the electrode layers and hence explains the increased strain in the actuators.

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## 1. Introduction

Electroactive polymers have enabled soft, lightweight actuators, sensors, and energy harvesting devices that are widely applicable to robotics, haptics, and biomimetic systems [1–3]. Among the various types of electroactive materials, fluoropolymers blended with ionic liquid (IL) allow device operation in air at low applied voltages. The polymer blends can be deposited over large area, patterned into arbitrary shapes through solution printing [4,5], and integrated into flexible organic electronics [6–8]. Here we use solution casting to change the IL concentration and enhance the performance of a bimorph actuator.

A bimorph actuator is comprised of a polymer gel electrolyte layer sandwiched between two electrodes, in which IL is incorporated in all three layers. Upon applying a bias, the IL cations and anions are redistributed by the electric field, and the ion migration leads to volume change in the structure because of steric repulsion and electrostatic effects between the charged ions [9,10]. Due to the size difference between cations and anions, one of the electrodes shrinks while the other one swells, resulting in a bending motion. This type of ionic gel actuator is based on the bucky gel actuators pioneered by Asaka et al. They have shown that the generated strain depends on the size and transport properties of the IL species [11,12], as well as on the fluoropolymer support [13], and the composition of the electrodes [14-16]. The interaction between electrode, polymer matrix, and electrolyte is key to the actuator's final performance. The electrodes are typically made from integrating conductive carbon nanoparticles and/or nanotubes into the fluoropolymer-IL blend. Structures with graphene hybrids [17,18] and graphene-stabilized silver electrodes [19] have shown improved actuator frequency response and displacement. Here a combination of graphene, graphite, and silver nanoparticles is used to raise electrode conductivity and is shown to be another viable choice for electrode materials. As for the polymer gel electrolyte film, previous studies have often treated it as a mere transport layer. However, the role of the polymer gel electrolyte layer extends beyond ions transport, and this paper investigates how the electrolyte composition affects the actuation mechanics in bimorph structures.

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<sup>&</sup>lt;sup>1</sup> Experiments were completed at Palo Alto Research Center, when this author was there on sabbatical.

<sup>1566-1199/\$ -</sup> see front matter @ 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.orgel.2013.11.026

In this work, we show that actuation strain is improved by tuning the IL concentration in the polymer gel electrolyte. The ratio of IL to fluoropolymer (IL/FP) is varied in the polymer gel electrolyte layer, as the electrode composition remains fixed. The actuator properties are measured and examined alongside simulation results. We demonstrate that adjustment of the polymer gel electrolyte composition provides a new method to increase actuator displacement.

### 2. Experiments

#### 2.1. Materials and fabrication procedure

All of the materials were used as received. The ionic liquid was 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIBF<sub>4</sub>, Aldrich). The fluoropolymer was fluoride-cohexafluoropropylene (PVDF-HFP, Kynar Flex 2801, Arkema). The electrode materials were graphite (Aldrich), graphene composite (Vorbeck F101), and silver nanoparticle ink (Sun Chemicals). *N*,*N*-dimethylacetamide (DMAc), 4methyl-2-pentanone (MP), and propylene carbonate (PC) were purchased from Aldrich and used as solvents.

Six different weight ratios of EMIBF<sub>4</sub>:PVDF-HFP were prepared for the polymer gel electrolyte layers, ranging from 0.3, 0.4, 0.6, 0.8, 1, to 1.5. The ratio of 1.5 was the maximum limit, because beyond that the ionic liquid started to form a separate band in the solution mixture. Each solution had 200 mg PVDF-HFP, 60–300 mg EMIBF<sub>4</sub>, dissolved in 6 mL MP and 0.5 g PC solvents. The solution was stirred for one day at 70 °C, and then 2 g was cast onto a circular mold with 25 mm radius. The mixture was dried in air at 50 °C for one day and in vacuum oven at 80 °C for three days. The resulting polymer gel electrolyte films were measured to be 40  $\mu$ m in thickness.

The electrode layers had a fixed composition made from a solution of 100 mg graphite, 200 mg EMIBF<sub>4</sub>, 100 mg PVDF-HFP, mixed in 10 mL DMAc solvent. The solution was stirred for one day, and then 1.5 g was cast onto a circular mold with 25 mm radius. The mixture was dried in air at 50 °C for one day and in vacuum oven at 80 °C for three days. The electrode films were measured to be 40 µm in thickness. The surface resistance of these films was 200  $\Omega/\Box$ , and the graphite films were made more conductive (150  $\Omega/\Box$ ) by spin-coating a micron-thick layer of the graphene composite ink. This composite conductor also acted as a blocking barrier to seal in the IL. A silver ink was spin-coated upon the blocking layer, and the silver film (50 nm) drastically reduced the electrode resistance to  $20 \Omega/\Box$ . We observed that without the silver film, the resistivity of the electrode was too high and the actuator device did not show movement. In general, resistivity of tens of ohms is required for effective actuation. While future experiments are needed to quantify the lifetime of actuators using non-noble metals, this method of using a blocking layer expands the choices of conductor materials, to allow low-cost metals in devices with ionic liquid.

The electrode films were cut into 20 mm by 5 mm strips. They were laminated [20] with the polymer gel electrolyte films to form actuator structures as illustrated in



**Fig. 1.** (a) Schematics of the actuator. (b) Chemical structures of the ionic liquid EMIBF<sub>4</sub> and the fluoropolymer PVDF-HFP.

Fig. 1. The lamination was done at 2 atm and 70 °C for one minute. It was observed that for films with EMIBF<sub>4</sub>:PVDF-HFP (IL/FP) ratio below 0.3, the adhesion was poor between the polymer gel electrolyte and electrodes. Thus, IL/FP = 0.3 was the minimum composition ratio. The total thickness of a typical device was around 120  $\mu$ m.

#### 2.2. Measurement procedure

Electrical and mechanical measurements were simultaneously captured. The electrical contacts to the bimorph actuator, as shown in Fig. 2 inset, were made from aluminum foils soldered to wires, and the contacts were located on the two isolated sides of a spring-loaded clip, which clamped down on the actuator electrodes. As voltage was applied to one electrode, the other electrode was connected to an electrometer (HP model 617) to monitor current through the actuator. The actuator motion was concurrently recorded by a video camera at 60 frames per second to measure displacement. The video sequence



**Fig. 2.** (a) Displacement and (b) current of actuators under  $\pm$ 4 V square waveform at 0.05 Hz. The amount of ionic liquid in the polymer gel electrolyte is at 0.3× or at 1× of polymer weight. (c) Photos of the bending actuators at 4 V.

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