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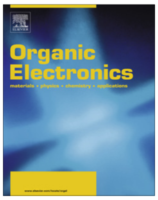

# Influence of ionic liquid concentration on the electromechanical performance of ionic electroactive polymer actuators

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ABSTRACT

We have investigated influence of ionic liquid concentration on the electromechanical response of ionic electroactive polymer actuators. Actuators were fabricated from ionomeric membrane and doped with different concentrations of 1-ethyl-3-methylimidazolium trifluoromethanesulfonate ionic liquid. Samples were investigated for their electromechanical and electrochemical characteristics; and it was observed that the maximum electromechanical strain of approximately 1.4% is achieved at 22 wt% ionic liquid content. Increasing ionic liquid concentration results in saturation of the electrode–ionomer interface and formation of ionic double/multi layers, which in turn result an inward accumulation of ions; hence, generate strain in an undesired direction that deteriorates the electromechanical response of the actuator.

## 1. Introduction

Recently, electroactive polymers have received immense attention and interest from the materials community because of their applicability to actuators, sensors and haptics [1–3]. Electroactive polymers are soft and lightweight; hence, enable realization of biomimetic and microrobotic devices. Among wide variety of electroactive polymers, ionic electroactive polymers (IEAP) have proven more practical for actuator applications due to their substantially low operation voltage (typically <5 V), light weight, relatively large strain, and bending (instead of linear) deformation [4,5].

IEAP actuators comprise of an ionomer membrane that is doped with an ion-rich electrolyte and coated with electrodes on each surface [6,7]. The electromechanical response is upon attraction/repulsion of ions and their accumulation at the oppositely charged electrode when subjected to an external electric field. Due to the volume difference between cations and anions, cathode and anode swell to different extents, thus a volume imbalance is generated in the actuator, which in turn causes a mechanical deformation. Change in the polarity of the electric field reverse the process and direction of bending [8–14].

Ions are sourced by either an aqueous electrolyte or ionic liquid (IL). Ionic liquids are preferred as their near zero vapor pressure allows longer shelf life, operation in air, and higher operation voltages without concerns about ionomer hydration or electrolysis of water in aqueous electrolytes [15–17]. Also, substantially higher ion concentration in ionic liquids, compare to that of aqueous electrolytes, and larger Van der Waals volume difference between molecular cations and anions (compare to atomic cations and anions in aqueous electrolytes) result in an enhanced performance of IEAP actuators doped with ionic liquids, compare to those doped with aqueous electrolytes. These characteristics along with scalable manufacturing and flexibility in design allow integration of IEAP actuators in

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flexible organic electronics, microrobotics, biomimetic devices and bioelectronics [18].

In the present study, we have demonstrated that the electromechanical performance of IEAP actuators is influenced by the concentration of ionic liquid, and that the concentration of ionic liquids can be tuned to achieve maximum actuation performance. Ionic liquids concentration in IEAP actuators was varied while electromechanical and electrochemical properties were characterized. It is shown that the ionic liquid concentration can be used as a means to control, improve and optimize actuation performance; and that at high concentrations of ionic liquid an ionic double/multi-layer forms at the ionomer–electrode interface which deteriorate the actuation strain.

## 2. Materials and methods

### 2.1. Sample preparation

Commercially available Nafion membrane of 25 μm thickness (Ion Power, Inc.) was used as the ionomeric membrane. To fabricate IPMCs, nanocomposites of the polycation poly(allylamine hydrochloride) (PAH) (Sigma Aldrich) and anionic functionalized gold nanoparticle (AuNP) (~3 nm diameter, Purest Colloids, Inc.) were grown on both sides of the Nafion membrane via LbL deposition of the ionic species, using a StratoSequence 6 (NanoStrata, Inc.) automated thin-film fabrication robot. The substrates were alternately immersed for 5 min each in aqueous solutions of PAH at a concentration of 10 mM at pH 4.0 and AuNP at a concentration of 20 ppm at pH 9.0 with three rinsing steps for 1 min each in de-ionized water after each deposition step. IPMCs were then soaked with 1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMI-Tf molecular formula: $C_7H_{11}F_3N_2O_3S$) (Sigma Aldrich) ionic liquid at 80 °C for various durations of time to intake desired concentrations of ionic liquid. Ionic liquid content was measured as the weigh percentage (wt%) of dry weight of the membrane, and calculated from Eq. (1).

$$W_e\ (\%) = \frac{W_f - W_d}{W_f} \times 100 \qquad (1)$$

where $W_e$ (%) is the weight-percent of the electrolyte; and, $W_d$ and $W_f$ are the weights of dry and doped samples, respectively. Gold leaf electrodes of 50 nm thickness were then hot-pressed at 95 °C under 1000 $lb_f$ for 25 s on both sides of the membrane to form IEAP actuators.

### 2.2. Electrochemical characterization

Impedance spectroscopy and current flow were measured and recorded using a VersaSTAT-4 potentiostat (Princeton Applied Research). The impedance spectroscopy studies were carried at frequencies between 1.0E5 Hz and 0.1 Hz, and a potential difference ($\Delta V$) of 10 mV. Current flow was monitored in response to a ±4 V step potential over 60 s intervals. Electrical conductivity ($\sigma$) of the doped membranes was calculated from Eq. (2),

$$\sigma = \frac{h}{RA} \qquad (2)$$

based on the geometry of the membranes where $h$ and $A$ represent thickness and area of the membrane, respectively; and $R$ is the resistance deduced from impedance spectroscopy measurements.

### 2.3. Electromechanical characterization

Actuators were cut into approximately 1.5 × 15 $mm^2$ pieces and tested under application of a 4 V step potential. Electromechanical response of the actuators was monitored and recorded using a charge-coupled device (CCD) video camera, mounted to an in-house fabricated micro-probe station, at 30 fps. Individual frames were then analyzed to measure the radius of curvature as a function of time ($r(t)$) and to calculate ($Q(t)$) and strain ($\varepsilon\%(t)$) values from Eqs. (3) and (4), respectively; where $Q$, $\varepsilon$ and $h$ are curvature, strain and thickness of the actuator, respectively.

$$Q(t) = \frac{1}{r(t)} \qquad (3)$$

$$\varepsilon\%(t) = \frac{h}{2r(t)} \times 100 \qquad (4)$$

## 3. Results and discussions

### 3.1. Current flow

Current flow corresponding to a 4 V potential difference between the outer electrodes was measured and recorded as a function of time. As presented in Fig. 1, magnitude of displaced charge (area under the curve) increases with the increasing concentration of ionic liquid in the samples; suggesting that first, current flow is due to mobilized ions; and second, more ions are displaced in samples containing higher concentration of ionic liquid. After approximately 55 s (see 55 < $t$ < 60 and 115 < $t$ < 120 on Fig. 1) all curves have asymptotically reach the $x$-axis (approximately zero current) indicating that the system is fully charged.

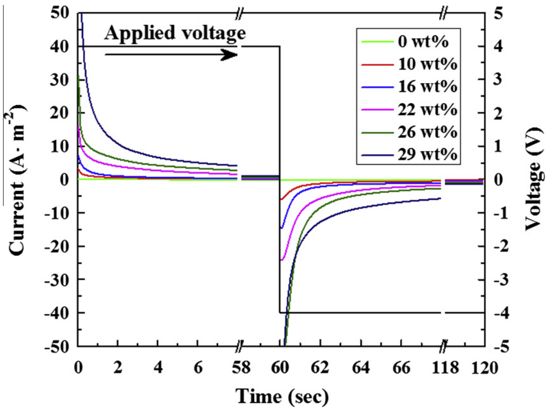

**Fig. 1.** Charging and discharging currents for samples containing different ionic liquid concentrations recorded as a function of time under a 4 V square wave.