Longitudinal size effects on electrical and actuation behaviors of ionic polymer-metal composite

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Abstract
The structural characteristics of ionic polymer-metal composite (IPMC) were analyzed, and the effects of IPMC length ranging from 1 to 5 cm (with a width of 0.5 cm) were evaluated in terms of electrical parameters and actuation behavior. It has been concluded that the electrical parameters of IPMC materials (including capacitance, electrode resistance in thickness-direction, and internal resistance) decrease, and the bending strain of the setpoint increases as the length increases. The simulation of the current response of IPMC to 2 V DC voltage shows that the error between simulated peak current using 1 cm-IPMC parameters and the measured value is 8.23 times higher than that of 5 cm-IPMC. The strain of the setpoint on 5 cm-IPMC sample is 5.65 times bigger than that of the 1 cm-IPMC sample.

Keywords
IPMC, electrical parameter, length effect, simulation, actuation behavior

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Introduction
Ionic polymer-metal composite (IPMC) is one of the most promising soft smart materials for electromechanical or mechanoelectrical transduction, because of its lightweight, flexibility, noiselessness, structural simplicity, fast response, and a large deformation under a low voltage.¹⁻⁴ The attractive inherent advantages of IPMC make it a wide range of medical and industrial applications in artificial muscles and soft robotic actuators as well as dynamic sensors.⁵⁻⁸ The typical structure of IPMC is composed of one ionically conductive electrolyte membrane (e.g. Nafion, Flemion, or Aciplex) plated with a metal (e.g. Platinum, Silver, or Copper) electrode on both sides. When an excitation voltage is applied to the electrodes, the uniformly distributed hydrated cations in the membrane are forced to move toward the cathode, resulting in the IPMC bending toward the anode side.⁹,¹⁰

The behavior of IPMC materials exhibits coupling among electrical, chemical, and mechanical properties. Since the late 20th century, a lot of studies have been focused on fabrication, modeling for transduction behaviors, and complicated application of IPMC materials.⁵,¹¹ However, only a few authors studied on the effect of the size on the response of IPMC materials. Li and Yip¹² investigated the effects of the thickness of IPMCs on their characterization and actuation performances. Yang et al.¹³ fabricated IPMCs with various

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thickness and studied the effect of substrate thickness on their displacements and blocking forces. Yilmaz et al.\textsuperscript{14} investigated nonlinear relationship between electrode thickness and electroactive characteristics of IPMC actuators. Wang et al.\textsuperscript{15} studied the effects of the dimensions on sensing performance of IPMC materials, including thickness, length, and width. Most of them focused on the size effects on the displacements and blocking forces of IPMC samples.

Measurement of the size effects on electrical, electrochemical, and actuation performances of IPMC materials under excitation voltages can provide comprehensive information of IPMC actuators. In this study, Nafion 117 membrane based IPMC material is fabricated and cut into samples with same width and different lengths to study the longitudinal size effects on their electrical and actuation behaviors.

**Materials and methods**

In this work, Nafion 117 membrane (provided by Dupont) based IPMC samples with Platinum (Pt) electrodes were fabricated by the electroless deposition process. Typically, the electroless deposition process mostly requires two steps: the adsorption and diffusion of Pt salt (e.g. Pt[NH$_3$]$_4$HCl) within the Nafion membrane, and the successive reduction of the platinum cations absorbed into the polymer to the metallic state using a reducing agent (generally NaBH$_4$ or LiBH$_4$).\textsuperscript{16–18} Samples with a width of 0.5 cm and lengths of 1, 2, 3, 4, and 5 cm were cut from the IPMC strips to study the size-effect in the electrical response. All prepared samples were stored in deionized water, and the fully hydrated IPMC samples were used for performance testing.

The electroless deposition imparts a complex interfacial layered structure to IPMC due to the penetration of metallic particles into the membrane.\textsuperscript{19–22} Figure 1(a) shows a cross-sectional SEM image of the IPMC with line EDX analysis of the metal (Pt). It indicates that the layer-up structure is composed of the surface metal electrodes, the gradient polymer-metal composites, and the polymer membrane. Different layers play different roles in IPMC transduction.\textsuperscript{23–26} A double-layer capacitor $C$ is formed at the interface of the surface electrode and the electrolyte, electrode resistances $R_{sa}$ ($R_{sb}$) in length-direction and $R_1$ in thickness-direction are introduced by the surface electrodes, and an internal resistance $R_2$ is introduced by the electrolyte. A phenomenological model to illustrate the roles of IPMC is also shown in Figure 1(a).

Due to the making process of IPMC, the surface metal electrodes (also called an outer electrode) is not plain but porous metal,\textsuperscript{27} as shown in Figure 1(b). In this study, voids in the surface Pt metal layer are air, which could not conduct electricity and could be considered as non-isolated inclusions. The electrical resistivity of the outer electrode could be represented as\textsuperscript{26,28}

$$\gamma_s = \gamma_{Pt} \frac{4}{1 - \phi}$$

where, $\gamma_s$ and $\gamma_{Pt}$ ($2.22 \times 10^{-7} \Omega \text{m}$) are the resistivity of the porous and compact platinum metals respectively, $\phi$ is the volume fraction of voids (porosity of the metal electrode). The sheet resistance $R_s$ could be obtained by $R_s = \gamma_s / t_0$, where $t_0$ is the thickness of the surface electrodes. As shown in Figure 1(c), the increase of voids in the metal electrode leads to an increase in electrical resistance.

According to our previous work,\textsuperscript{26} the EDX line analysis of metal element for IPMC samples could be used to obtain the thickness of the surface electrodes, and the surface and cross-sectional SEM images as shown in Figure 1(b), segmented by Otsu’s thresholding...
method, can be used to obtain the porosity of the metal electrode of IPMC samples by using ImageJ software. Specifications of the IPMC samples are listed in Table 1. The surface electrode resistance $R_{sa}$ is considered to be the same as $R_{sb}$ when no voltage is applied on the IPMC sample. The value of the IPMC electrode calculated using equation (1) is $2.26 \Omega/\text{sq}$, which is very close to the measured value $2.41 \pm 0.158 \Omega/\text{sq}$ (measured 10 times using an ST-2258C multifunction digital four-probe tester).

The imperfect electrical conductivity of the metal electrode leads to a decrease in the electric field applied to the IPMC sample along its length ($x$-direction) and causes a nonuniform bending curvature of the sample. Therefore, a distributed RC electric circuit24,25 with a series of similar circuits was developed to describe the performance of IPMC, as illustrated in Figure 1(d). The parameters $R_c = R_{sa} + R_{sb}$, $C$, $R_1$, and $R_2$ in the single unit circuit are assumed to be time-invariant and uniform. It should be noted that the surface resistance of the anode (cathode) electrode decreases (increases) with increasing curvature of IPMC sample, but the sum of the two surface electrode resistances $R_s$ does not change with curvature of IPMC consulting our previous research.14 In other words, the assumption of the single unit circuit is suitable for bending IPMC actuators.

Table 1. Specifications of IPMC samples.

| Specification                  | Description               |
|-------------------------------|---------------------------|
| Ionic polymer                 | Nafion 117                |
| Electrode                     | Pt                        |
| Cations                       | Na+                       |
| Solution                      | Water                     |
| Cross-section of IPMC sample  | $5 \times 0.2 \text{mm}^2$|
| Thickness of metal electrode, $t_0$ | $0.99 \pm 0.083 \mu\text{m}$ |
| Porosity of metal electrode, $\phi$ | $0.611 \pm 0.0316$ |
| Electrode resistance $R_{sa}/R_{sb}$ | $2.41 \pm 0.158 \Omega/\text{sq}$ |

Figure 2. (a) Current response of the 5 cm-IPMC under 2 V DC voltage and (b) dependence of the electrical parameters $R_1$, $R_2$, and $C$ with the length of IPMC samples.

The change of the three parameters $R_1$, $R_2$, and $C$ of IPMC with the length is shown in Figure 2(b). The results show a significant dependence of the electrical parameter per unit length (1 cm) with the length of IPMC samples. All three electrical parameters of per unit length decrease and the trend of the decrease become slowly as the length of IPMC increase. When the length of the IPMC samples changed from 1 to 5 cm, the changes in $R_1$, $R_2$, and $C$ are from $4.75 \times 10^{-4}$ to $2.88 \times 10^{-4}$ $\Omega/\text{cm}$, from $7.50 \times 10^{-5}$ to $3.31 \times 10^{-5}$ $\text{F/cm}$, and from $8.93 \times 10^{-4}$ to $3.23 \times 10^{-4}$ $\Omega/\text{cm}$, respectively.

Results

A testing technique with voltage step pulses proposed by Punning et al.24 was used to study the size effect on the values of the parameters $R_1$, $R_2$, and $C$. A voltage of 2 V was applied on the five fully hydrated IPMC samples through two pieces of silver clamps to measure the responses of electric current in the air at 50% RH and 26°C respectively. The contact clamps covered the whole surface electrodes of all the samples to avoid the influence of the surface electrode resistance on the results.

Figure 2(a) shows the typical current response of the IPMC sample (5 cm-IPMC). At the very first moment when capacity $C$ is totally discharged, the current comes to the peak $i_A$ (2.87 mA). After charging the whole pseudocapacitor in $t_C$ seconds (5.7 s), the electric current remains at a stable level $i_B$ (0.299 mA). The values of the parameters $R_1$, $R_2$, and $C$ can be obtained by analyzing the circuit shown in Figure 1(d), based on the response of electric currents.
The current response of 5 cm-IPMC to 2 V DC voltage was simulated by the model shown in Figure 1(d) to illustrate the importance of the parameters \( R_{sa} \) (\( R_{sb} \)), \( R_1 \), \( R_2 \), and \( C \) for IPMC modeling. The simulated equivalent circuit using different parameters measured from IPMC samples with different lengths was separated into 10 segments to predict electrical currents \( i_{A_{\text{sim}}} \) and \( i_{B_{\text{sim}}} \) (listed in Table 2). The results show that the parameters have a significant influence on the accuracy of model simulation. The error \( e_A \) (\( e_B \)) between simulated current \( i_{A_{\text{sim}}} \) (\( i_{B_{\text{sim}}} \)) from simulation using 1 cm-IPMC parameters and measured value \( i_A \) (\( i_B \)) is 84% (149.5%), which is 8.23 (13.4) times higher than that of 5 cm-IPMC, 9.1% (10.4). Water lose of IPMC samples, which is inevitable during the testing, may leads to the size effects of the length on the electrical parameters. The smaller the sample is, the faster the rate of water loss is, and the lager the error of electrical parameters is.

To explore the actuation behavior of the samples with the same cross-section but different lengths, the electrochemical performances of hydared IPMC samples were characterized by cyclic voltammetry (CV) in aqueous electrolyte with a three-electrode system on Autolab electrochemical workstation (PARSTAT MC). The CV curves of the IPMC samples in a 1 M KOH solution at a 100 mV/s scan rate in a potential window from 0.6 to 0 V are shown in Figure 3. No remarkable peak appears in all the curves for the IPMCs with different lengths. The quasi-rectangular CV curves signify the ideal double-layer capacitor characteristic.

The specific capacitances in CV measurements were calculated using the equation

\[
C_{sp} = \frac{1}{\Delta V v S} \int_{V_1}^{V_2} I dV
\]  

Where \( \Delta V \), \( v \), and \( S \) are the potential window, scan rate, and the weight taken of the IPMC samples, respectively. Namely, the specific capacitances from the integrated area of CV curves are plotted against scan rates, as presented in the inset of Figure 3. The capacitances are 3.91, 3.60, 1.47, 1.73, and 1.41 mF/g at a scan for the IPMC samples with lengths of 1, 2, 3, 4, 5 cm. The result shows that the capacitance decreases as the length of the IPMC sample increase, which is consistent with the previous testing result. The capacitance obtained from the 1 cm-IPMC sample exhibits 3.91 mF/g at a scan rate of 100 mV/s, which is 2.8 times higher than that of the 5 cm-IPMC sample (1.41 mF/g). However, a little change in capacitance with length when the length of the sample is longer than 3 cm.

The measurements of displacement were performed to test the length effect on the mechanical responses of a fully hydrated IPMC actuator in the air at 50% RH and 26°C. All the samples were cantilevered using two parallel Ag plates at one end, and the effective lengths of the beam actuators were 0.5, 1.5, 2.5, 3.5, and 4.5 cm, respectively. The displacements of the points at a 5 mm distance away from the fixed end under DC voltage inputs were detected by a laser displacement sensor. In this configuration, the IPMC cantilever actuators demonstrate significant bending deformation toward the anode. Based on the assumption that no torsional deformations exist, the measured displacements were transformed into bending strain (difference of strain between two surfaces of IPMC sample) by using the following equation.

\[
\varepsilon = \frac{28d}{l^2 + \delta^2}
\]  

where \( d \) is the thickness of the IPMC, \( \delta \) is the extreme deflection of the set point on the IPMC strip, and \( l \) is the length from the fixed end of the IPMC sample to the set point.

Figure 4 shows the extreme strains of the set points on IPMC samples with different lengths under voltages of 1, 2, and 3 V DC. It is obvious the strain of the set-point increases with the increase of the length of the

| \( L \) (cm) | \( i_{A_{\text{sim}}} \) (mA) | \( e_A \) (%) | \( i_{B_{\text{sim}}} \) (mA) | \( e_B \) (%) |
|---|---|---|---|---|
| 1 | 5.28 | 84.0 | 0.746 | 149.5 |
| 2 | 3.41 | 18.8 | 0.446 | 49.2 |
| 3 | 3.12 | 8.7 | 0.412 | 37.8 |
| 4 | 3.15 | 9.8 | 0.354 | 18.4 |
| 5 | 3.13 | 9.1 | 0.311 | 10.4 |

Figure 3. Cyclic voltammetry analysis of the IPMCs with different lengths at a scan rate of 100 mV/s.
testing sample regardless of excitation voltage. Under 1 (2 or 3) V DC voltage, the strain of the set point on 5 cm-IPMC sample is $13.3 \times 10^{-3}$ ($16.1 \times 10^{-3}$ or $19.5 \times 10^{-3}$), which is 5.65 (5.44 and 1.2) times bigger than that of the 1 cm-IPMC sample ($2 \times 10^{-3}$, $2.5 \times 10^{-3}$, or $8.8 \times 10^{-3}$).

**Discussion**

In summary, the effects of sample length on electrical parameters and actuation behaviors of IPMC were systematically examined, based on the analysis of the structural characteristics of IPMC materials. The porous metal electrodes, the polymer membrane, and the polymer-metal composites making up the layered structure of IPMC play the roles of electrode resistance, internal resistance, and capacitance in transduction, respectively. The values of the electrical parameters per unit length, which have a significant influence on the accuracy of model simulation, decrease with the increase of the length of the IPMC sample. The specific capacitance in CV measurement decreases and the bending strain of the set point on the IPMC sample increases, as the length of the sample increases.

**Declaration of conflicting interests**

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