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Recent Developments About IPMCs (Ionic Polymer-Metal) Composites: A Review of Performances for Different Conditions

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Abstract: It is of great significance for the production of micro robots and new sensors to develop actuators with "muscle" properties. As a kind of electroactive polymers (EAPs), IPMC (ionic polymer-metal composite) can exhibit significant deformation for very low electrical excitation. These composites, known as the "artificial muscle", can be regarded as intelligent bionic materials. With regard to the mechanism of deformation of IPMC, a large number of experimental studies have proved that the variety of electrodes and water contents relating to IPMC have great influence on its electro-mechanical and mechanical properties. Recent research results about IPMC were summarized here to provide a reference for the design and manufacture of these materials. Our conclusions show that, among the main electrode material, though Pt and Au show excellent stability and conductivity, the utilization is limited due to high cost. While Ag, as electrode material, has better application prospect, due to good conductivity and tensile properties. But the optimal voltage under electrical excitation need to be considered when used in specific conditions, or it will cause rapid failure of Ag-IPMC. In addition, IPMC displays high sensitivity to the water content, the service life of IPMC can significantly be prolonged by increasing the degree of humidity of the working environment. So the prospect of IPMC is appropriate to be used in underwater robots, as drivers or actuators.

Keywords: IPMC, metal electrode, water content, deformation law, application strategy.

1 Introduction

With the widespread application of micro robot, bionic robot in engineering and medical, there is an increasing demand of material of high flexibility, high load response and high sensibility to environment. Ionic polymer-metal composites (IPMCs) with polymer as the base material, show high flexibility, low working voltage (less than 5 V), high rate of deformation, and can work under water [Kim, Kim, Nam et al. (2011); Kim, Pugal, Wong (2014)]. Because of these properties, IPMC is widely used in the research and development of underwater robot [Shen, Wang and Kim (2015); Palmre, Fleming, Hubbard et al. (2013)], fluid flow sensor [Lee and Kim (2006)], and is one of the highlights in the field of biomedical applications, such as artificial muscle materials and other biomimetic applications [Lu, Kim, Lee et al. (2008); Shahinpoor, Bar-Cohen, Simpson et al. (2005); Jung, Nam and Choi (2003); Naji, Safari and Moaven (2016)].

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IPMC is a composite obtained by depositing noble metals such as Pt and Au on the substrate of ion exchange polymer membrane (the current researches mainly choose Nafion as matrix material, as shown in Fig. 1). The intermediate layer contains a large number of fixed ions and mobile counterions, as well as a certain amount of solvent water [Kim (2003); Lee and Kim (2006)]. At present, the deformation mechanisms of IPMC under electric excitation mainly contain the hydrated cation movement mechanism [Tadokoro, Yamagami, Takamori et al. (2000)], the cationic electrostatic interaction mechanism [Sia and Li (2000)], and external metal electrode and anionic Coulombic force mechanism [Bonomo, Fortuna, Giannone et al. (2005)]. Through experiment and theoretical calculation, Tadokoro et al. [Tadokoro, Yamagami, Takamori et al. (2000)] found the electric excitation deformation mechanism of IPMC was, when the voltage was applied on the IPMC surface, the counterions directional moved under the effect of electric field, which caused the movement of water molecules. Then the solvent water pressure gradient changed, causing a lot of water accumulated in the cathode caused the cathode to expand, at the same time, the anode was shrinking, resulting in the bending deformation of IPMC. This mechanism has been widely recognized.

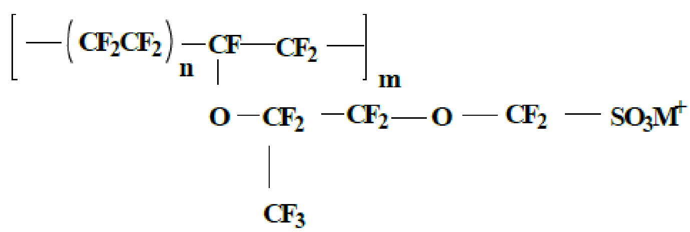


Figure 1: Nafion structural formula [Kim and Shahinpoor (2003)]

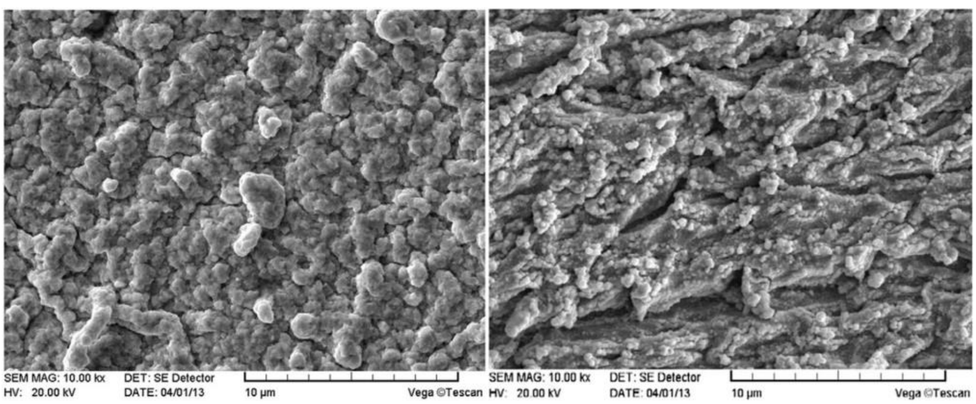
The deformation quantity of IPMC is the size of the end deformation under certain voltage, which is an important property of IPMC, and reflects the induction effect on electric excitation [Biswal, Bandopadhya and Dwivedy (2013)]. Kanno et al. [Kanno, Kurata, Tadokoro et al.] studied the displacement response property of IPMC, by the method of least square fitting, and established the function relationship between voltage and end displacement; then, through experimental and theoretical researches, they found that the deformation of IPMC were closely related to water content and the variety of metal electrodes. The noble metal electrodes on the upper and lower surfaces of IPMC are usually Ag, Au, Pt and Pd. The electrode layer dense degree of different metal electrode is different, and will have an impact on the resistivity of IPMC, lead to the change of deformation of IPMC. In addition, the deformation of IPMC is caused by the movement to the cathode of water molecule under the action of the electric field, causing the sharply expansion of cathode and contraction of anode, which makes the sample bend to the anode, so different water content will also cause different deformation. In order to provide theoretical support for deformation properties of IPMC during design and fabrication, an overview of the effect of electrode variety and water content on the deformation of IPMC was presented, to provide a summary of researches of recent years on IPMC deformation property, and pointed out the development trend of IPMC.

2 Effect of electrode variety on the deformation of IPMC

2.1 Fabrication and properties of IPMC of different electrodes

The main electrode materials of IPMC are Au, Pt, Ag and Pd. Many different methods have been used to combine the noble metal with Nafion to prepare IPMC.

The main method to manufacture IPMC is impregnation-reduction (IR) method. Onishi et al. [Onishi, Sewa, Asaka et al. (2001)] used sodium sulfite and Au^+ solution to carry on plating reaction and got the dendrimers structure of Au electrode generated in the ion polymer. This structure increased the junction superficial area of ionic polymer and electrode, meanwhile the resistivity was very low, and the prepared Au-based IPMC showed long service life of reciprocating bending as well as large bending deformation. After treatment of roughening and cleaning on the surface of Nafion 117 membrane, Wang et al. [Wang, Chen, Wang et al. (2014)] immersed the Nafion membrane into $Pd(NH_3)_4Cl_2$ solution to transmit the palladium cation from the solution close to the inner surface, and then stabilized the palladium cation to metal electrode through reduction process by $NaBH_4$ solution. Wang also manufactured Au-IPMC by similar process, the difference was that the gold cation solution was $[Au(phen)Cl_2]Cl$ and the reducing agent was Na_2SO_3. The prepared Pd-IPMC and Au-IPMC were shown in Fig. 2.



(a) (b)

Figure 2: SEM of (a) Pd-IPMC and (b) Au-IPMC prepared by IR method [Wang, Chen, Wang et al. (2014)]

To realize the properties of light-weight and brilliant flexible behavior, Khan et al. [Khan, Inamuddin and Jain (2016)] prepared SPVA (sulfonated poly vinyl alcohol) solution through the dissolution of PVA in demineralized water and then carry on sulfonation reaction by 4 ml of 4-sulfophthalic acid after filtering. The SPVA-PANI (polyaniline) membrane was then got by the interaction with polyaniline. After roughening by mild sandpaper, the membrane was immersed in the aqueous solution of $Pt(NH_3)4Cl_2$ and NH_4OH to coat Pt metal to prepare SPVA-PANI-Pt IPMC. In the determination of IPMC microgripping system, the end displacement was 14.5 mm under 5.25 V voltage, showed great potential in the application of micro robot, Fig. 3 showed SEMs of the SPVA-PANI-Pt IPMC membranes before and after actuation. Chang et al. [Chang, Chen, Zhu et al. (2012)] chose Nafion ion exchange membrane as substrate, $Pd(NH_3)_4Cl_2$ and $NaBH_4$ as reactants, used sandpaper to polish the surface, 3 reduction plating and 2 electroless plating were carried out, after ion exchange to prepare Pd-IPMC. Kim [Kim (2008)] took Pd metal particles as buffer layer, and then carried on Pt electroless plating, after EDS analysis, found that the composite metal electrode IPMC Pd metal particles penetration depth reached 30 μm. When Palmre et al. [Palmre, Kim, Pugal et al. (2014)] manufactured IPMC of both electrical and mechanical properties, large superficial area Pd-Pt electrodes were prepared. EDS analysis showed that Pd particles reached a penetration depth of 100-200 μm in the Nafion membrane.

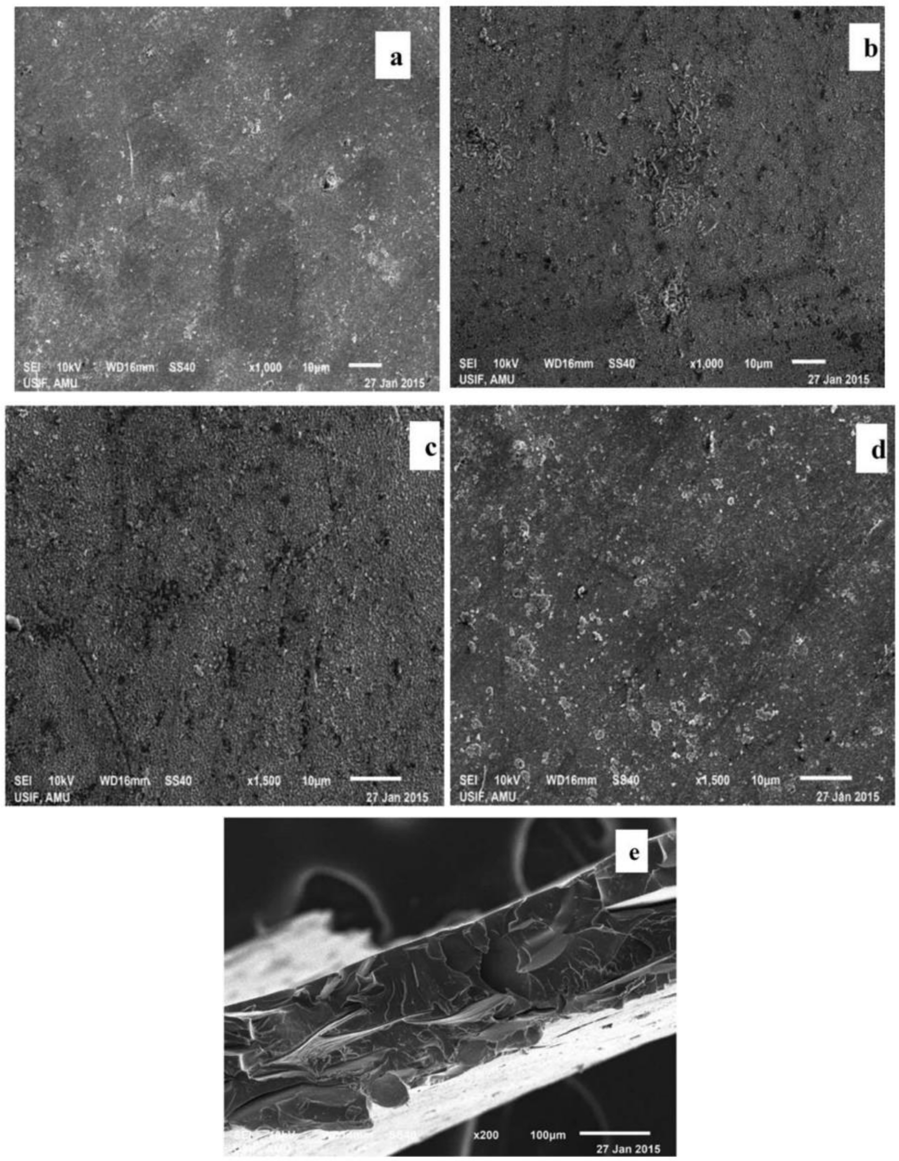


Figure 3: SEMs of the SPVA-PANI-Pt IPMC membranes before (a, c), after actuation (b, d) and cross-sectional view (e) under different magnifications [Khan, Inamuddin and Jain (2016)]

Chung et al. [Chung, Hong, Fung et al. (2006)] used Ag nanoparticle to electroless plate Ag. Ma et al. [Ma and Zhang (2007)] reduced Ag^+ to Ag simple substance, and a layer of silver electrode was formed on the surface of the Nafion membrane, to achieve the high conductivity electrode. There was electron transfer in the reaction process, but there was no external power supply and no extra energy. So, it was a controllable reduction reaction of metastable silver ammonia complex on the surface of Nafion membrane, and the $Ag(NH_3)_2^+$ was reduced to Ag simple substance and deposited on Nafion membrane surface. The microstructure of Ag-IPMC were shown in Fig. 4.

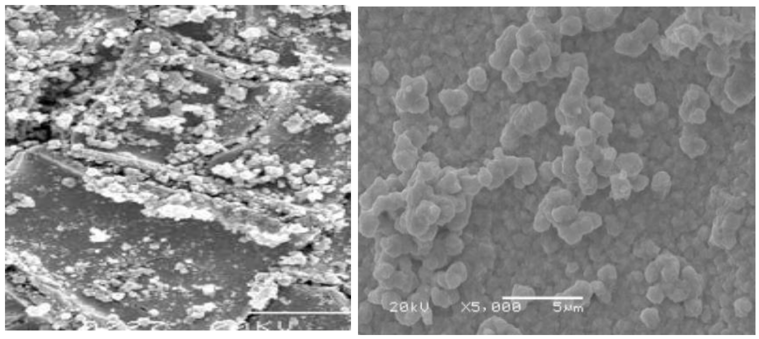


Figure 4: Microstructure of Ag-IPMC under different magnifications [Chung, Hong, Fang et al. (2006); Ma and Zhang (2007)]

The resistivity of different electrodes materials is different, and the density and water storage capacity are different, so IPMCs of different noble metal electrode show different deformation. The deformation laws of Au-IPMC and Pd-IPMC were studied by Wang et al. [Wang, Chen, Wang et al. (2014)]. They found that under the conditions of the same relative humidity, the deformation of Au-IPMC was significantly higher than that of Pd-IPMC. That was mainly because when electrode materials were different, Au had better conductivity than Pd, while the electrode layer was more compact, which further reduced the resistivity. Liu et al. [Liu, Wang, Bian et al. (2010)] made the prepared Pt-IPMC sample drive by DC voltage. When the voltage reached 3V, the deformation displacement of Pt-IPMC sample reached 90°. Bian et al. [Bian, Xiong, Chen et al.] contrasted Ag-IPMC and Pt-IPMC samples by experiment. When the applied DC voltage was 0.2 V, Ag-IPMC sample reacted to the excitation; when the driving voltage increased to 0.4-1.0 V, the deformation of Ag-IPMC showed steady state deformation of about 90°; when the driving voltage increased to 1.2 V, the deformation velocity and deformation rate rapidly increased, resulting in sample curling; while Pt-IPMC reacted to the excitation when the driving voltage reached 1.0 V, and showed a steady state deformation of about 90° when the driving voltage was 5 V. It proved that the driving deformation property of Ag-IPMC was better than that of Pt-IPMC, reflecting in the deformation property and speed of response.

Based on the above analysis, although the stability and conductivity of Pt and Au are very good, the cost are much higher. Pd is more suitable for composite materials electrode. While the preparation process of Ag-IPMC is simple, no pollution, and Ag has good conductivity and tensile properties as electrode, so Ag is more suitable for the large-scale production of IPMC.

2.2 Displacement and force of Ag-IPMC under electric excitation

Ag-IPMC produces large deformation under electrical excitation, and the output force is more than several times the weight of its own. In order to provide a reasonable reference for the application of Ag-IPMC and test the properties of Ag-IPMC, a clamping device test platform had been designed, as shown in Fig. 5. The platform mainly included clamping parts, DC power supply, force measuring device and data processing software [Meredith, Priam and Mary (2011); Punning, Kruusmaa and Aabloo (2007)].

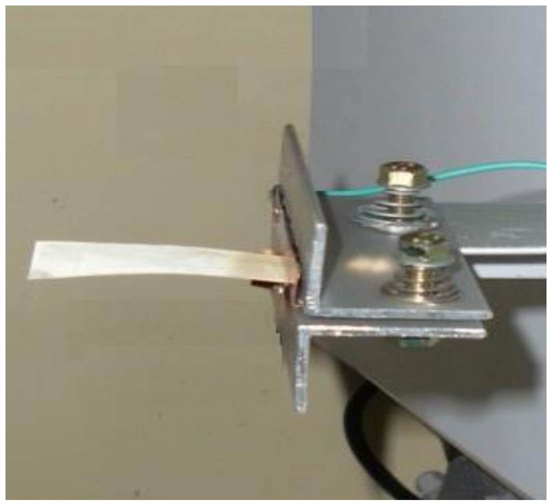


Figure 5: Clamping device test platform of Ag-IPMC [Meredith, Priam and Mary (2011); Punning, Kruusmaa and Aabloo (2007)]

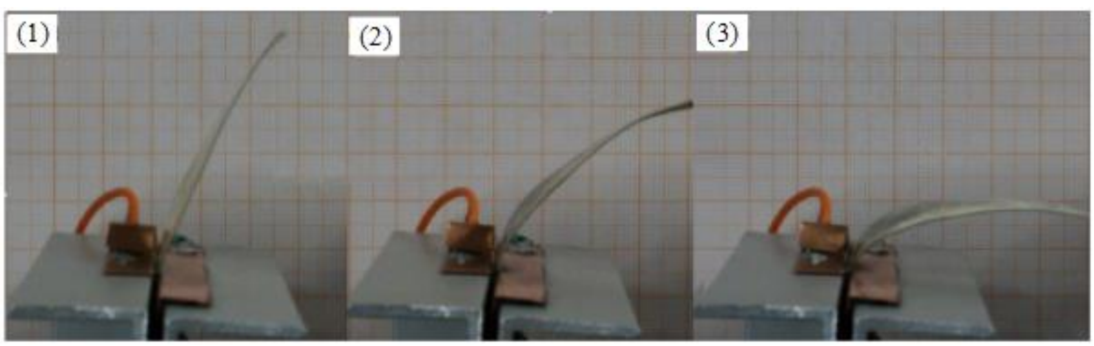


Figure 6: Transient state of Ag-IPMC deformation [An, Xiong and Gu (2010)]: (1) 0.2 s (2) 0.4 s (3) 0.6 s

The Ag-IPMC sample was fixed on one end of clamping device and the other end was free state. The experimental model of cantilever structure was established. The bending deformation of IPMC was measured by high-precision laser displacement measuring system. According to the electrodynamic characteristics of Ag-IPMC, the 0.25 Hz and 3 V sinusoidal driving voltages were applied on both sides of the electrodes [An, Xiong and Gu (2010)]. The Ag-IPMC sample showed bending deformation under electrical excitation, and the deformation was shown in Fig. 6. It was obvious that Ag-IPMC exhibited cantilever beam flexural structure under electrical excitation.

When the input voltage frequency was certain, the relationship between the displacement of free end and the input voltage was shown in Fig. 7. With the increase of voltage, the displacement of free end increased linearly. The maximum output displacement was 32 mm; but when the voltage exceeded 3 V, the displacement began to decline. The changes of the displacement showed nonlinear and instability relationship. At the same time, the sample surface turned black, which meant that the sample tended to failure state [Vahabi, Mehdizadeh, Kabganian et al. (2011); Lee, Park and Kim (2005)]. In summarize, the driving voltage should not exceed the maximum stability value (which was 3 V in this test), and when less than the maximum value, Ag-IPMC free end displacement showed stability change.

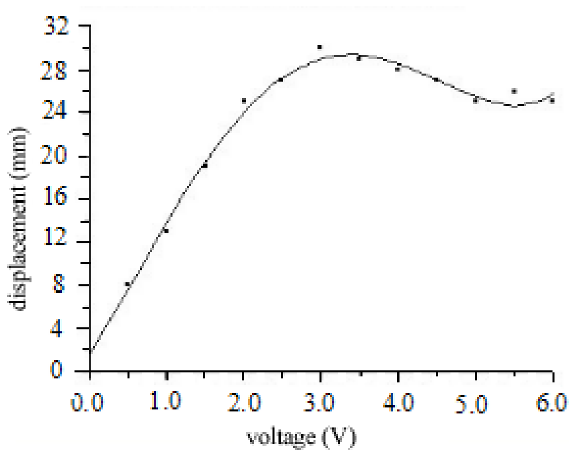


Figure 7: Relationship of displacement and voltage [Vahabi, Mehdizadeh, Kabganian et al. (2011)]

In the test, the Ag-IPMC samples were set under the voltage of 1 V, 2 V, 3 V, 4 V and 5 V to measure the output force. The results were shown in Table 1. From the data in the table, the output force increased with the increase of voltage before the voltage came to 3 V, and the maximum value appeared when the voltage was 3 V. It would accelerate the water molecule electrolysis and electrode oxidation when continued increasing the voltage. At the same time, the output force reduced and the sample failed rapidly [Lee, Park and Kim (2005)].

Table 1: The relationship of output force and the voltage [Lee, Park and Kim (2005)]

Voltage（V）	1	2	3	4	5
Force（mN）	12.5	20.2	28.1	18.6	11.2

In summarize, Ag-IPMC will have an optimal voltage under electrical excitation. When the voltage is less than the optimal voltage, the displacement and output force of the free end will increase linearly with the increase of voltage, and the maximum value will be obtained when the voltage is optimal. When the voltage is applied more than the optimal voltage, the displacement becomes unstable, and the output force will decrease, which causes the rapid failure of Ag-IPMC.

3 Effect of water content on the deformation of IPMC

3.1 Deformation and failure mechanism of IPMC

The hydrated cation movement mechanism is the most widely recognized mechanism to explain IPMC deformation under electric excitation, its principle was shown in Fig. 8, which is due to that the hydrophilic cation carrying hydrated ions to gather in the cathode to form pressure difference, then make IPMC bend to the anode [Gong, Fan and Wang (2009)]. Thus, it can be seen that the amount of deformation of IPMC is closely related to the water content.

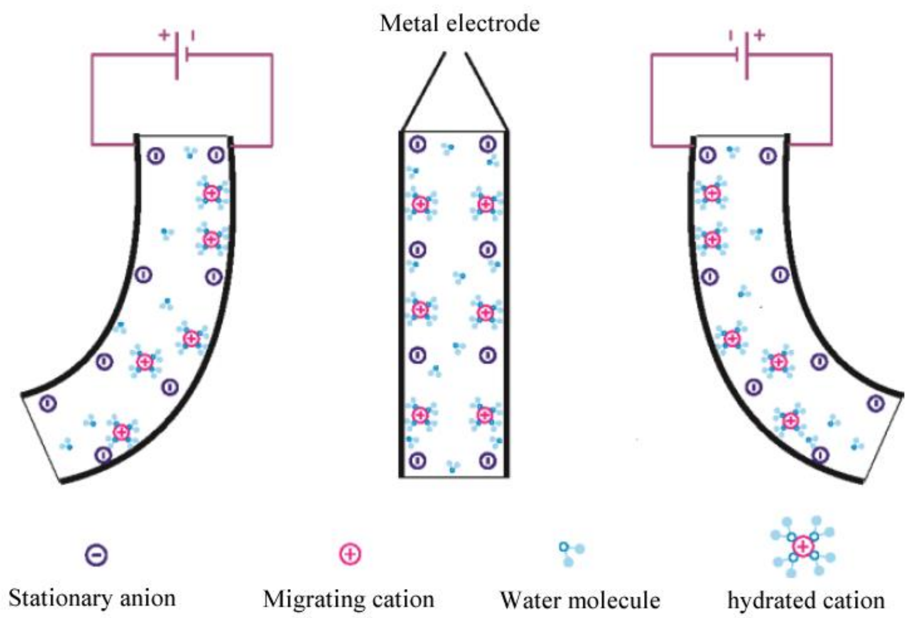


Figure 8: Hydrated cation movement mechanism [Gong, Fan and Wang (2009)]

Lu et al. [Lu, Kim, Lee et al. (2008)] found that because of moisture dissipation, a large number of nano-cracks occurred in the metal layer deposited on the surface of IPMC actuator. Lee et al. [Lee, Park and Kim (2011)], Park et al. [Park, Yoon, Lee et al. (2010)] observed IPMC actuator after multiple actuation, and found that the electrode surface also showed fragmented cracks, as shown in Fig. 9. These cracks cause water molecules to leak out from the electrode crack, affecting the hydration of IPMC and the conductivity of the electrode, driving properties greatly reduces. The study of water content and IPMC deformation law can provide theoretical support and practical guidance for the working environment and preparation of IPMC.

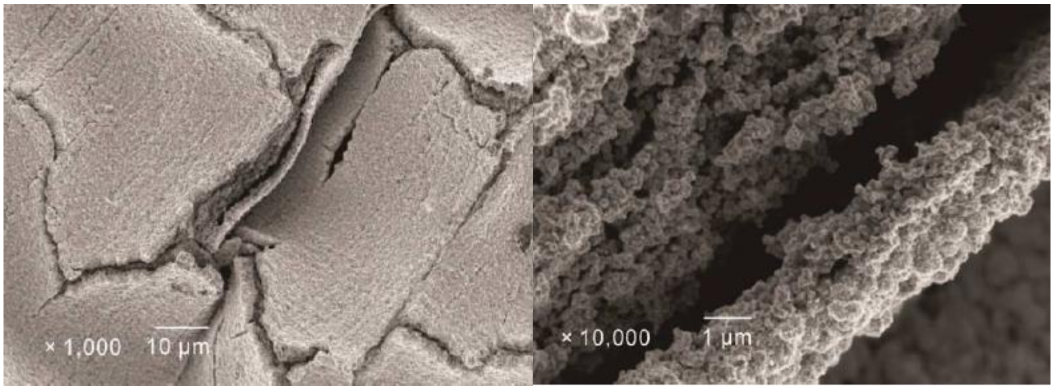
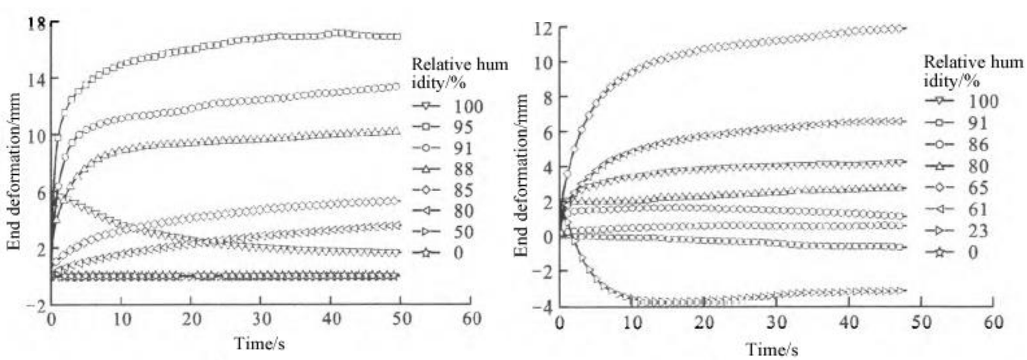


Figure 9: SEM of fragmented cracks of electrode [Park, Yoon, Lee et al. (2010)]

An et al. [An, Xiong and Gu (2009)] measured the geometrical size of IPMC samples under different water content, and the relationship between tensile properties, modulus of elasticity and water content was studied. The theoretical basis was provided for the mechanical modeling of IPMC. In the study of Zhu et al. [Zhu, Asaka, Chang et al. (2013); Zhu, Chang, Takagi et al. (2014)], they proved stresses of IPMC actuator was quite sensitive to water content, with the decrease of water content, relaxation deformation gradually decreased due to the increase of the osmotic pressure and the decrease of total static stress. Wang et al. [Wang, Chen, Luo et al. (2014)] carried on tests to compare Au-IPMC and Pd-IPMC, and got the same conclusion. The relaxation deformation occurred when under high water content and the relaxation deformation disappeared under low water content They also found that under different relative humidity, the performance of Au-IPMC was better than Pd-IPMC, and the deformation of Au-IPMC could reach 17.4 mm, as shown in Fig. 10. In the study of Guo et al. [Guo, Liu, Cheng et al. (2015)], the theoretical steady state results of properties of Ag-IPMC were compared with the experimental results. They found that due to the decrease of water content, the end displacement of IPMC decreased gradually.



(a) Au-IPMC (b) Pd-IPMC

Figure 10: Au and Pd electrode IPMC deformation law [Wang, Chen, Luo et al. (2014)]

3.2 Electrical excitation attenuation test of Ag-IPMC

Liu et al. [Liu, Wang, Bian et al. (2010)] tested the water content of Ag-IPMC in the air by weighing method. Liu put the Ag-IPMC sample on the analysis balance tray and kept the air flow on the surface of the sample. Then they connected the analytical balance with the computer, and set the time as 2 min to send a weighing data. The water content of Ag-IPMC sample was calculated by data processing, and the water content curve of Ag-IPMC sample in the air was shown in Fig. 11.

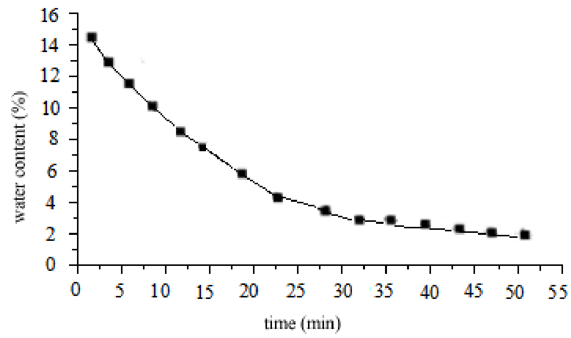


Figure 11: water content curve of Ag-IPMC sample in the air [Liu, Wang, Bian et al. (2010)]

According to Fig. 11, water content of Ag-IPMC sample in air decreased continuously because of evaporation. After about 20 min, the water content decreased and tended to maintain stable at 10-15%. The electro kinetic test of the sample after the loss of water showed the decrease of activity, which meant that the Ag-IPMC sample failed mainly due to evaporation of water in the membrane.

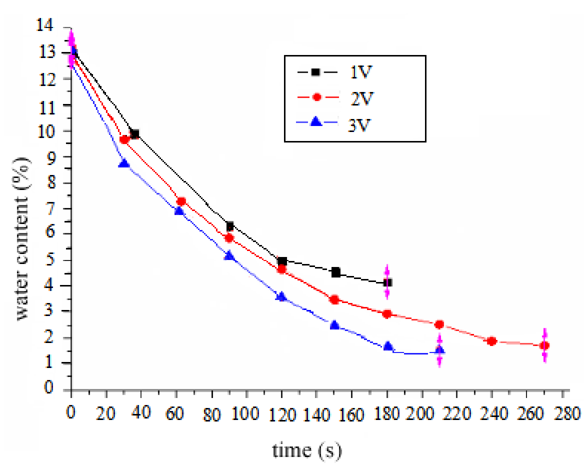


Figure 12: Water content curve of Ag-IPMC sample in air under electric excitation

We used the voltage of 1 V, 2 V and 3 V to drive the Ag-IPMC in the air. The ambient humidity was 50%, and the temperature was 23°C. The mass was measured every 40 s. The average value of several tests was collected to draw water content curve, as shown in Fig. 12. The end point of the curve was when the IPMC could not be driven under the constant voltage. From the figure, the greater the excitation voltage was, the faster the water loss rate was. When the voltage is 1 V, 2 V and 3 V, the minimum water content of stopping movement was 5.153%, 2.708% and 2.5327% respectively, indicating that the higher the voltage was, the higher the attenuation degree of water content of the membrane surface was.

The electric excitation of Ag-IPMC under different voltage excitation was carried out in water. The relationship between the driven time and the water content of Ag-IPMC was shown as Fig. 13. When the excitation voltage was 1 V, the water content rate was 9.736% when Ag-IPMC stopped movement; when the excitation voltage was 2 V, the rate was 6.435%; when the excitation voltage was 3 V, the rate was 5.7%. The higher the electrical excitation voltage, the faster water content decreased.

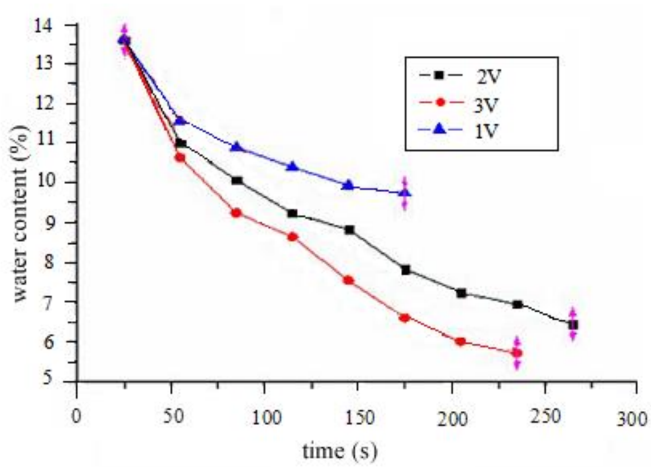


Figure 13: Water content curve of Ag-IPMC sample in water under electric excitation

In comparison of the attenuation of Ag-IPMC in air and in water, the water content in water was much more than that in air when Ag-IPMC stopped movement. In another test, the Ag-IPMC which stopped movement in the test in water would be activated in the air. Therefore, the reason that Ag-IPMC stops in water is not due to lack of water, is because of the resistance of water.

3.3 Application strategy of IPMC

Summarize the above measurement and fitting analysis of IPMC properties under different water content, comparing the response velocity, acceleration and displacement, we concluded that IPMC actuating performance of different water content were different. With the increase of water content, its actuating performance and response velocity became better. The choice of electrode material did not well compensate for the loss of IPMC actuating performance caused by water content loss.

Because IPMC has properties similar to natural muscle, with advantages of low driving voltage, large displacement, fast response, and very low vibration and noise, and because of the high dependence of IPMC on the environment water content, so IPMC is more suitable for application in underwater propeller. Such as Tan et al. (Fig. 14(a)) [Tan, Kim, Usher et al. (2006); Tan (2011)], Eamex company (Fig. 14(b)), Zhang et al. (Fig. 14(c)) [Zhang, Guo and Asaka (2006)], Su et al. (Fig. 14(d)) [Su, Ye and Guo (2010)], Hao et al. (Fig. 14(e)) [Hao, Xu and Liu (2009)], Shen et al. [Shen, Wang, Liang et al.] developed tail fin muscle IPMC robotic fish. Such as Chen et al. [Chen, Shatara and Tan (2009)] developed a kind of bionic fish with IPMC tail fin, as shown in Fig. 14(f), the robotic fish was manufactured by excellent waterproofing materials, the control circuit of the robot fish was enclosed in fish body, the maximum cruising speed could reach 2cm/s. In addition, Nakabo et al. [Nakabo, Mukai and Asaka (2007)] designed the snake robot (Fig. 14(g)), ERI company developed the flapping wing device (Fig. 14(h)) [Shahinpoor, Bar-Cohen, Xue et al. (1998)], and NASA Jet Propulsion Laboratory (JPL) developed artificial joint multi DOF Manipulator (Fig. 14(i)) [Bar-Cohen (2002)].

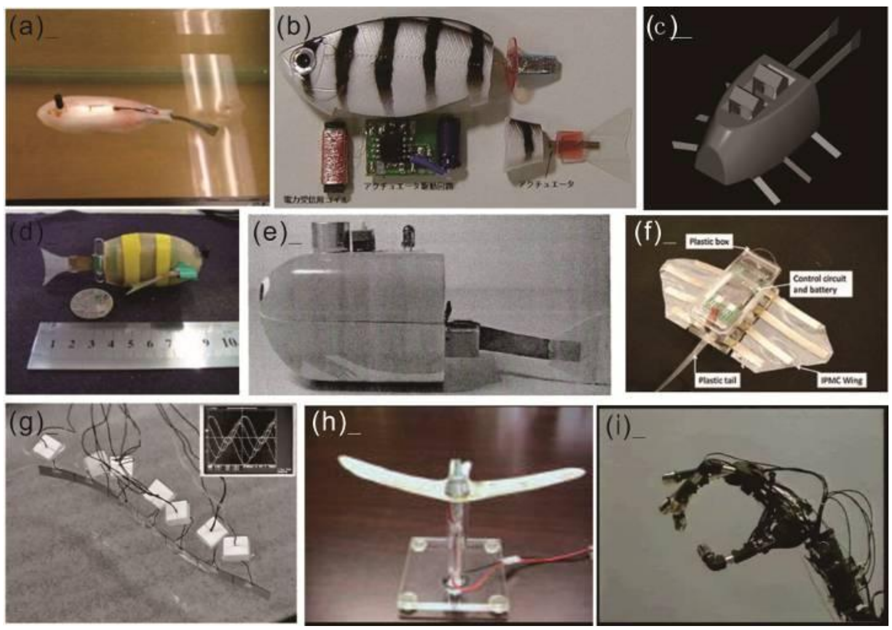


Figure 14: Application of IPMC in robot actuator (a), (b), (c), (d), (e) and (f) tail fin muscle IPMC robotic fish (g) snake robot (h) flapping wing device (i) artificial joint multi DOF Manipulator

To sum up, as the IPMC is highly sensitive to water content, the response velocity and end deformation properties of IPMC decrease rapidly under the condition of water content loss. In view of the character of IPMC, it is necessary to study the preparation method of IPMC with stronger water retention ability, and to improve the relative humidity of the working environment of IPMC. The underwater working environment can effectively maintain IPMC water content in a certain degree, at the same time IPMC can adapt to underwater equipment of underwater complex environment, and realize the lightweight and sensitivity, so IPMC will have important engineering value in the underwater propeller field.

4 Summary and outlook

Compared observation of a large number of experiment of different electrode materials and water content of IPMC samples, analyzed the deformation mechanism of IPMC from many aspects such as surface microstructure and chemical composition, summarized the past qualitative and quantitative analysis of IPMC deformation properties of IPMC, provided a reference for the research of IPMC deformation properties. Firstly, although recently IPMC is mainly used for experiments and research, but with the development of IPMC, it is necessary to consider the preparation of IPMC electrode materials from both properties and cost. While Ag-IPMC has similar property with Au and Pt, and the cost is lower, so is more likely to become the mainstream of raw materials to manufacture IPMC; in addition, as the IPMC is highly sensitive to water content, it is obliged to strictly control the water content during the design and fabrication of IPMC, to prevent water loss, as far as possible to improve the water content of IPMC working environment, if IPMC is applied to underwater equipment, can effectively prolong the service life of IPMC, improve the reaction velocity and deformation efficiency.

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