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Research article

Bending Controllable IPMC Requires the Particular Structure Silver Electrodes as Well as Dehydration Treatment and Imposed Charge Quantity Control

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Abstract

There has been significant anticipation that IPMCs could serve as effective bending mode soft actuators. However, practical IPMC actuators need to be precisely controllable in their bending, which requires overcoming various challenges. We investigated IPMCs made from different types of ion exchange membranes and electrodes. The results suggest that the following factors are critical for achieving a highly bendable and precisely controllable IPMC:

(i) The formation of a specific silver electrode structure on the IPMC surface.
(ii) Dehydration treatment of the IPMC.
(iii) Control of the charge quantity applied to the IPMC.

Keywords: IPMC; Ion exchange membrane; Silver coating; Dehydration; Bending; charge control

Introduction

Despite a long research history and a strong demand of a practical soft actuator, fabrication of a practical soft actuator is a big challenge even at present. Soft actuators are typically categorized as electroactive polymers (EAPs) in a wet state. Various types of EAPs exist, including hydrogels, conducting polymers, and Ionic Polymer-Metal Composites (IPMCs) [1-8]. Of course, some soft actuators operate not in a wet state but in a dry state. For example, dielectric elastomer actuator (DEA) is a fully dry EAP [9-11]. Although DEAs require quite a high voltage (kV) for activation, the current needed is quite low (~μA), resulting in low required activation energy, which is a significant advantage of DEAs. Super-coiled polymer (SCP) actuators are another type of dry-state soft actuator [12,13]. These actuators consist of a highly twisted polymeric fiber that is thermally controlled by an electrically heat-generating wire attached to the SCP body.

All these soft actuators share several beneficial characteristics, such as low mass, large deformation, low energy consumption, and a soft matrix, among others. Among these soft actuators, we have particularly focused on IPMCs. IPMC is a bending-mode electroactive polymer that was invented by Oguro three decades ago [14,15]. Despite its simple structure, in which an ion exchange membrane is coated with two thin metal layers, the IPMC can exhibit significant bending under very low voltages, as low as a few volts.

Based on our previous study of IPMCs, we observed that the dry-state silver-coated IPMC exhibits relatively good bending controllability through the control of the applied charge. To verify this hypothesis, we investigated whether the silver layer's redox reaction enhances the bending controllability of the IPMC in its dried state. We fabricated five different IPMCs using five distinct types of ion exchange membranes, all of which had silver surface coatings serving as electrodes. The bending characteristics of these IPMCs under electrical stimulation were then examined.

Specimen Preparation

Five types of IPMCs were fabricated as described below.

CMV: Selemion CMV is a cation exchange membrane manufactured by Asahi Glass Co., Ltd. (Japan). In its wet state, it carries immobile negative charges. The surface of this membrane was coated with silver using the silver mirror reaction. The silver-coated Selemion CMV was then cut into small strips (20 mm in length and 2 mm in width). This IPMC, used in the present study, is hereafter referred to as CMV.

Four additional types of IPMCs were fabricated using the following ion exchange membranes: Selemion CMVN, Selemion AMV, Selemion AMVN (all manufactured by Asahi Glass Co., Ltd., Tokyo), and Neosepta (bipolar type, manufactured by ASTOM Corp., Tokyo). These ion exchange membranes underwent the same silver surface coating process as CMV. The resulting IPMCs are hereafter referred to as CMVN, AMV, AMVN, and BP, respectively. Like CMV, CMVN carries immobile negative charges in its wet state, while AMV and AMVN carry immobile positive charges. BP contains both immobile negative and positive charges.

Experiment

We prepared IPMCs in both wet and dehydrated states. The wet-state IPMCs were prepared by simply immersing the fabricated IPMCs, as described in the section "Specimen Preparation", in deionized water, while the dehydrated IPMCs were prepared by leaving the fabricated IPMCs in air overnight.

Each IPMC was clamped horizontally, and electrical stimulation was applied. The induced bending of the IPMC resulted in a vertical displacement of its tip, which was measured as a function of time. This vertical displacement was then converted into bending curvature.

Results and Discussions

CMV Characteristics

We have studied the characteristics of CMV for many years. The dehydrated CMV exhibits significant bending under electrical stimulation, and its bending curvature is almost proportional to the total charge applied. However, this proportional relationship is often significantly impaired when the CMV is in the wet state. These characteristics of CMV are presented here for the first time.

Applying the triangular voltage shown in Figure 1 to the IPMC of CMV can induce bending. Figure 2 shows the relationship between "CMV curvature (C) vs. the total charge applied to the CMV (Q)."

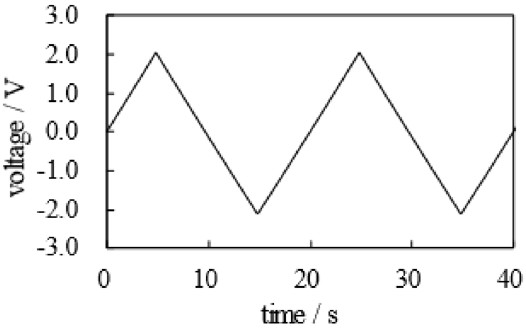

Figure 1: Voltage vs. time imposed on CMV.

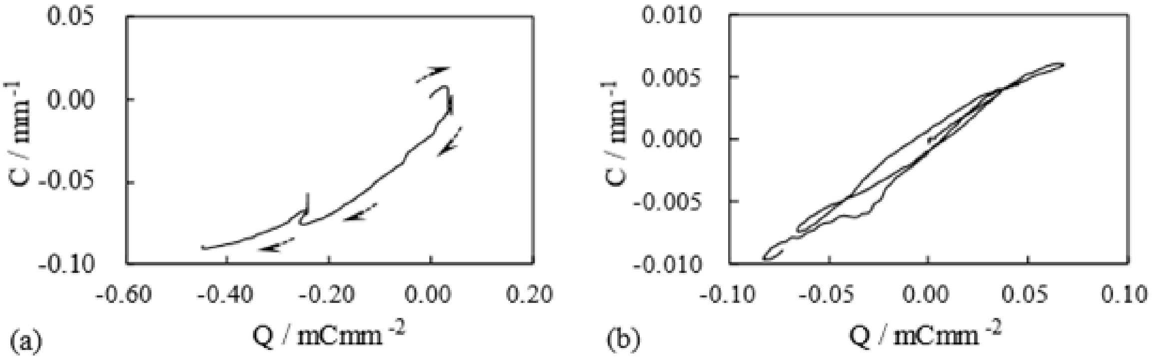

Figure 2: Curvature (C) vs. Charge (Q) when the voltage in Fig. 1 is imposed on the CMV (a) CMV in the wet state where the arrows indicate the direction of time (b) CMV in the dehydrated state

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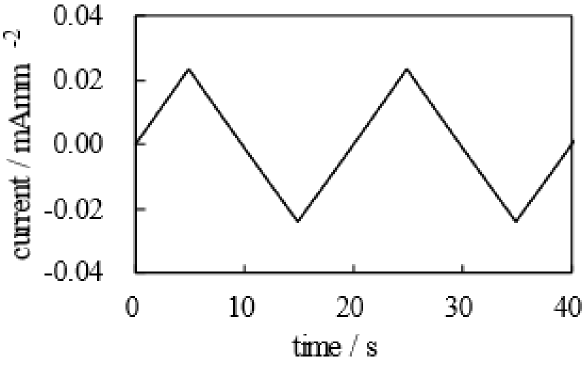

Figure 3: Current vs. time imposed on CMV.

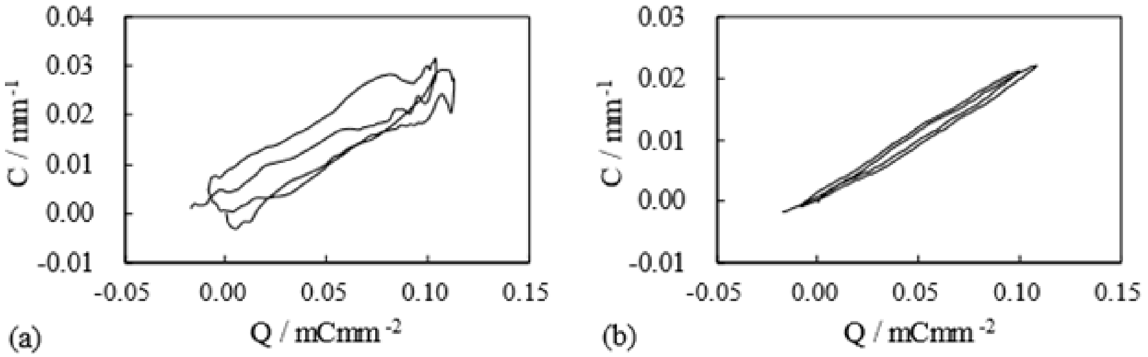

Figure 4: Curvature (C) vs. Charge (Q) when the current in Fig. 3 is imposed on the CMV (a) CMV in the wet state (b) CMV in the dehydrated state

Owing to the control of the current (or charge) applied, both wet and dehydrated CMVs exhibit oscillatory bending curvature. C vs. Q of the dehydrated CMV exhibits almost perfect linearity. Therefore, it is expected that the curvature of the dehydrated CMV can be controlled by the total charge applied. Although the wet CMV does not exhibit as good linearity, as shown in Figure 4 (a), its C vs. Q trajectory is not unpredictably disorderly but relatively orderly oval. Thus, even the bending of the wet CMV can be relatively well controlled by charge quantity. Now we wonder if such a better bending controllability by the control of charge quantity imposed and by the dehydration treatment is universal for any type of IPMCs.

Characteristics of IPMCs Other than CMV

Wet State IPMC Characteristics

First, the bending characteristics of wet CMVN, AMV, AMVN, and BP in response to the applied voltage were investigated, as shown in Figure 1. The results of C vs. Q for these IPMCs are displayed in Figure 5. The C vs. Q relationships for CMVN and AMVN deviate significantly from linearity, making it impossible to control their bending through voltage alone. In contrast, BP exhibits a linear relationship, indicating that its bending can be controlled by the total charge applied. Although the trajectory of AMV is not linear, it follows an orderly oval shape. Therefore, the bending of AMV can be controlled by charge quantity.

For the further discussion to be made at the end of the the section "Bending of IPMCs under the charge control", the time dependence of charge (Q vs. t) when C vs. Q shown in Figure 5 was observed is shown in Figure 6.

Dehydrated State IPMC Characteristics

Next, we conducted the same experiments as described in the section "Wet state IPMC characteristics", but using dehydrated IPMCs instead of wet IPMCs. The C vs. Q relationship for CMVN and AMVN improved to a shape closer to an oval with dehydration treatment, although it still does not exhibit perfect linearity. For AMV and BP, the C vs. Q relationship (see Fig. 7(b) and (d)) appears to deteriorate compared to the wet AMV and BP (see Figure 5(b) and (d)). However, the vertical axis of all diagrams in Figure 7 is much narrower than in Figure 5. Therefore, it is not inaccurate to say that dehydration treatment improves the C vs. Q relationship for all types of IPMCs. The horizontal axis in Fig. 5 (total charge applied to the IPMC) covers a relatively narrow range compared to Figure 7. This indicates that dehydration treatment reduces the current induction in IPMCs, and this smaller current (charge quantity) results in reduced bending of dehydrated IPMCs compared to wet IPMCs. Dehydration inhibits ionization in IPMCs, making current induction less effective. Consequently, conclusions:

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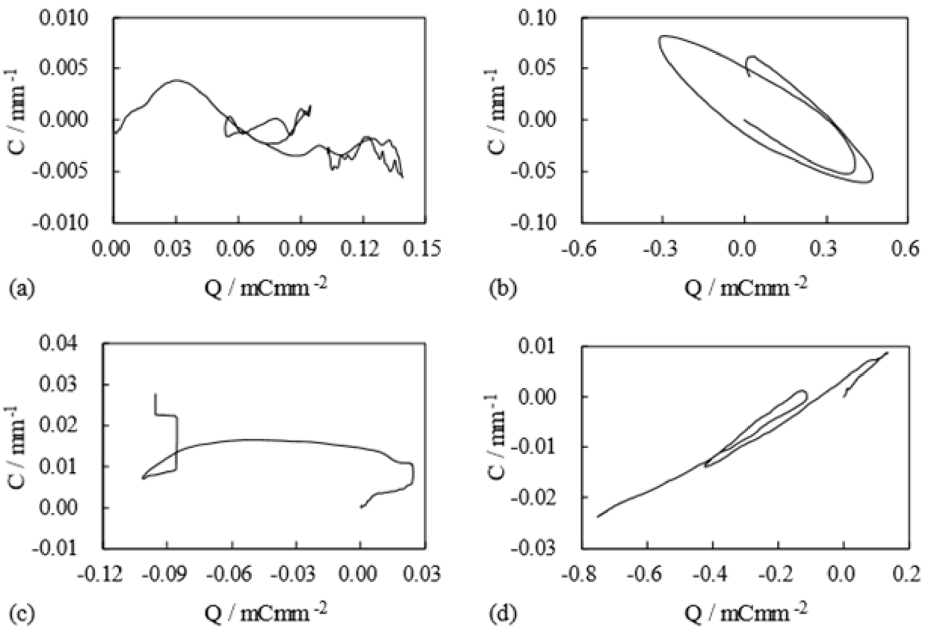

Figure 5: Curvature (C) vs. Charge (Q) when the voltage in Fig. 1 is imposed on (a) CMVN, (b) AMV, (c) AMVN and (d) BP where all the IPMCs were in the wet state

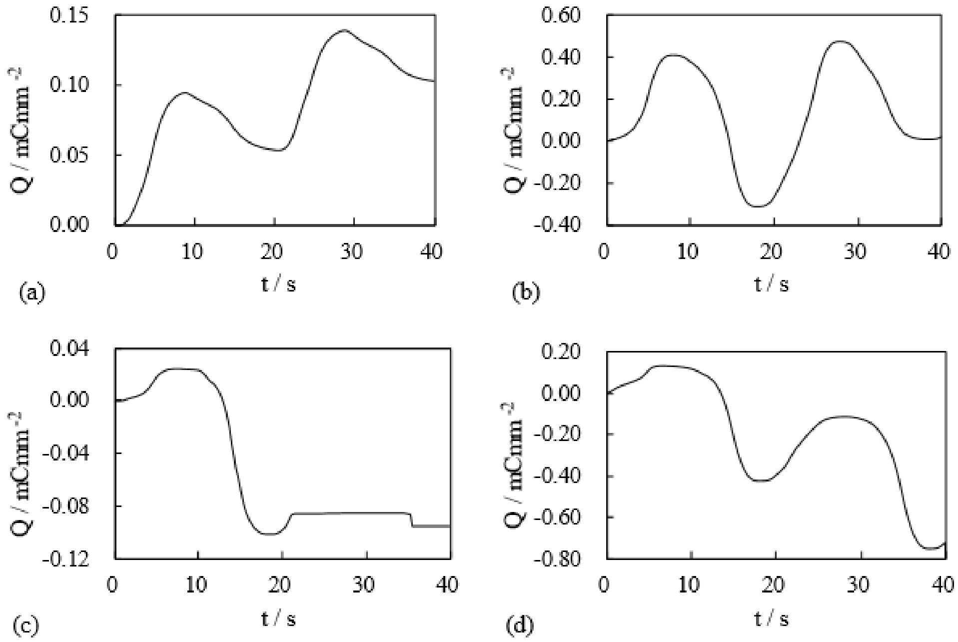

Figure 6: Charge (Q) vs. Time when C vs. Q shown in Fig. 5 was observed (a) CMVN, (b) AMV, (c) AMVN and (d) BP

(i) The bending of IPMCs is governed by the current (or charge) applied.

(ii) Dehydration treatment can improve the linearity of the C vs. Q relationship for IPMCs, but it also reduces the degree of bending. Table 1 summarizes the characteristics of IPMCs under various condition.

Bending of IPMCs Under the Charge Control

Based on the discussion in Sections the section "CMV Characteristics'' and Characteristics of IPMCs other than CMV", it is strongly speculated that the C vs. Q relationship for any IPMC can be improved by dehydration treatment and that bending curvature can be controlled by charge quantity. The same current as shown in Figure 3 was applied to the dehydrated CMVN, AMV, AMVN, and

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BP. Figure 8 displays the C vs. Q relationships for these IPMCs. BP exhibited perfect linearity, while CMVN, AMV, and AMVN did not show a linear relationship but did exhibit an oval shape.

An additional question arises: Is the bending of wet IPMCs controllable through charge control? To investigate this, we applied the current depicted in Figure 3 to the wet IPMCs to observe their bending characteristics. The results are shown in Figure 9. Generally, wet IPMCs exhibit greater deformation compared to dehydrated IPMCs, although CMVN does not follow this trend. The linearity of the C vs. Q relationship for BP deteriorates with dehydration treatment, as evidenced by comparing Figure 8(d) with Figure 9(d).

Table 1: C vs. Q of IPMC

Figure	**state[†1]**	**input[†2]**	**CMVN**	**AMV**	**AMVN**	**BP**
Figure 5	wet	voltage	disorderly	oval	disorderly	linear
Figure 7	dehy	voltage	oval	oval	oval	near-linear
Current of all the IPMCs is low due to their less ionization by the dehydration.						
Figure 8	dehy	current	oval	oval	oval	linear
Figure 9	wet	current	oval	oval	oval	near-linear

[†1] IPMC condition, wet or dehydrated

[†2] electrical input to the IPMC

[†3] C (IPMC curvature) vs. Q (total charge imposed on the IPMC)

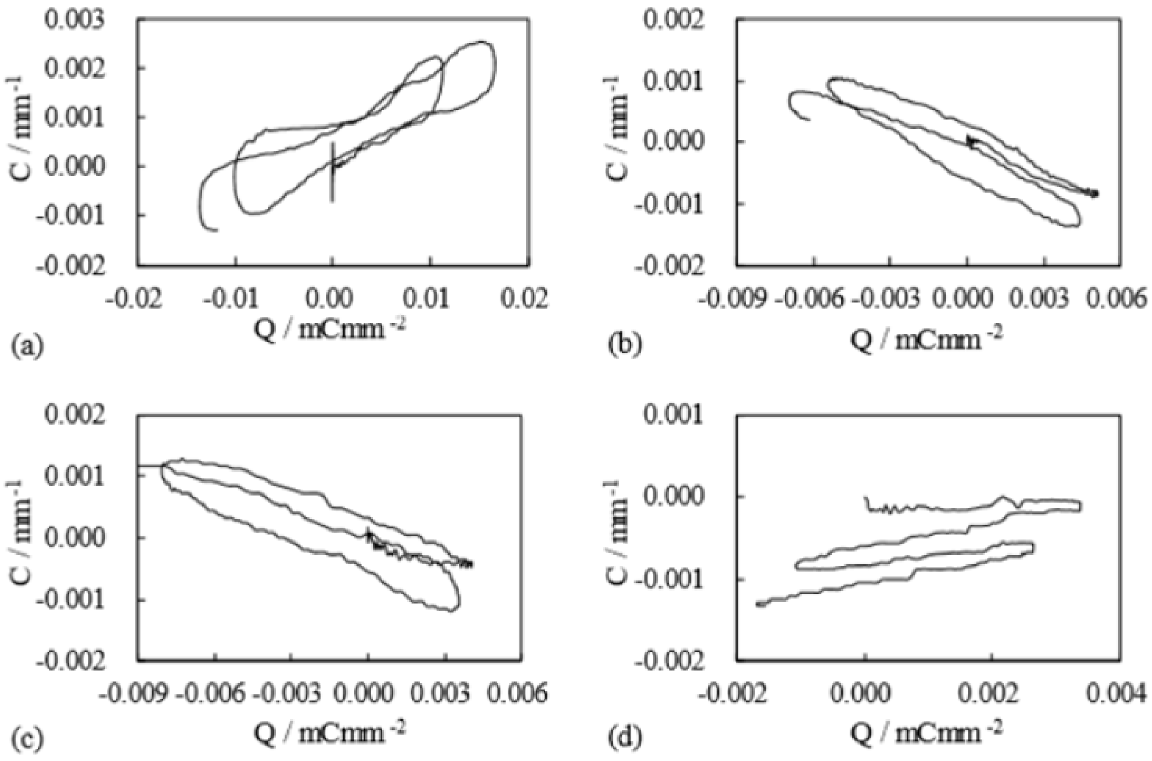

Figure 7: Curvature (C) vs. Charge (Q) when the voltage in Fig. 1 is imposed on (a) CMVN, (b) AMV, (c) AMVN and (d) BP where all the IPMCs were in the dehydrated state

As anticipated, the C vs. Q relationship generally exhibits a specific pattern—either linear or oval—as summarized in Table 1. Therefore, it is reasonable to conclude that the bending of IPMCs is generally controllable through charge quantity control (or current control). The silver-coated dehydrated IPMC can be effectively deformed by controlling the charge (or current).

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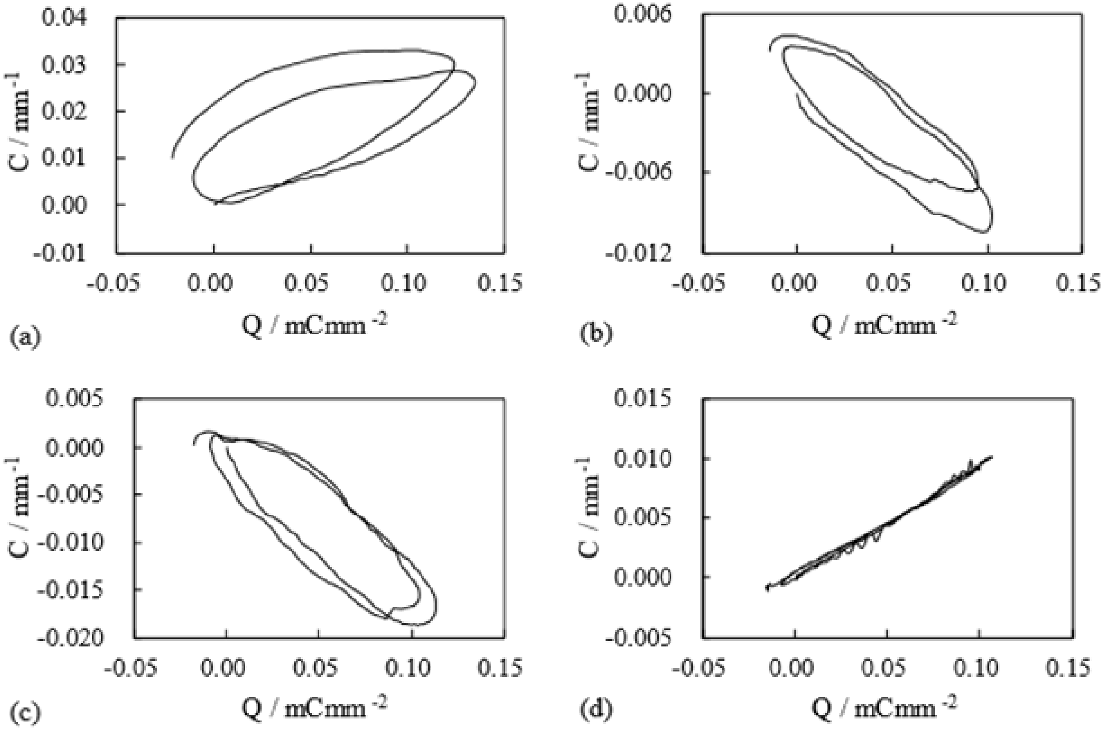

Figure 8: Curvature (C) vs. Charge (Q) when the voltage in Fig. 3 is imposed on (a) CMVN, (b) AMV, (c) AMVN and (d) BP where all the IPMCs were in the dehydrated state

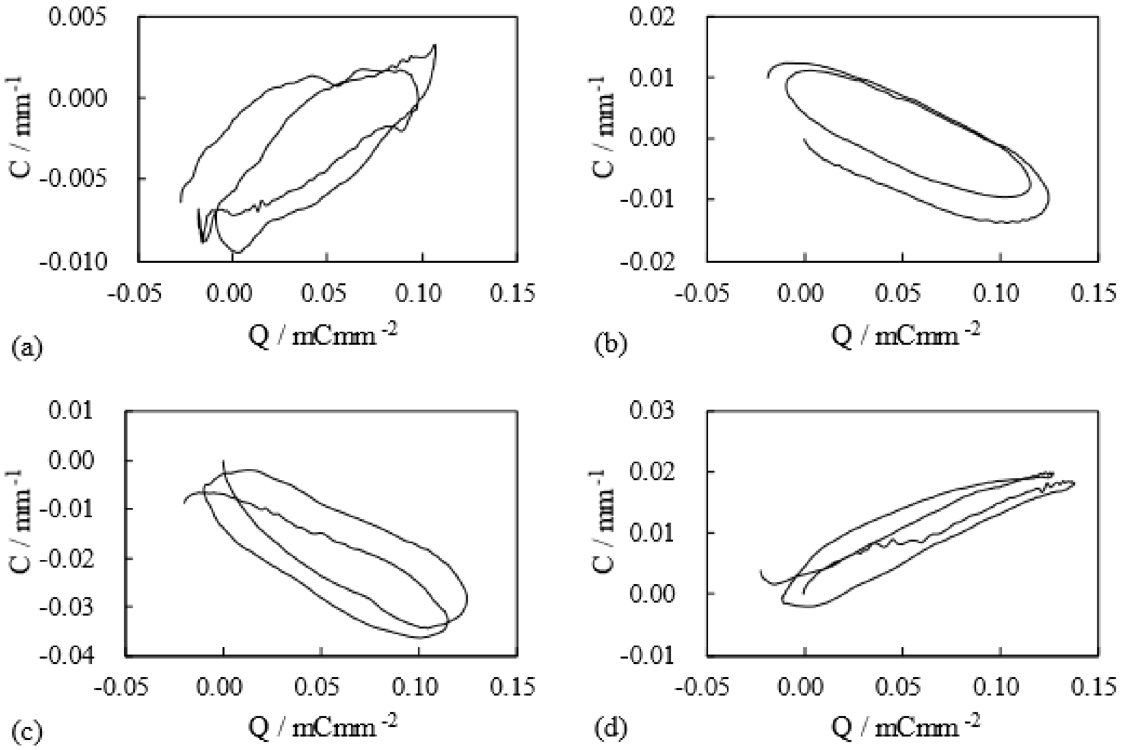

Figure 9: Curvature (C) vs. Charge (Q) when the voltage in Fig. 3 is imposed on (a) CMVN, (b) AMV, (c) AMVN and (d) BP where all the IPMCs were in the wet state

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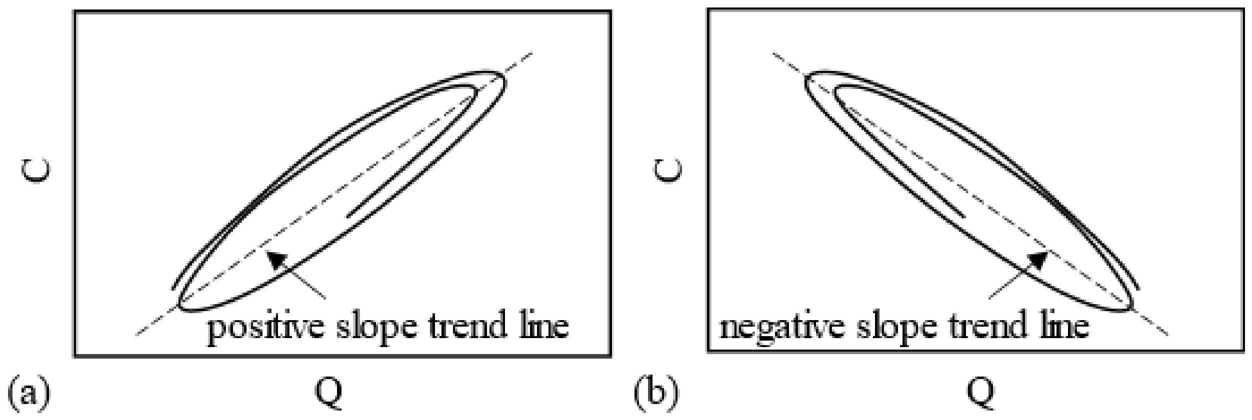

Figure 10: Model of C vs Q (solid curve) and its trend (dashed line) and the definition of its slope (a) Positive slope of Type I and III (b) Negative slope of Type-II

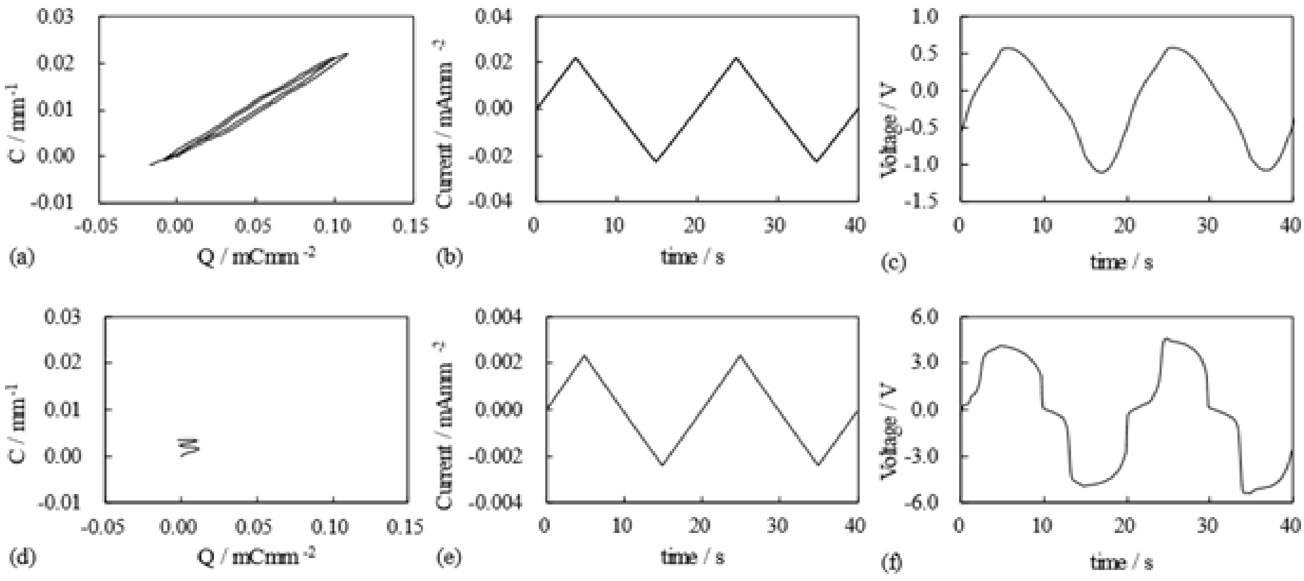

Figure 11: Characteristics of dehydrated CMV in response to the current shown in Fig. 3: (a) C vs. Q (b) Time course of current imposed (c) Time course of induced voltage; Characteristics of dCMV under the current shown in Fig. 3: (d) C vs. Q (e) Time course of current imposed (f) Time course of induced voltage

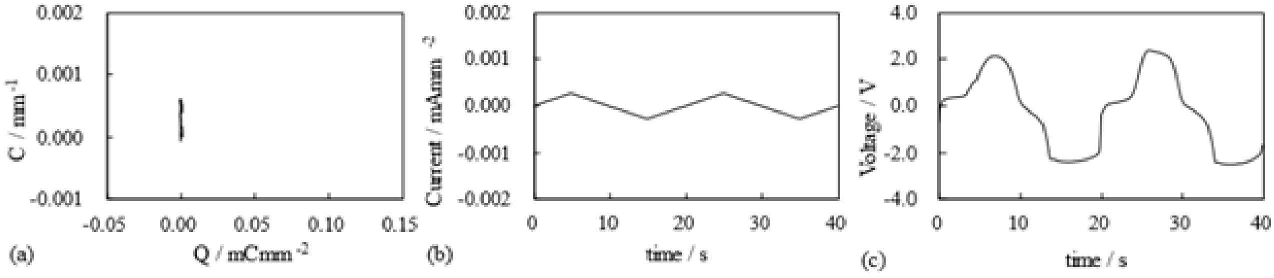

Figure 12: Characteristics of pCMV in response to the current shown in Figure 3: (a) C vs. Q (b) Time course of current imposed (c) Time course of induced voltage

Charge quantity control is quite effective for bending control. However, a comparison of the diagrams in Figure 9 for wet IPMCs with those in Figure 8 for dehydrated IPMCs suggests that dehydration treatment may not significantly enhance bending control. This implies that dehydration treatment does not markedly improve IPMC bending controllability. Nevertheless, the characteristics of wet IPMCs can sometimes be quite unstable, as described below.

Figure 6 shows the time dependence of charge for the diagrams in Figure 5. The bending curvature of CMVN in Figure 5(a) is unpredictably irregular, while the charge shown in Figure 6(a) is less irregular. Although this measurement was performed using voltage control rather than charge (current) control, it is still expected that dehydration treatment would improve the reliability of IPMC bending controllability.

Bending Direction in Response to the Imposed Charge Quantity

We have studied CMV, CMVN, AMV, AMVN, and BP IPMCs, which can be categorized into three types based on the immobile charges they contain:

[Type-I]: CMV and CMVN contain negative immobile charges, as they are cation exchange membrane-based IPMCs.

[Type-II]: AMV and AMVN contain positive immobile charges, as they are anion exchange membrane-based IPMCs.

[Type-III]: BP is an IPMC that contains both positive and negative immobile charges.

Examining the C vs. Q relationships for these IPMCs (excluding BP), as shown in Figures. 2, 5(b), 7(a)–(c), 8(a)–(c), and 9(a)-(c), we observe that for Type-I IPMCs (CMV and CMVN), the C vs. Q relationships exhibit a positive slope. In contrast, for Type-II IPMCs (AMV and AMVN), the C vs. Q relationships exhibit a negative slope (see Figure 10). This indicates that the type of immobile charge in the IPMC governs the bending direction. BP contains both negative and positive immobile charges but exhibits a positive slope in the C vs. Q relationship, similar to Type-I IPMCs (CMV and CMVN). CMV and CMVN contain sulfonic groups, while AMV and AMVN contain amino groups. The sulfonic group is a strong electrolyte, whereas the amino group is a weak electrolyte. Thus, the degree of ionization depends on the type of electrolyte. The positive slope of the C vs. Q relationship for BP suggests that the influence of the negative immobile charges on BP's bending is significantly greater than that of the positive immobile charges. Interestingly, the C vs. Q relationship for BP exhibits perfect linearity, as shown in Figure 8(d). While the sign of the immobile charge affects the bending direction, it is strongly speculated that the characteristics of the polymer matrix also play a significant role in the precisely controllable bending behavior of IPMCs.

Influence of Electrode Structure on the IPMC Bending

From the experimental results described so far, (i) silver coating, (ii) dehydration, and (iii) charge control are fundamentally important for achieving well-controllable bending in IPMCs. Regarding (i) silver coating, electroless plating is typically used to form electrodes on an IPMC. This process employs aqueous chemical solutions, which inevitably hydrate the IPMC, even though dehydration treatment is performed to improve bending controllability. Finding an alternative method for forming electrodes without using aqueous solutions would be beneficial. As an alternative to silver electroless plating, we fabricated IPMCs coated with silver-based electrically conductive paint and studied their characteristics.

IPMCs with the Electrode of Paint Cotaning Silver Powder

Dotite (Fujikura Kasei Co., Ltd. (Tokyo)) is an electrically conducting adhesive containing silver. We prepared five types of IPMCs by coating Selemion CMV, Selemion CMVN, Selemion AMV, Selemion AMVN, and Neosepta (bipolar type) with Dotite. The resulting IPMCs will be referred to as dCMV, dCMVN, dAMV, dAMVN, and dBD, respectively. All the IPMCs were in the dehydrated state. We attempted to apply the current shown in Fig. 3. However, the voltage required to achieve such a current was found to be extremely high, reaching tens of volts. This high voltage requirement undermines one of the key advantages of IPMCs: low energy consumption. We detail these points below.

Figure 11(a) shows the C vs. Q for CMV in the dehydrated state (not in the wet state) under the current depicted in Figure 11(b), where Figure 11(a) corresponds to Figure 4 and Figure 11(b) corresponds to Figure 3. Figure 11(c) represents the time course of the imposed voltage when observing the current shown in Figure 11(b). With these diagrams, Figures. 11(a)–(c) in mind, we now consider Figures. 11(d)–(f) for dCMV. Figure 11(e) suggests that the current imposed on dCMV was only 1/10 of the current imposed on dehydrated CMV, but the induced voltage in dCMV was approximately 6 times greater than that in dehydrated CMV. Therefore, applying the same level of current to dCMV as used for dehydrated CMV requires an extremely high voltage. Consequently, we used a lower current as shown in Figure 11(e) deliberately. However, this is not preferable for achieving practical IPMC characteristics. We need IPMCs that can be activated with both low voltage and low current. Unfortunately, the induced curvature was extremely small even with this low imposed current, as shown in Figure 11(d). All other types of IPMCs (dCMVN, dAMV, dAMVN, and dBD) exhibited similar characteristics to dCMV. Therefore, Dotite coating cannot serve as effective electrodes for IPMCs.

IPMCs with the Electrode of Paint Containing Copper and Silver Powder

Polycalm PTP-G1501 (Plascoat, Kyoto, Japan) (hereafter referred to as "Pcalm") is an electrically conductive paint containing copper and silver. We prepared five types of IPMCs by coating Selemion CMV, Selemion CMVN, Selemion AMV, Selemion AMVN, and Neosepta (bipolar type) with Pcalm. The resulting IPMCs will be referred to as pCMV, pCMVN, pAMV, pAMVN, and pBP, respectively. These IPMCs underwent the same experiments described in the sections "IPMCs with the electrode of paint cotaning silver powder". Figure 12(a) shows the C vs. Q for pCMV in the dehydrated state under the current depicted in Figure 12(b). Figure 12(c) represents the time course of the imposed voltage when observing the current shown in Figure 12(b). Figure 12(b) indicates that the current imposed on pCMV was even less than 1/40 of the current imposed on the dehydrated CMV, but the induced voltage in pCMV was approximately 4 times greater than that in the dehydrated CMV. Therefore, applying the same level of current to pCMV as used for dehydrated CMV requires an extremely high voltage. As a result, we deliberately used a lower current as shown in Figure 12(b), similar to the current used for dCMV (Figure 11(e)). However, this is not preferable for achieving practical IPMC characteristics, just as with dCMV. All other types of IPMCs (pCMVN, pAMV, pAMVN, and pBP) exhibited similar characteristics. Therefore, Pcalm coating cannot serve as effective electrodes for IPMCs.

Paint Type Electrode and Plating Type Electrode

Contrary to our expectations, neither Dotite- nor Pcalm-coated IPMCs exhibited effective bending. According to the diagrams of their currents shown in Figures 11(e) and 12(b), the current induced in dCMV and pCMV was quite low. This suggests that the Selemion CMV matrices in dCMV and pCMV were less ionized compared to even the dehydrated CMV. Why were the dCMV and pCMV so significantly less ionized (or highly dehydrated)? Previously, we found that the bending curvature of dehydrated CMV under electric stimulation heavily depended on the environmental absolute humidity [16,17]. Since Dotite and Pcalm are polymeric paints, they are inherently hydrophobic. Thus, dCMV and pCMV are virtually coated with a water repellent, as illustrated in Figure 13(a). On the other hand, the CMV surface is coated with silver particles due to the formation of silver layers through the electroless plating process. In other words, many tiny particles accumulate on the surface of Selemion CMV, as shown in Figure 13(b). Consequently, water in the air can penetrate through the silver particle layers of the dehydrated CMV and reach the Selemion CMV matrix, making the dehydrated CMV slightly more hydrated than dCMV and pCMV.

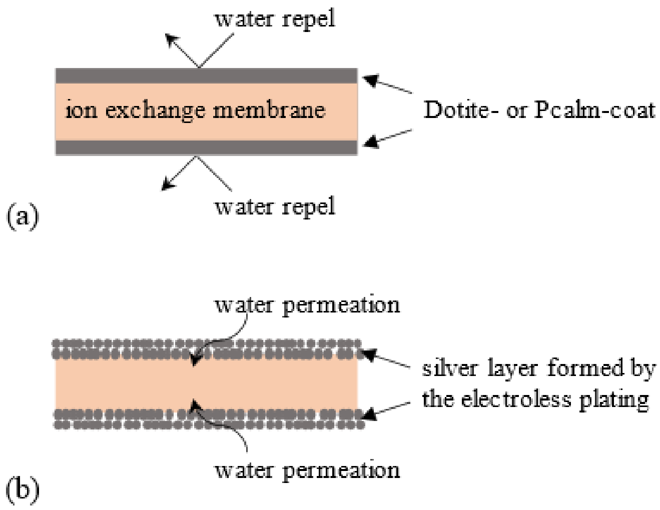

Figure 13: Side view of IPMC (a) Dotite- and Pcalm-coated IPMC where these coatings are hydrophobic. (b) Silver-coated IPMC where the silver coat was formed by the electroless plating. Water can penetrate through this silver coating.

Conclusion

Various types of IPMCs were fabricated and their characteristics were studied in this work. Our study suggests that achieving effectively and precisely deformable IPMCs requires:

(i) Formation of silver electrodes through electroless plating,

(ii) Dehydration treatment of the IPMC, and

(iii) Control of the charge quantity applied to the IPMC.

Additionally, we found that the slope of C vs. Q for IPMCs in the dehydrated state is governed by the sign of the immobile charges contained in the IPMC body. However, precise control of IPMC bending is still challenging to implement. We believe that what is essential is to establish a method for fabricating precisely structured ion exchange membranes. The commercially available ion exchange membranes typically used in IPMC studies are not precisely structured. Consequently, achieving precise IPMC bending control from the outset is difficult. Therefore, future work should focus on developing methods to fabricate precisely structured ion exchange membranes.

Author Contributions

Conceptualization, H.T., I.K. and S.T.; methodology, H.T., I.K. and S.T.; validation, H.T., I.K., S.T., W.L. and M.S; formal analysis, H.T., I.K., S.T., W.L. and M.S; data curation, H.T., I.K. and S.T.; original draft preparation, H.T.; review and editing, H.T. and M.S.; supervision, H.T.; funding acquisition, W.L. and M.S.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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