

The Performance of Nafion-Based IPMC Actuators Containing Polypyrrole/Alumina Composite Fillers

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Abstract: A polypyrrole (PPy)/alumina composite filler prepared via in-situ polymerization of pyrrole on alumina particles was incorporated into Nafion® to improve the performance of ionic polymer-metal composite (IPMC) actuators. The IPMCs with the pristine PPy without alumina support did not show bending displacements superior to that of the bare Nafion-based IPMC, except at a high PPy content of 4 wt%. This result was attributed to the low redox efficiency of the PPy alone in the IPMC and may have also been related to the modulus of the IPMC. However, at the optimized filler contents, the cyclic displacement of the IPMCs bearing the PPy/alumina filler was 2.2 times larger than that of the bare Nafion-based IPMC under an applied AC potential of 3 V at 1 Hz. Even under a low AC potential of 1.5 V at 1 Hz, the displacement of the PPy/alumina-based IPMCs was a viable level of performance for actuator applications and was 2.7 times higher than that of the conventional Nafion-based IPMC. The generated blocking force was also improved with the PPy/alumina composite filler. The greatly enhanced performance and the low-voltage-operational characteristic of the IPMCs bearing the PPy/alumina filler were attributed to the synergic effects of the neighboring alumina moiety near the PPy moiety involving electrochemical redox reactions.

Keywords: ionic polymer-metal composites, actuators, polypyrrole, alumina, Nafion.

Introduction

Ionic polymer-metal composite (IPMC) actuators produce bending motion from the migration of solvated free ions, which are loosely bonded with sulfonic or carboxyl anions terminating flexible side chains. Fast and large bending deformations, low-voltage operation (> 2 V), and sensing capability can be achieved in IPMC actuators.¹ With these advantages, rigorous studies have been conducted in the field of IPMC for high-performance actuators and sensors during the last decade.²

For further enhancement of the actuation performance of IPMC in terms of bending displacement and speed, and generated force, a great deal of effort has been addressed to the developments of the polyelectrolyte for efficient movement of ions between the pair of electrodes.³⁻⁹ The targeted polyelectrolyte for IPMCs should possess high ionic conductivities and sufficient mechanical strengths to result in high bending displacement and speed, and the generated force.^{3,4} However, due to the difficulty in the synthesis of

reliable high-performance membranes, the modification of commercial membranes is rather preferred. Nam *et al.* reported IPMCs prepared with Nafion/montmorillonite (MMT) nanocomposite membranes as a modified polyelectrolyte of IPMC.⁴ IPMCs based on the Nafion/MMT membranes possessing enhanced moduli and electrical properties showed improved generated forces in the frequency range of 5-20 Hz. Nguyen and Yoo chose silica and MMT as fillers of Nafion.⁵ The resulting IPMCs displayed largely enhanced displacements and blocking forces due to the reinforced moduli and water-retention abilities of the modified polyelectrolytes as well as the extra metal cations introduced via modification of the fillers. Lee *et al.* fabricated high-performance IPMCs using Nafion/multi-walled carbon nanotube (MWCNT) composite membranes.⁶ They described that the dispersity of MWCNT in the Nafion matrix, which extends to enhanced electroosmotic effects via the combination effect of double-layer electrostatic and quantum chemical effects, played a key role in showing the high actuation performance. The electrode of IPMCs should also possess high toughness, low surface resistances, and large capacitances formed along the interface of the electrode particulate/electrolyte through its mod-

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ification or replacement of material.^{5,7-9} These requirements for IPMCs are similar to the ones for the applications of proton exchange membrane fuel cells (PEMFCs) and direct methanol fuel cells (DMFCs) due to their architectural similarity.¹⁰⁻¹²

In this study, Nafion composite membranes modified with polypyrrole (PPy) or PPy/alumina composite filler were engaged as the polyelectrolytes of IPMCs to improve the performance of IPMCs. The key concept of current work is based on i) the activation of mobile Li^+ ions via the reduction/oxidation of PPy inside the IPMC and ii) the further activation of the Li^+ ions with an aid of the hygroscopic alumina bonded with the highly hydrophobic PPy moiety, which may help the IPMC to bend more rapidly, largely, and powerfully. Related to conducting polymer/metal oxide composite fillers prepared via *in-situ* polymerization, the combinatorial compatibility of PPy and alumina in terms of adhesion has encouraged us to adopt PPy and alumina as the electrochemically reactive material and the hygroscopic ceramic support, respectively.¹³ As an example showing the difficulty in preparation of conducting polymer/metal oxide composites, Yang *et al.* reported that since it is very difficult to form the hydrophobic PPy particles on hydrophilic silica by direct *in-situ* polymerization, they modified the surface of silica via complicate methods using chitosan, and then prepared PPy/silica nanocomposites through *in-situ* self-assembly polymerization.¹⁴ Morphology of the PPy and the PPy/alumina composite fillers prepared in this study is observed via a scanning electron microscope and a transmission electron microscope. From the characterization of Nafion membranes bearing the pristine PPy or the PPy/alumina composite filler, the actuation performances of IPMCs based on the Nafion composite membranes are shown and analyzed through cyclic voltammetric techniques.

Experimental

Materials. Pyrrole (purity: 98%), hydrated ferric sulfate ($\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$, purity: 97%), and sodium dodecyl sulfate (SDS, purity: 99+%) were purchased from Aldrich, and α -alumina (ALO14PB, particle size: 1 μm) was purchased from Kojundo Chemical Laboratory Co., Ltd. in Japan. A 20 wt% Nafion dispersion (DE-2021, DuPont), with a total acid capacity of 0.95 meq/g (0.95 mmol H^+ per 1 g polymer), was used as an ion-conducting matrix of IPMC.

Preparation of Polypyrrole and Polypyrrole/Alumina Composite Fillers. PPy-coated α -alumina filler was synthesized by *in-situ* polymerization of pyrrole on alumina particles. The following procedure was carried out to synthesize the PPy/alumina composite filler. First, a calculated amount of alumina powder was mixed with deionized water and the mixture was milled to acquire finer particles by an attrition mill using 0.5 and 1.0 mm-sized zirconia balls. The resulting alumina slurry comprising 2.08 g of alumina and 9.24 g of

deionized water was obtained. The alumina/water mixture was poured into a 0.1 M aqueous SDS solution of 20.58 g, and then it was placed in an ultrasonic homogenizer for 15 min. 2.08 mL of pyrrole monomer was then added to the mixture and constantly stirred for 30 min. After that, 6 g of $\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$, dissolved in 30 mL of deionized water, was added dropwise to the above mixture and maintained for 5 h with vigorous stirring at ambient temperature. The resulting PPy/alumina composite filler was filtered and then washed with deionized water and methanol repeatedly, followed by vacuum drying at 60 °C. Pristine PPy filler was also synthesized by the same procedure without the addition of alumina.

Preparation of Composite Membranes and IPMCs.

The content of PPy in the PPy/alumina composite filler was 37.7 wt% (PPy:alumina = 1:1.65, w/w), which was obtained through thermogravimetric analysis. Two types of Nafion composite membranes containing the pristine PPy or the PPy/alumina composite filler were fabricated. In both cases, the content of PPy was fixed to 1, 2, 3, and 4 wt%. Unlike the Nafion membranes containing the pristine PPy filler, the Nafion membranes containing the PPy/alumina composite filler possessed not only the PPy component of 1-4 wt% but also the consequent additional alumina component (1.65 times of the loaded PPy by weight). A calculated amount of filler was added to the Nafion solution and mechanically stirred for over 12 h, and then placed in an ultrasonic homogenizer for 1 h. After that, the mixture was poured into a Teflon mold and dried in an oven at 40 °C overnight, followed by an annealing step at 130 °C for 2 h to endow them with insolubility in water. Platinum electrodes were formed on both membrane surfaces for fabrication of IPMCs by an electroless plating method as described in the literature.³ After the plating process, Li^+ -form IPMC was obtained by immersion in a 1 N aqueous LiOH solution for 12 h.

Characterization. Morphology of the prepared fillers was elucidated by a scanning electron microscope (SEM, JSM-6380, Jeol) and a transmission electron microscope (TEM, JEM-1011, Jeol). A complex impedance analyzer (IM6ex, Zahner) was used for the evaluation of electrochemical behaviors in IPMC. Cyclic *I-V* test via a two-electrode system was performed in an air environment by connecting the counter- and reference-electrode leads to one side of the IPMC and by connecting the working-electrode lead to the other side, with a potential range of -3 to 3 V and a scan rate of 50 mV/s. Cyclic voltammograms were obtained using a three-electrode system, in which the working- and counter-electrode leads were connected to the respective sides of the IPMC in a 0.1 N aqueous LiClO_4 solution, while the reference lead was connected to an Ag/AgCl reference electrode (1 M KCl). The applied potential respect to Ag/AgCl was cycled between -0.8 and 1.3 V with a scan rate of 50 mV/s. Actuation test was carried out by measuring the horizontal displacement of IPMC strips, with dimensions of 25 mm in length and 5 mm in width. The IPMC strips were

supported vertically by a gold grip and fixed to 5 mm length on both sides, and thus the actual effective length of the samples was 20 mm. Electric power was applied with an AC source/analyzer (Model 6811B, Agilent). The displacement measurement was performed using a laser sensor (LB-11, Keyence) and a video camera connected to a computer by a data acquisition (DAQ) system (SCB-68, National Instrument).

Results and Discussion

IPMC actuation is based on ion and solvent flux in the polyelectrolyte covered with two metal electrode layers. A higher ionic flux results in a larger bending actuation to the IPMC, and the driving force is referred to electro-osmosis.¹ The redox reaction of PPy¹⁵ may facilitate the ion transport because the redox reaction causes the injection/ejection of mobile ions in/out of the PPy chains in aqueous environment and the resultant ions increase the ionic flux. The hydrophobic nature of PPy, however, prevents the access of water, which limits the injection/ejection of mobile ions. Thus, to reduce the hydrophobic characteristics of PPy, alumina, a hygroscopic metal oxide, was employed as a support for PPy via *in-situ* polymerization.

Figure 1 shows the morphology of the pristine PPy and the PPy/alumina composite fillers prepared in this study. Fine PPy particles which were doped with SDS anions (PPy-SDS) had the diameter of 150-200 nm with spherical morphology, as observed in Figure 1(a). In the PPy/alumina composite filler, alumina particles (dark shaded) measured the diameter of 300-400 nm and were partially covered with PPy (light shaded) agglomerates possessing high tortuosity rather than sphere, as shown in Figure 1(b). Granular features of the two fillers in a wide view were also observed from the SEM pictures given in Figures 1(c) and 1(d). The

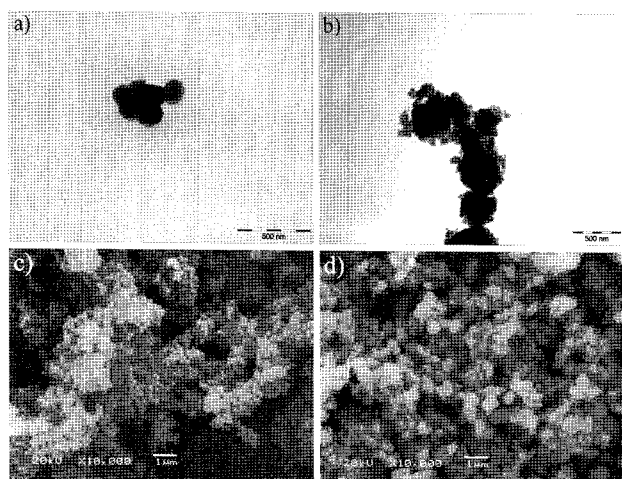
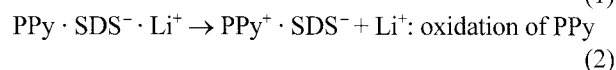
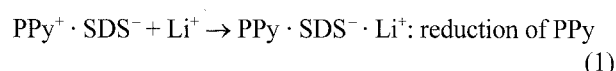


Figure 1. TEM photographs of a) pristine PPy and b) PPy/alumina composite fillers. c) and d) are SEM photographs of a) and b) samples, respectively.

PPy/alumina composite filler dispersed in the Nafion matrix could enable the highly hydrophobic PPy to exist in an abundantly humidified regime due to the stronger hygroscopic effect of alumina as well as to acquire a large specific surface area (electrochemically active area), whereas the PPy alone might experience the lack of water due to the hydrophobic nature. Ions can be transported freely only in moisture regimes. Therefore, the PPy/alumina composite filler in the IPMC may induce a larger injection/ejection of mobile ionic species (Li^+ ion) into/out of the PPy chains via the reduction/oxidation of PPy under an applied potential, as given in eqs. (1) and (2),¹⁵ respectively.



In addition, the redox reaction of PPy is expected to occur actively at the electrolyte-electrode interface of IPMC, with a less extent towards the middle section of the electrolyte. It is due to the highest potential difference occurred at the interfaces in the electrolyte.¹⁶ Under applied voltages exceeding the redox potential of PPy, the injection of Li^+ ions on PPy at the cathodic interface of IPMC and the ejection of Li^+ ions from PPy at the anodic interface may promote the ion transport. Especially in the case of IPMCs comprising the PPy/alumina composite filler, more efficient redox reactions are expected due to the presence of hygroscopic alumina. The conceptual schematics on the actuation mechanism of IPMCs with the pristine PPy and the PPy/alumina composite fillers were presented in Figure 2.

Nafion composite membranes comprising the PPy and the PPy/alumina composite fillers and their IPMCs were prepared, and their properties and characteristics were summarized in Table I. Electrical conductivity of the synthesized PPy was 9.5 S/cm, and that of the PPy/alumina composites was 3.7 S/cm. This indicates the connection of PPy domains for the pathway of electrons in the bulk state of the PPy/alumina composites, with high loading of PPy on alumina particles. The hygroscopic effect of alumina moiety was evidenced from higher water-retention abilities of the PA series membranes bearing the PPy/alumina composite filler, compared to the bare Nafion membrane and the P series membranes with the pristine PPy. Meanwhile, water uptake was higher for both P and PA series membranes than for the bare Nafion. Generally, water in ionic polymers favorably influences the proton conductivity because ionic species are transported by means of well-known Grotthus 'hopping' and vehicle mechanisms.^{17,18} In this study, however, proton conductivity slightly decreased with loading of filler in both cases. In the dry state, Young's moduli of the membranes were also measured. The modulus of the PA series membranes greatly increased with loading of filler, while the P series membranes showed less

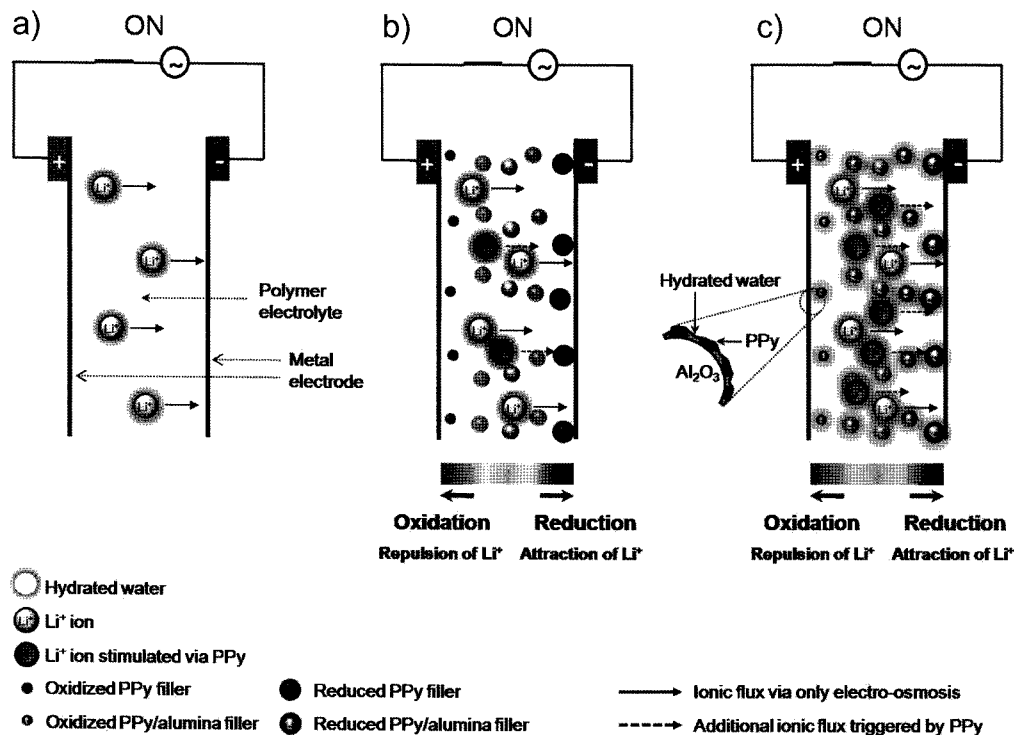


Figure 2. Conceptual schematics on the actuation mechanism of IPMCs with pristine PPy or PPy/alumina composite filler. a) is assigned to the IPMC prepared without adding fillers, and b) and c) are assigned to the IPMCs with the pristine PPy and the PPy/alumina composite fillers, respectively.

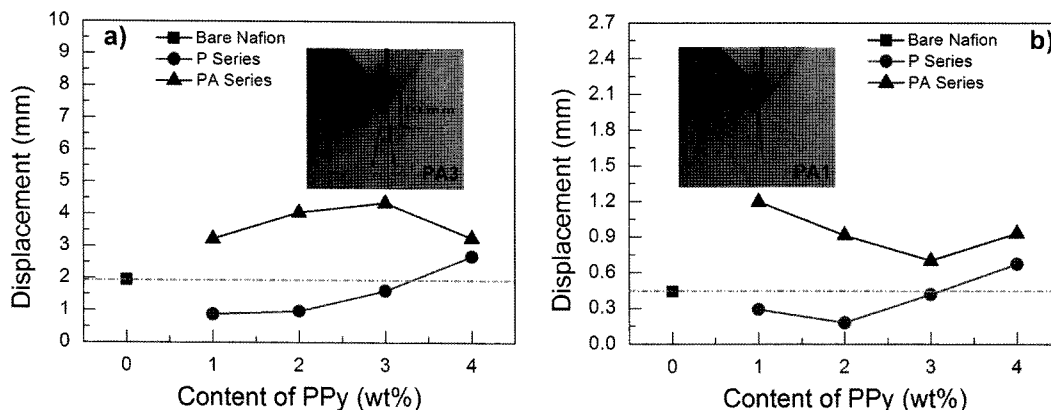


Figure 3. Cyclic displacement of Nafion-based IPMCs containing the pristine PPy or the PPy/alumina composite filler. The IPMC samples were vertically fixed by a gold grip and the actuation test was carried out by measuring the cyclic horizontal displacement (y) of IPMC strips at 10 mm distance from the grip under rectangular voltage pulses of a) 3 V and b) 1.5 V at a frequency of 1 Hz. The insets of a) and b) display the bending shapes of PA3 and PA1 samples at each applied potential, respectively.

significant change in modulus.

Under 3 V and 1.5 V rectangular voltage inputs of 1 Hz, the bending performance of the IPMCs was evaluated by measuring the cyclic horizontal displacement (y) of IPMC strips at 10 mm distance from the grip, as shown in Figures 3(a) and 3(b). At both 3 V and 1.5 V, the displacement of the P series IPMCs was lower than that of the bare Nafion-based IPMC except for the PPy content of 4 wt%. It was also noted that the displacement vs. filler content varied with the elastic modulus, which was confirmed by mechanical

data in Table I. However, the PA series exhibited enormously improved displacements at all compositions compared to the bare Nafion-based IPMC. The bending displacement of the PA series IPMCs at 3 V increased with the increase in filler content up to 3 wt% (at 3 wt%, 2.2 times larger than that of the bare Nafion-based IPMC), and then decreased at 4 wt% probably due to its high modulus,²⁰ while the displacement at 1.5 V showed the largest displacement at the low content of 1 wt% (2.7 times larger than that of the bare Nafion-based IPMC) with a fluctuating tendency of actua-

Table I. Properties and Characteristics of Fillers, and Their Membranes and IPMCs

Electrolyte	Filler			Membrane				IPMC		
	Type	EC ^a S/cm	Code	PPy Content wt%	Alumina Content wt%	Water Content wt%	Water Retention ^c wt%	PC ^d S/cm	Modulus (dry) MPa	Thickness (wet) μm
Nafion	100% PPy	9.5	P1	1	0	40.0	39.8/9.0	0.087	272	458
			P2	2	0	41.5	38.5/8.9	0.084	282	508
			P3	3	0	41.2	38.2/8.1	0.083	257	437
			P4	4	0	41.6	42.3/11.8	0.083	253	479
Nafion	PPy/Al ₂ O ₃ composites (PPy:Al ₂ O ₃ = 1:1.65)	3.7	PA1	1	1.65	41.5	52.2/9.8	0.086	284	395
			PA2	2	3.30	42.0	54.5/10.4	0.081	329	422
			PA3	3	4.95	41.7	58.4/10.1	0.080	347	453
			PA4	4	6.60	41.5	59.7/10.5	0.078	382	434
Nafion	-	-	N	0	0	38.8	41.3/9.6	0.087	232	433

^aDC electrical conductivity of pelletized samples obtained using the standard four-probe method at room temperature. ^bWater content was estimated from the weight change of the membrane between the fully dry and wet states at room temperature. ^cThe residual water content after 20-min-exposing the swollen membrane in air/the content equilibrated in air (temp.: 25 °C, relative humidity: 19%). ^dProton conductivity obtained using a custom-made cell for the normal four-probe technique at room temperature.¹⁹

tion with PPy content. The largest displacement of the PA1-IPMC under 1.5 V was attributed to the electrochemical and mechanical properties optimized at the low content of 1 wt% in terms of the bending performance of IPMC. It is worth mentioning that the cyclic displacement of the PA1-membrane-based IPMC at 1.5 V was almost a comparable level to that of the conventional Nafion-based IPMC at 3 V. However, convergency of the cyclic displacements with increasing content of filler, especially at the highest loading of 4 wt%, was observed. This indicates that a higher loading of the PPy filler may yield a larger displacement. The blocking force under DC 3 V was also measured at the tip of IPMC strips using a load cell. In Figure 4, the maximum blocking force of the PA3-IPMC at 3 V was almost 2 times higher than that of the bare Nafion-IPMC and the values were 0.82 g_f and 0.42 g_f, respectively.

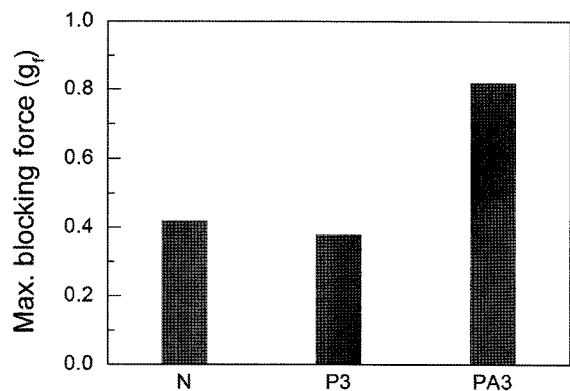


Figure 4. Maximum blocking force of Nafion-based IPMCs containing the P3 or PA3 filler. These data were obtained at the tip of IPMC strips under DC 3 V.

Cyclic voltammograms of IPMCs obtained using a two-electrode system show the trend of ionic movements (Li⁺ ion) induced by the applied potential, with the electrolysis of water inside the IPMC. In the cyclic voltammetry, a higher current density implies not only a higher capacitance but also a larger ionic movement, which usually produce a higher bending performance of the IPMC strip.⁵ Based on this statement, cyclic *I-V* curves of the electrochemically active IPMCs bearing PPy were compared with that of the bare Nafion-based IPMC in Figure 5. The P4- and PA4-membrane-based IPMCs were chosen to detect the deviation effectively. Unlike the bare Nafion-IPMC, the IPMCs with PPy displayed an extra current density at low poten-

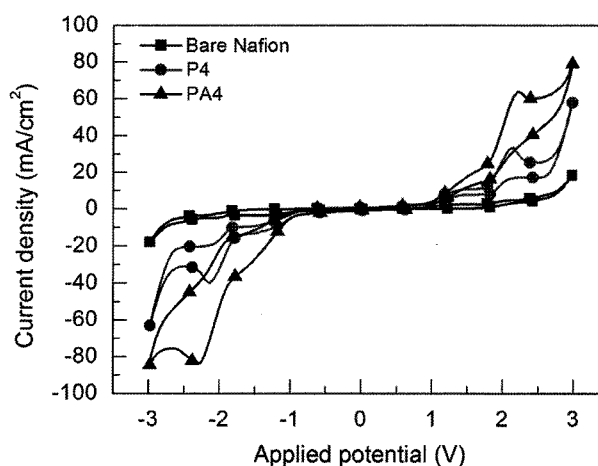


Figure 5. Cyclic *I-V* curves for Nafion-based IPMCs containing the P4 or PA4 filler. The measurement was performed using a two-electrode system in air and the curves were the 3rd cycle of scan with a potential range of -3 to 3 V and a scan rate of 50 mV/s.

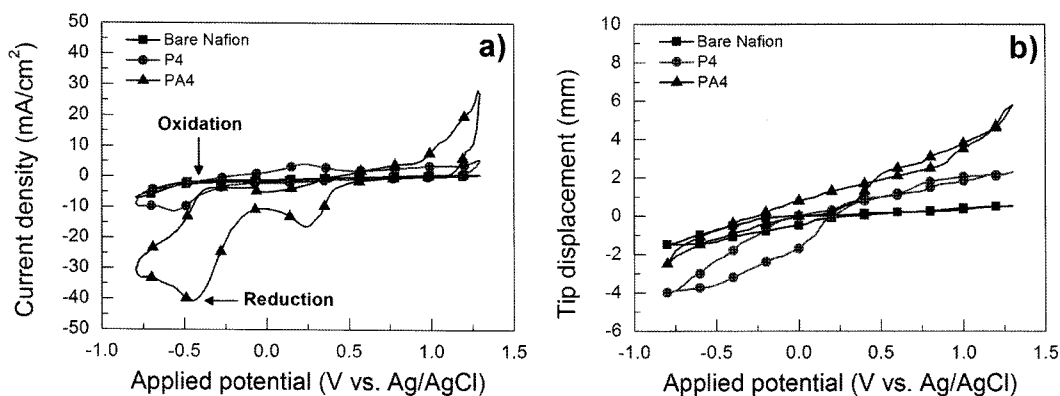


Figure 6. a) Cyclic voltammograms and b) the consequent bending displacements for Nafion-based IPMCs containing the P4 or PA4 filler. The cyclic voltammograms were obtained using a three-electrode system in a 0.1 N aqueous LiClO₄ solution with an Ag/AgCl reference electrode (1 M KCl), after 30 cycles of scan with a potential range of -0.8 to 1.3 V and a scan rate of 50 mV/s. The consequent bending displacement was obtained measuring the horizontal displacement of IPMC strips at the tip of 20 mm away from the grip under the above cyclic potential scan.

tials of ca. $\pm(0.9-1.7)$ V, which is too low to induce the transport of Li⁺ ions in IPMC.⁵ Moreover, the extra current density of IPMCs with the PPy/alumina composite filler was higher than that with the pristine PPy filler, which indicates more ionic movement induced by higher efficiency of the redox reaction. This result clearly explains the high actuation performance of the PA series IPMCs at the low voltage of 1.5 V. In addition, from the fact that the slopes of current density for the IPMCs containing PPy are higher than that for the bare Nafion-based IPMC in the higher potential range of $\pm(1.7-3.0)$ V, it is viewed that the redox reaction of the PPy moiety dispersed in the IPMCs also contributes to the electro-osmotic drag of lithium ion.

To confirm the effect of PPy on the performance of IPMCs, the reduction/oxidation reaction of PPy inside the IPMCs and the resulting extra displacement were compared in the range of -0.8 to 1.3 V versus Ag/AgCl employing a three-electrode system, as seen in Figure 6. As was reported in the literature,¹⁵ the redox reactions of PPy in the P4- and PA4-membrane-based IPMCs were found between -0.6 to -0.3 V versus Ag/AgCl, although the oxidation peak was somewhat obscure probably due to the overlap with ionic fluxes induced by the electro-osmotic drag of lithium ion and the coexisting back diffusion in the potential range. Within the range of -0.8 to 1.3 V, another peaks and higher current densities for the IPMCs bearing PPy were observed, which is attributed to ionic fluxes stimulated from the injection/ejection of Li⁺ ions through PPy, as previously mentioned. From these results, it was postulated that the addition of PPy into the polyelectrolyte layer of IPMCs contributes to the bending actuation. It was also observed that the current density followed the order of PA4 > P4 > bare Nafion and the consequent tip displacements of the P4- and PA4-membrane-based IPMCs were 6.30 and 8.30 mm, respectively, while the bare Nafion-based IPMC recorded 2.03 mm. Again,

the displacements produced by the P4- and PA4-membrane-based IPMCs were 3.1 and 4.1 times higher than that of the conventional Nafion-based IPMC, respectively. From examination of these cyclic voltammograms and bending displacements, it was evidenced that the PPy/alumina composite material as a filler of Nafion-based IPMCs enables the IPMC to perform not only a largely enhanced bending actuation but a low-voltage operation, compared with the sole PPy, owing to the neighboring hygroscopic alumina moiety furnishing the highly hydrophobic redoxible PPy with a much more water as well as an enlarged electrochemically active area.

Conclusions

An evolution of IPMC has been achieved using the Nafion composite membranes containing the PPy/alumina composite filler as the polyelectrolytes of IPMC. The new IPMCs with the PPy/alumina composite filler, which compared with both IPMCs with the pristine PPy filler and without the PPy filler (conventional IPMC), exhibited a largely enhanced actuation performance in terms of bending displacement, response rate, and blocking force. Furthermore, even under a low applied potential of 1.5 V, a viable level of actuation performance could also be approached by IPMCs with the PPy/alumina composite filler. From the cyclic voltammetric analysis and the simultaneous displacement test, the high performance of the IPMCs with the PPy/alumina composite filler was attributed to the PPy's redox reaction of high efficiency and the consequent contribution to the major bending mechanism of IPMC, namely, the electro-osmotic drag of lithium ion.

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References

- (1) M. Shahinpoor, Y. B. Cohen, J. O. Simpson, and J. Smith, *Smart Mater. Struct.*, **7**, R15 (1998).
- (2) A. J. Duncan, D. J. Leo, and T. E. Long, *Macromolecules*, **41**, 7765 (2008).
- (3) M. J. Han, J. H. Park, J. Y. Lee, and J. Y. Jho, *Macromol. Rapid Commun.*, **27**, 219 (2006).
- (4) J. D. Nam, H. R. Choi, Y. S. Tak, and K. J. Kim, *Sensor Actuat. A-Phys.*, **105**, 83 (2003).
- (5) V. K. Nguyen, J. W. Lee, and Y. T. Yoo, *Sensor Actuat. B-Chem.*, **120**, 529 (2007).
- (6) D. Y. Lee, M. H. Lee, K. J. Kim, S. Heo, B. Y. Kim, and S. J. Lee, *Surf. Coat. Technol.*, **200**, 1920 (2005).
- (7) B. J. Akle, M. D. Bennett, and D. J. Leo, *Sensor Actuat. A-Phys.*, **126**, 173 (2006).
- (8) M. Shahinpoor and K. J. Kim, *Sensor Actuat. A-Phys.*, **96**, 125 (2002).
- (9) C. K. Chung, P. K. Fung, Y. Z. Hong, M. S. Ju, C. K. Lin, and T. C. Wu, *Sensor Actuat. B-Chem.*, **117**, 367 (2006).
- (10) M. K. Song, Y. T. Kim, J. M. Fenton, H. R. Kunz, and H. W. Rhee, *J. Power Sources*, **117**, 14 (2003).
- (11) G. Y. Moon and J. W. Rhim, *Macromol. Res.*, **15**, 379 (2007).
- (12) G. Y. Moon and J. W. Rhim, *Macromol. Res.*, **16**, 524 (2008).
- (13) T. Uchikoshi, S. Furumi, T. S. Suzuki, and Y. Sakka, *J. Ceram. Soc. Jpn.*, **114**, 55 (2006).
- (14) X. Yang, T. Dai, and Y. Lu, *Polymer*, **47**, 441 (2006).
- (15) S. Skaarup, K. West, L. M. W. K. Gunaratne, K. P. Vidanapathirana, and M. A. Careem, *Solid State Ionics*, **136-137**, 577 (2000).
- (16) E. T. Enikov and G. S. Seo, *Sensor Actuat. A-Phys.*, **122**, 264 (2005).
- (17) C. Y. Chen, J. I. G. Rodriguez, M. C. Duke, R. F. D. Costa, A. L. Dicks, and J. C. D. Costa, *J. Power Sources*, **166**, 324 (2007).
- (18) Z. Jiang, X. Zheng, H. Wu, and F. Pan, *J. Power Sources*, **185**, 85 (2008).
- (19) S. C. Byun, Y. J. Jeong, J. W. Park, S. D. Kim, H. Y. Ha, and W. J. Kim, *Solid State Ionics*, **177**, 3233 (2006).
- (20) M. Shahinpoor and K. J. Kim, *Smart Mater. Struct.*, **10**, 819 (2001).