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Current research status of ionic polymer–metal composites in applications of low-voltage actuators

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The ionic polymer–metal composite (IPMC) is a smart material that displays exceptional electrochemical, mechanical, and optical properties. Its strong electromechanical conversion capability, small size, light weight, wide deformation range, simple actuation structure, and ability to operate at low voltages make it highly promising in applications of biomimetic actuators, such as bionic land-based robots, biomedical devices, and underwater actuators. First, the working principle and preparation process of IPMCs are introduced. Second, the properties and features of IPMCs are briefly reviewed. Moreover, the latest research developments in preparing modified and new composite materials for IPMC actuators to enhance various properties are highlighted. The current challenges in developing new IPMC materials are summarized. In addition, the influence of electrode materials, and macroscopic and microscopic features on the performance of IPMCs are summarized. Finally, hot application spots of the IPMC in different subfields of biomimetic actuators are briefly introduced and prospects for future studies are outlined.

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1. Introduction

The ionic polymer–metal composite (IPMC) is a promising smart material due to its unique characteristics. These include light weight, small size, strong electromechanical conversion capability, high deformability, simple actuation structure, low voltage operation, and environmentally friendly features.¹ The material was first discovered in 1992 by three research groups from Japan and the United States.^{2–4} Unlike the driving methods used with shape memory alloys (SMA),^{5,6} an IPMC can exhibit faster response speeds and better deformation capabilities. Its biomimetic bending response makes it an ideal material for use in human–machine integrated technologies, active skin, and artificial muscles.^{7–9} For example, Ma *et al.*¹⁰ successfully

developed a biomimetic *Forsythia suspensa* based on an IPMC driver and simulated the natural opening and closing process of *Forsythia suspensa*. Ming *et al.*¹¹ developed an intelligent glove based on IPMC sensing, which can be widely used in the field of pulse monitoring. In addition, Lee *et al.*¹² developed a non-invasive throat sensor based on an IPMC, which can accurately identify movement patterns such as swallowing and coughing in the throat. These innovative studies provide strong support for the application of IPMC technology in biomimetic engineering and medical fields. IPMCs are materials with active deformability, mainly composed of ion exchange membranes, metal electrodes, and water molecules sandwiched between them.⁸ When an external voltage (normally) ranging from 1–5 V is applied to the electrode layers, IPMCs undergo bending deformation. However, currently, available commercial base films for IPMCs are expensive and lack moisture retention, making it crucial to develop advanced IPMCs with low cost and excellent performances.

This article introduces the working principle, preparation process, and the properties and features of IPMCs. An analysis of the advantages and disadvantages of various IPMC preparation methods is also introduced. The latest research developments in preparing modified and new composite materials and electrode materials for IPMC actuators to enhance various properties are introduced in detail and highlighted. Hot application spots of the IPMC in different subfields of biomimetic

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actuators are briefly mentioned and prospects for future studies are outlined.

2. Working principle of IPMCs

As shown in Fig. 1(a)–(l), the IPMC exhibits a bending response towards the positive voltage direction upon application and promptly reverts to its initial configuration upon voltage removal. The deformation mechanism of ionic polymer–metal composites (IPMCs) involves the conversion of mechanical, electrical, and chemical energy. Nevertheless, there remains a lack of consensus regarding the nature of the deformation mechanism. Presently, three primary hypotheses try to elucidate the behavior of IPMCs: the hydrated cation migration effect, the electrostatic force effect, and the coulomb force effect. Among these hypotheses, the hydrated cation migration effect is the most widely accepted mechanism.

2.1. Hydration cation migration effect

The hydration cation migration effect pertains to the complex interplay between positively charged ions and water molecules in an aqueous solution. When these ions are introduced into a water-based medium, they are immediately surrounded by water molecules, resulting in the formation of stable hydrated shells. The interaction between hydration ions enables these shells to move through the medium, influencing the motion and arrangement of the ions.

The deformation mechanism of the IPMC is shown in Fig. 2. When immersed in an electrolyte solution, cations migrate toward the polymer membrane of the IPMC. Simultaneously, water molecules bind to the cations, forming hydrates. This process can cause the IPMC to swell and bend, which is known as hydrated cation migration.^{13,14}

2.2. Electrostatic interaction

Electrostatic interaction results in the attraction or repulsion of the ions, as shown in Fig. 3, which plays a critical role in

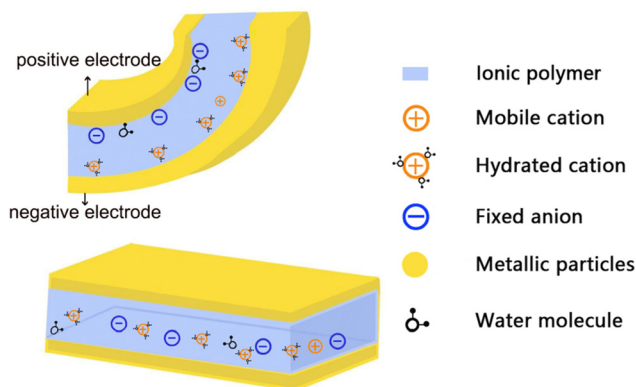


Fig. 2 Schematic diagrams of the hydration cation migration effect of an IPMC.¹⁵

regulating the motion and aggregation behaviors of the ions. In response to an external electric field, cations, which are located within the base layer of the intermediate membrane of the IPMC, migrate toward the negative electrode. Anions, which are attached to the high-molecular-weight chains, also move toward the cationic direction. As a result, the loss of charge balance within the IPMC causes the anode to be in a state of deficit, leading to the relaxation and contraction of the high-molecular-weight chains. The concentration of cations at the cathode creates an electrostatic attraction that causes the high-molecular-weight chains to elongate and bend. This mechanism results in the elongation and bending deformation of the IPMC.¹⁶

2.3. Coulombic force interaction

The coulombic force interaction depicted in Fig. 4 arises from electromagnetic forces between charged particles. Its magnitude is directly proportional to the charges of the particles and inversely proportional to the square of the distance between them. The coulombic force is repulsive when both particles have the same charge, and attractive when their charges are of

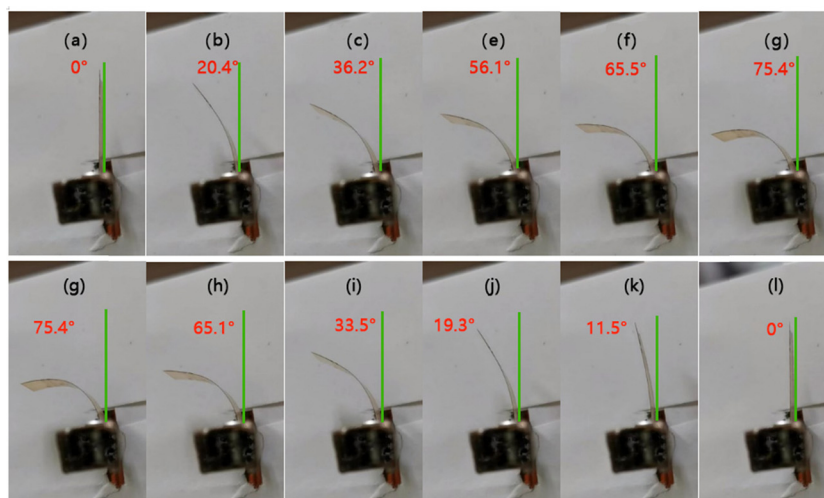


Fig. 1 (a)–(f) Bending response images of an IPMC actuator under the same DC voltage; (g)–(l) recovery process diagram after removing the voltage.



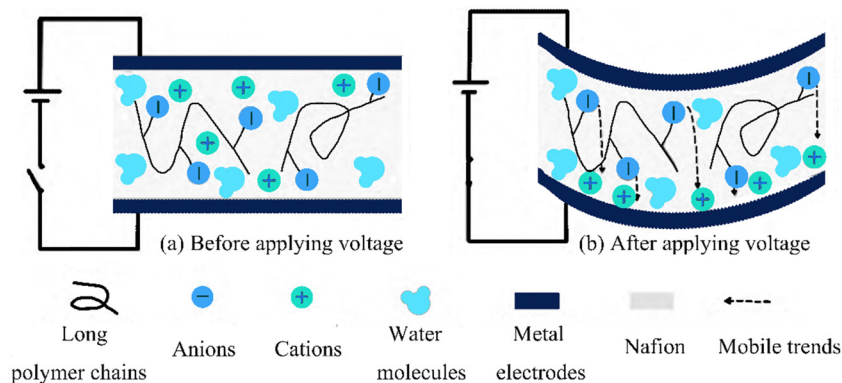


Fig. 3 Schematic diagrams of electrostatic interactions of an IPMC.

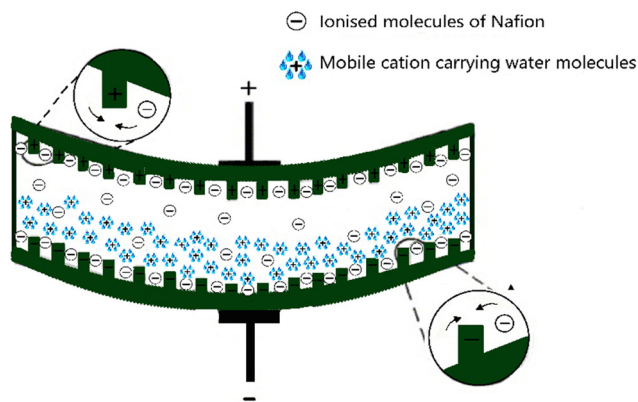


Fig. 4 Schematic diagram of the coulombic force interaction of an IPMC.¹⁷

opposite signs. In an IPMC, the application of an electric field to the ionomer layer initiates the movement of ions within the layer. This movement results in the coulombic force between positively and negatively charged particles, which causes a volume change in the ionomer layer. As a result, the IPMC undergoes bending or stretching.¹⁷

2.4. Back-relaxation

In addition to the aforementioned driving mechanisms, a lesser-known yet interesting phenomenon associated with IPMC actuators is the “back-relaxation”, first documented two decades ago by Asaka and colleagues¹⁸ in sodium-based samples immersed in a Na_2SO_4 and H_2SO_4 solution. They believe that there are double electric layers at the interface of platinum and polyelectrolyte film. Applying a voltage creates different interfacial tension and osmotic pressure at each interface. Due to these differences, bending stresses need to be considered. This mechanism is thought to have no relaxation process. This revelation has spurred further investigation into this physical phenomenon. Vunder *et al.* introduced a novel ionic-electroactive polymer actuator model and conducted experimental validation using three distinct ionic-electroactive polymer materials.¹⁹ Their study offers additional insights into the

significance of back-relaxation in ionic electroactive polymer actuators. Presently, within the academic sphere, there is a consensus that the physical mechanism underlying back-relaxation is linked to the gradual flow of water within the polymer matrix.²⁰ Different from any of these theories, under the physical model,²¹ M. Porfiri *et al.* postulated that the frequently overlooked Maxwell stress in IPMC models has a significant role in elucidating back-relaxation. Through a straightforward temporal analysis of the model, they unveiled a swift deformation towards the anode succeeded by a gradual back-relaxation.²² The inference drawn suggests that back-relaxation might be merely an initial comprehension of the potentiality induced by Maxwell stresses. These theories' complexity may challenge the physical interpretation of the phenomenon and the calibration of simulation parameters from experimental data.

3. Preparation process of IPMCs

The preparation process for IPMCs involves five steps: (1) raw material preparation; (2) base film preparation; (3) ion exchange membrane treatment; (4) electrode plating; and (5) post-treatment (*e.g.*, washing and drying). The primary structure of an IPMC consists of a polymer intermediate layer sandwiched between two electrode layers. Base film preparation and electrode plating are critical steps in the preparation of IPMCs. Ions migrate within the polymer matrix in the polymer layer when an external voltage is applied to the electrode layers of the IPMC due to the conductivity of the ions. To achieve optimal actuation performance for ion exchange membrane-based polymer composites, it is necessary to ensure the density and uniformity of metal particles attached to the surface of the polymer film in the electrode layer during preparation. This paper will focus on discussing the classification and research progress of methods used in preparing the base film and the electrode plating, which are two critical steps in the preparation of IPMCs.

3.1. Preparation of the base membrane

3.1.1. Solution casting method. The solution casting method is a technique in which a resin solution is poured onto a flat substrate, leveled, and formed into a film by evaporating



the solvent as shown in Fig. 5(a). This process involves three steps: (1) solution preparation; (2) solution casting; and (3) solvent evaporation. The method offers benefits such as uniform and easy control of the thickness of the film, but drawbacks include a lengthy preparation time and challenges in identifying appropriate solvents for high molecular weight polymer materials.

Liang Yang *et al.*²³ used the solid solution casting method to produce 158 IPMCs with different thicknesses. Additionally, they chemically plated Pt-IPMCs with five platinum electrodes. The resulting ion exchange membranes were displayed with high uniformity, transparency, and flatness, without any aggregates or bubbles. The morphology, displacement, and blocking force of Pt-IPMCs were assessed under DC voltage.

Zhang Xiaojun *et al.*²⁴ employed the solution casting method to produce a mesoporous IPMC actuator, which comprised of multi-layered multi-walled carbon nanotubes/Nafion membrane. The incorporation of an ordered silica spherical network into the Nafion membrane led to a mesoporous structure that considerably enhanced the electromechanical performance of the IPMC actuator. Liu *et al.*²⁵ developed an IPMC actuator for minimally invasive surgery (MIS) using the solution casting method. They assessed its fundamental characteristics, including displacement and response speed. The experiments suggest that IPMC-infused tubes have great potential to be used in MIS.

3.1.2. Hot pressing. As shown in Fig. 5(b), to prepare an IPMC by using the hot pressing method, a high molecular

weight polymer in multi-layers is subjected to pressure and heat until the polymer reaches a viscoelastic state. After hot pressing, the film is formed in one layer, due to enhancement of the adhesion between interfaces of the multi-layers. The compressed and cooled films are purified through continuous heating and cleaning to eliminate surface impurities.¹⁶ The absence of a boundary layer on the cross-section of the film indicates that the film formation is successful. Hot pressing has many advantages, such as fast film-forming speed, high preparation efficiency, and compatibility with various materials. One drawback of hot-pressing is the requirement for a hot-press machine, which requires significant equipment investment. To improve the driving performance of the IPMC, Jie Ru *et al.* used the hot pressing method to create a composite matrix membrane by placing a Nafion layer between SCNT/Nafion hybrid layers.^{26,27} This research introduced a nanoporous carbon nanotube film electrode with superior conductivity to replace expensive Pt or Au electrodes.

An actuator was produced under vacuum conditions by hot pressing a Nafion/eMIMBF4 electrolyte layer between two electrode films, causing a durable and controllable cycle life of at least 20 000 cycles. This material has the potential to be used as an artificial muscle. Yin Guoxiao *et al.*²⁸ also tested a similar hot-pressing technique to create cylindrical Pt-IPMCs using Nafion rods of different diameters. As the diameter of the IPMC increases, there is a significant increase in both blocking force and anti-relaxation ability. Specifically, under DC driving voltage, a 3.0 mm diameter IPMC exhibits a strong blocking force

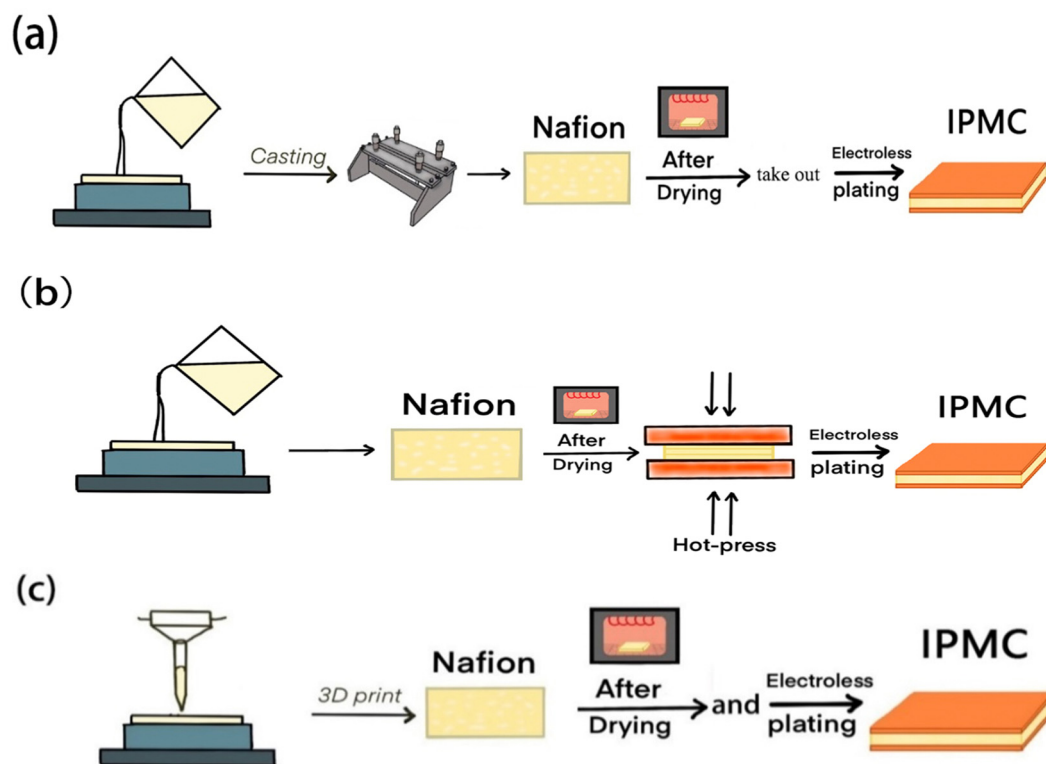


Fig. 5 Schematic diagrams of solution casting (a), hot pressing (b), and 3D printing (c), respectively.



of 323.9 mN and an exceptional power density of up to 139.41 W m⁻³. Additionally, it has demonstrated that the thick cylindrical IPMC can lift objects of 400 times its own weight, which shows its potential for use in soft actuator applications.

3.1.3. 3D printing. Additive manufacturing technology 3D printing can be employed to create base membranes of the IPMC as shown in Fig. 5c. 3D printing is a rapidly advancing technology that can make actuators with intricate geometries.²⁹ Its advantages include a shortened production cycle, high material utilization, and the ability to print complex shapes. However, there are several limitations associated with 3D printing, such as high production cost, slow production speed, low precision, and the need for professional training and operation to ensure quality and efficiency. In a study conducted by Wang Yuyang *et al.*,³⁰ digital light processing (DLP) 3D printing was used to design IPMC actuators made of polyacrylic acid–ethyleneimine. In another study, Hiroyuki Nabae *et al.*³¹ developed a method for manufacturing finer 3D structures that are useful in micro and soft robots. Water and propanol were used as solvents to 3D print honeycomb-structured films, which were combined with an electroplating process to manufacture IPMC actuators.

To broaden the application scope of IPMC actuators, a reliable and adaptable manufacturing process is required to make arbitrary 3D structures. Asuka Ishiki *et al.*³² proposed and investigated a manufacturing method that uses paper and fabric as base materials for IPMC actuators. Compared to PF-IPMCs made of fabric, PF-IPMCs created using paper (tissue, wiping, and towel) exhibited a high deformation ratio, blocking force, and adsorption rate, where the potential value of PF-IPMCs was highlighted.

3.2. Plating electrodes

The most common method for producing IPMC electrodes is chemical deposition.³³ This involves the deposition of an electrode layer on the surface of the base membrane by chemical reaction. This method has many advantages such as low cost, a fast formation speed of the electrodes, and a large deposition area. However, the disadvantages mainly involve poor control over the quality of the thin film and deposition thickness. As a result, alternative methods to replace the chemical plating have emerged, including sputtering deposition, electroplating, and direct assembly.³⁴ Although these methods offer certain benefits when depositing non-precious metals onto polymer films, no single method can create high-performing IPMCs. Therefore, some researchers have combined different methods to create IPMCs with superior performance.²⁸

3.2.1. Sputtering deposition. Sputtering deposition is a commonly used method for producing thin films. The deposition process involves exposing the surface of the target material to a high-speed ion beam in a vacuum environment, which expels atoms that are then deposited on the substrate surface to create a thin film. The method has several advantages, including the ability to produce well-formed, uniform, and dense thin films. However, disadvantages include the need for specialized equipment and a high-vacuum environment, which results in

increased costs and makes it difficult for small-scale production.

In a study conducted by Brault *et al.*,³⁵ sputtering deposition was used to deposit platinum (Pt) onto porous carbon materials that were subsequently integrated into fuel cell electrodes. The resulting Pt dispersion exhibited concentrations of up to 2 μm in the porous carbon medium, leading to enhanced electrode conductivity.³⁶ Hasani *et al.*³⁷ used sputter deposition to produce metal-stacked IPMC electrodes with wrinkled surface morphology. This IPMC exhibits a significant ability to withstand tensile strains from Nafion swelling or IPMC bending, which can significantly reduce the occurrence of microcracks during bending and prevent electrode delamination.

Siripong *et al.*³⁸ introduced a new method based on the sputter deposition for producing Ni–Au–Nafion IPMC materials, which have proved to be both efficient and economical. The method involves the use of DC sputtering to deposit a finely-textured gold electrode layer onto Nafion film, followed by electroplating a thin and flexible nickel layer using a solution containing nickel salt and boric acid. However, the utilization of the ion beam during the sputtering process can affect the water adsorbed on the surface of the Nafion film. This leads to the rearrangement and redistribution of water molecules, which may compromise the mechanical properties and stability of the IPMC. Therefore, the choice between chemical plating and sputtering deposition should be based on specific application requirements, as both techniques have their own strengths and weaknesses.

3.2.2. Electroplating method. To improve the driving performance of IPMCs, researchers have utilized a combination of chemical plating and electroplating methods to create high-performance IPMCs. Electroplating is a surface treatment method that employs electrochemistry. During the plating process, the metal surface that is subjected to the coating process acts as the cathode, while the electroplating metal serves as the anode. Both the cathode and anode are submerged in an electrolyte. Under the electric current, the anode metal is dissolved as the ions, which are subsequently deposited and reduced onto the cathode, *i.e.*, the metal surface, resulting in the formation of a metal coating layer.³⁹

The technology of electroplating offers several benefits for the production of IPMCs, such as improved corrosion resistance, increased rigidity, and enhanced conductivity. Nevertheless, it also has some disadvantages, including high energy consumption, elevated costs, and the generation of defects. To overcome these limitations, Colin Noonan *et al.*⁴⁰ solved these limitations by electroplating platinum onto a substrate made of cellulose nanofiber (CNF)–Nafion, which significantly reduced electrode resistance and improved conductivity. Jin-Han Jeon *et al.*⁴¹ proposed a novel method. They prepared IPMC actuators with multiple electrodes to simulate biological locomotion and flapping movements. This method combines electroplating and chemical reduction using mask technology. The advantage is that chemical reduction ensures the composite of the polymer and platinum particles while electroplating accurately and easily controls the thickness of the electrode with high uniformity.



3.2.3. Direct assembly method. The direct assembly method (DAP) can be used to prepare IPMCs.⁴² Its advantages include high preparation efficiency and low cost. However, it requires high technical expertise, and control of the preparation process is challenging. The method involves several steps: firstly, a specific amount of ionic polymer is dissolved in a solvent to create an ionic polymer solution. Secondly, the electrode solution is applied to the surface of a trimmed metal foil, and the ionic polymer solution is coated on the electrode, *i.e.*, the metal foil. Finally, the assembly is placed on a hot plate to produce a uniform ionic polymer metal composite film on the metal foil.⁴³

Palmre *et al.*⁴⁴ conducted a study on a nanoporous carbon-based electrode assembled on high-strain polymer-based IPMC actuators using the direct assembly method. The actuator exhibited a higher maximum output force than IPMCs prepared using normal methods, due to the utilization of high-porosity carbon material as the electrodes. Oh, *et al.*⁴⁵ investigated the effect of the thickness of the Nafion membrane on the performance of IPMC actuators, particularly the elastic modulus, using the direct assembly technique, and multi-walled carbon nanotubes (MWCNTs) were employed as the electrode material. In conclusion, the direct assembly method is a productive, high-quality, energy-efficient, and eco-friendly approach to creating IPMC electrodes.

4. Properties and features of IPMCs

Several noteworthy characteristics of IPMCs are important for the applications due to the ability to achieve large-angle bending at very low applied voltages. These characteristics include high water uptake (WU),⁴⁶ which facilitates ion transport, low water loss (WL), high ion exchange capacity (IEC), and high proton conductivity (PC).⁴⁷ This paper provides a concise overview of these essential features of IPMCs.

4.1. Proton conductivity

The proton conductivity (PC) of a material refers to the mobility of the ions inside the ionic polymer, indicating the capacity of charges to move within the substance. Ions move through the water in IPMCs and pores are presented in the ionic polymer film, contributing to ionic conductivity. The actuation ability of IPMCs can be enhanced by increasing the PC value, which promotes the excessive migration of H⁺ ions in their hydrated form. Researchers can use a formula⁴⁸ to determine the PC value of an IPMC film.

$$\sigma = \frac{L}{R \times A} \quad (1)$$

where σ , L , R , and A are the proton conductivity, membrane thickness (cm), resistance (Ω), and cross-sectional area (cm²), respectively.

Additionally, an IPMC, a composite material with proton-conductive properties, may change proton conductivity during the deformation process. This change stems from structural

adjustments, ion movement, and proton transfer within the IPMC actuator.

4.2. Ion exchange capacity

The ion exchange capacity refers to the ability of functional groups in a material to exchange with ions in a solution/water. When an external voltage is applied to an IPMC, ions migrate within the ion exchange membrane, altering the material's charge distribution and causing deformation. Hence, the strength of ion exchange directly affects both the response rate and extent of deformation of the IPMC to external electric fields. The formula to calculate IEC is as follows:⁴⁹

$$\text{IEC} = \frac{V \times M}{W} \quad (2)$$

where V , M , and W are the volume of NaOH consumed, the molarity of NaOH, and the weight of the dry film.

4.3. Water uptake

Water uptake (WU) refers to the ability of a base membrane to absorb water within a given time. In the process of IPMC water uptake, water molecules infiltrate the material, altering its electrical conductivity and electrochemical properties, subsequently impacting the IPMC's deformation response. Generally, higher water uptake may significantly affect the deformative performance of IPMCs.⁵⁰ Excessive water uptake can lead to uneven water distribution within the material, increasing the resistance between electrodes, and thereby reducing the response speed and deformation extent to the electric field. Consequently, regulating the water uptake of IPMC materials is crucial for maintaining their stable deformation performance at a high level. The formula to calculate WU is as follows:⁵¹

$$\text{WU} (\%) = \frac{W_2 - W_1}{W_1} \times 100\% \quad (3)$$

where W_1 is the weight of the base membrane before water absorption, and W_2 is the weight of the membrane after the water absorption for a specified time.

4.4. Water loss

The water loss refers to the degree of water loss in IPMCs, mainly caused by natural evaporation and electrolysis processes, and it is closely related to the deformation of IPMCs. An increase in water loss of an IPMC leads to internal structural changes, affecting its ion transport performance and electrochemical reactions. Consequently, higher water loss may diminish an IPMC's deformation capabilities or degrade its performance. This relationship necessitates consideration in the design and utilization of an IPMC, especially in applications requiring long-term stability and reliability. Excessive water loss (WL) inside the IPMC film has a negative impact on its conductivity and response rate, thereby affecting the performance of the IPMC actuator. To determine the percentage of water lost, the following formula is used:⁵¹

$$\text{WL} (\%) = \frac{W_1 - W_2}{W_1} \times 100\% \quad (4)$$



where W_1 and W_2 are the weight of the hydrated IPMC film before and after applying a voltage for a specific duration, respectively.

4.5. Electrode interface surface area

The interfacial surface area of the electrode is defined as the contact area between the electrode and the electrolyte, which is the area covered by the metal particles deposited on the film. Increasing the surface area of the electrode can enhance the response rate of IPMCs.⁴⁵

4.6. Response rate

The response rate of an IPMC refers to the speed at which the actuation of the IPMC responds to the applied electric field, typically occurring within milliseconds. The response rate is influenced by many factors, such as electric field strength (*i.e.*, voltage), water content, temperature, and thickness of the IPMC film.⁵²

4.7. Blocking force

The IPMC blocking force refers to the maximum load or displacement that an IPMC can withstand when exposed to an electric field. This property indicates the ability of an IPMC to resist external force. If the external force exceeds the blocking force of an IPMC, it undergoes permanent deformation and loses control, leading to performance degradation and failure in the end. Liang Yang *et al.* established a mathematical model of IPMC displacement and blocking force and proved the rationality of the model by measuring the displacement and blocking force of a Pt-IPMC,⁵³ and defined that the maximum blocking force of an IPMC can be expressed as follows:

$$F = a_0 + a_1U + a_2U^2 + a_3U^3 + a_4U^4 \quad (5)$$

where a_i ($i = 0, 1, 2, 3, 4$) is a constant. U and F are the input voltage and the maximum blocking force of the IPMC, respectively.

In summary, blocking force is a crucial material property and the strength of the blocking force is typically proportional to properties such as elastic modulus, hardness, toughness, and ion conductivity.^{24,37,52,54}

4.8. Tip deflection/displacement

Fig. 6 shows a simple diagram of testing methods for measuring deflection, and displacement, and high frame rate cameras are used to capture the motion trajectory of IPMCs, measuring tip displacement through displacement sensors.

The tip deflection/displacement refers to the extent of bending or displacement at the tip end of the IPMC actuator when the voltage is applied, as shown in Fig. 3, and the tip deflection increases with increasing working time. It is a typical evaluation to assess the driving force and response efficacy of IPMCs.^{30,48}

In addition, the maximum tip displacement and voltage also have the following relationship:⁵³

$$S = b_0 + b_1U + b_2U^2 + b_3U^3 + b_4U^4 \quad (6)$$

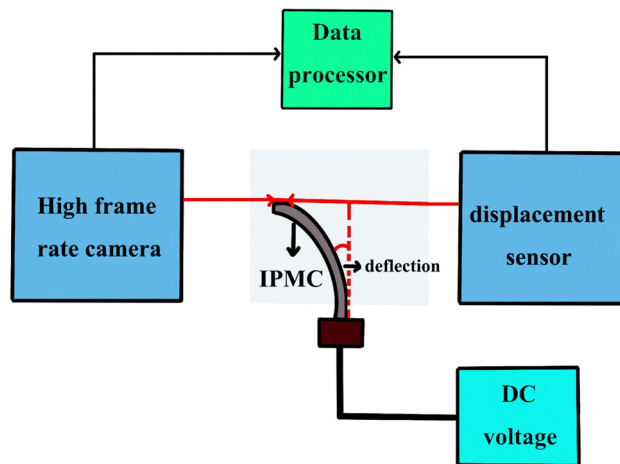


Fig. 6 Schematic diagrams of experimental setups used for measuring tip deflection and displacement.

where b_i ($i = 0, 1, 2, 3, 4$) is a constant. U and S are the input voltage and the maximum bending displacement of the IPMC, respectively.

5. New materials and properties of IPMC actuators

5.1. Modification of base membrane

The limitation of the IPMC actuators operated in the air using pure Nafion membranes is attributed to their poor hydrophilicity, which restricts their potential applications.⁴⁶ Researchers have explored various modifications on conventional commercial-base membranes by adding additives to improve output force and extend the stable actuation time of the IPMC actuators. For instance, Horiuchi Tetsuya *et al.*⁵⁴ developed a three-layer IPMC actuator that incorporates chemically modified carbon nanotubes (CNTs) into the Nafion layers. To enhance the performance of the composite material layers, the researchers inserted a pure Nafion layer between the outer layers of the CNT-Nafion composite layers. This resulted in a capacitance that was 2.2 times higher than that of traditional IPMC actuators. Inspired by this research work, Zhang *et al.*⁵⁵ used a template technique to integrate sulfonated graphene oxide (SGO) as a conductive material into a Nafion membrane using deposited Pt electrodes to create a modified membrane. The actuation tip displacement of this modified membrane was 1.7 and 1.9 times larger than that of the macroporous Nafion membrane-based IPMC (IPMC-M) when voltages of 6.2 V and 6.6 V were applied, respectively. Moreover, the IPMC-SGO-M exhibited a blocking force approximately 2.6 and 2.3 times higher than that of IPMC-M at the same applied voltages. These results emphasize the substantial influence of SGO modification on the performance of the actuators. Nah *et al.*⁵⁶ developed a new fibrous mat membrane made of a copolymer of polytetrafluoroethylene and perfluorovinyl ether sulfonic acid salt (Nafion). The researchers used the electrospin method to create Nafion fibrous mats. Many features of this newly created IPMC, such as the morphology, ionic conductivity, and actuation behavior were



compared to those of the conventional Nafion films. The fibrous mats demonstrated a dramatically high PC value, approximately three times higher than that of traditional similar-sized thin films. Meanwhile, the fiber mat-based actuators exhibited a 52% faster response rate compared to Nafion film-based actuators. Based on the above research finding, Palmre, Leila Naji, *et al.*^{57,58} conducted studies to modify Nafion-Pt IPMCs by using Pd-Pt electrodes and creating Nafion hybrid membranes to enhance IEC. The modified Nafion demonstrated a 52% increase in IEC and a 64.3% increase in WU. Furthermore, it exhibited an eight-time increase in tip displacement compared to pure Nafion films. However, the increase in tip displacement resulted in the permanent deformation of IPMCs, emphasizing the necessity of enhancing the anti-deformation capability of IPMCs. Therefore, Liang Chang *et al.*⁵⁹ developed a serrated structure on the IPMC actuator and optimized the size ratio of the serrations using numerical simulation methods. This study found that the IPMC with a 2:3 serration size ratio exhibited a 70% increase in blocking force and at least a 360% increase in tip deflection.

5.2. IPMCs based on new composite materials

The commonly used ion exchange membranes in IPMCs are the Nafion[®], Aciplex[®], and Flemion[®] series. These membranes have favorable properties such as high resistance to chemical degradation and extensive industrial implementation. However, the use of IPMCs using conventional ion exchange membranes is hindered by various constraints, including high production expenses, inadequate efficacy, and limited operational duration, which impede the widespread utilization of IPMC actuators.

5.2.1. IPMCs based on novel base membrane materials.

Although the Nafion membrane has high mechanical strength and chemical stability, its high cost limits its large-scale production and widespread application as an IPMC actuator. Moreover, its overall performance as a durable actuator, in particular, its blocking force, still needs to be improved. Therefore, it is essential to develop a new generation of IPMCs that are not only low-cost but also offer superior overall performance including prolonged working cycles, blocking force, and being eco-friendly.⁶⁰

The study conducted by Mohammad Luqman *et al.*⁶¹ examined the driving performance of sPS-IPMCs. The researchers found that an sPS-IPMC had a higher current density of 810 mA cm⁻² compared to Nafion's 456 mA cm⁻². Additionally, the sPS-IPMC had a larger tip displacement of approximately 44 mm, while Nafion had a tip displacement of only 23 mm. The response rate of the sPS-IPMC was around 10.3 mm within 3 seconds, which was higher than that of Nafion (2.9 mm). Finally, the blocking force of the sPS-IPMC was roughly 2.76 gf, which was higher than that of Nafion (1.51 gf). The findings suggested that the sPS-IPMC provides a better actuation performance than the Nafion-based IPMC. Moreover, the use of new materials in IPMC production, as studied by Sideris and Ru,^{62,63} has resulted in a significant reduction in production costs and an improvement in performance parameters such as PC and WU. Despite their advantages, the output force of these

IPMCs is lower than that of the Nafion membrane. To overcome this limitation, Man Jae Ha *et al.*⁶⁴ developed a polystyrene sulfonic acid (PSSA) thin film that was coated with platinum electrodes *via* a redox reaction. The resulting IPMC demonstrated a displacement output that was several times greater than that of Nafion, and its response rate was also enhanced. Aifen Tian *et al.*⁶⁵ used the solution casting method to fabricate pure PFSA, GO/PFSA, and SiO₂-GO/PFSA membranes, which were coated with Pt electrodes. The study showed that the addition of SiO₂-GO significantly improved the WUP and IEC of the PFSA membrane, leading to an amplified displacement by 3.2 times and a doubled blocking force. Mohammad Luqman *et al.*⁶⁶ synthesized a cost-effective Kraton (KR)-copper nanoparticle (CuNP) composite membrane, which was subsequently coated with platinum. The prepared IPMCs were then analyzed for their IEC, PC, WU, and WL. The results showed that sulfonated polyvinyl alcohol/alumina/graphene/Pt (SPVA-Al-GR-Pt) composite membranes could be a cost-effective substitute for commercial perfluorinated polymer-based IPMC actuators. Ajahar Khan *et al.*⁶⁷ successfully developed IPMC actuators that exhibit high IEC and WU. This breakthrough development of novel material-based IPMC actuators is expected to replace traditional Nafion polymer-based IPMC actuators in the future.

Recently, Mohammad Luqman *et al.*^{68,69} studied the preparation of various membranes, including SPEEK-PANI-Pt, sulfonated polyvinyl chloride (SPVC)-phosphotungstic acid (PTA), and SA/SPVA-PANI-Pt composite cation exchange membranes. Pt was used as the electrodes. The SPEEK-PANI-Pt membrane demonstrated superior IEC and response rate while maintaining a good PC compared to the Nafion membrane. Likewise, the SPVC-PTA film demonstrated superior characteristics compared to Nafion film, including higher WU, lower WL, and improved PC and IEC⁷⁰. Besides, the SA/SPVA-PANI-Pt composite cation exchange membrane exhibited comparable performance to the Nafion membrane. Tang Y. *et al.*⁷¹ developed a series of sulfonated polysulfone (SPSU) membranes. The resulting actuator demonstrated that SPSU membranes possess a higher ion exchange capacity and water absorption than commercial Nafion 117 ion exchange membranes, and IPMC actuators made from SPSU films exhibit significant bending strain, rapid response, and excellent fatigue resistance and the electro-mechanical behavior of SPSU ion exchange membranes can be controlled by adjusting their sulfuration degree. Mohd Imran Ahamed *et al.*⁷² fabricated a composite ion exchange membrane of poly(styrene sulfonic acid)-zirconium phosphate (PEDOT:PSS-ZrP) using the solution casting method. The membrane exhibited good displacement, superior WU capabilities, and reduced WL rate.

Table 1 summarizes the primary performance benefits and characteristics of novel membrane materials compared to conventional Nafion in recent years. Research and development on new membrane materials can significantly improve the output force and response rate of IPMCs. Additionally, using various operational media and waterproof coatings has the potential to extend the operation time of the IPMC actuators.



Table 1 Some of the composite materials as new base membrane materials of the IPMC actuators developed in recent years and their key performance benefits and characteristics

Ref.	Base membrane materials-electrode material	Key performance benefits	Other characteristics	Authors
24	MCNT-Nafion-Pt	Increase in tip displacement, blocking force, response rate, and stability	Increase in E and hardness by 3.3 and 4.4 times, respectively	Xiaojun Zhang, <i>et al.</i>
30	PAA-Cu	Improvement in PC and mechanical properties	Larger tip displacement	Yuyang Wang, <i>et al.</i>
46	SH-SiO ₂ -Nafion-Pt	Increase in displacement by 1.51–3.33 times	Increase in working hours by 4.46 times	Yubing Han, <i>et al.</i>
47	ThP-PANI-Pt	Higher WU and PC	Cost-effective and environmentally friendly	Ajahir Khan, <i>et al.</i>
48	SPEES-SWNT-Pt	Higher IEC, PC, and current density	Large tip displacement	Khan Ajahir, <i>et al.</i>
51	PPy-Al ₂ O ₃ -Nafion-Pt	Increase in displacement by 2.2 times	Increase in tip displacement, response rate, and blocking force	Jang-Woo Lee, <i>et al.</i>
52	Nafion-SiO ₂ -Pt	Increase in tip displacement by 2 times	Increase in blocking force by nearly 3 times	Vinh Khanh Nguyen, <i>et al.</i>
54	CNT-Nafion-Au	Increase in displacement by 1.3–2.3 times	Increase in capacitance by 2.2 times	Horituchi Tetsuya, <i>et al.</i>
58	SGO-Nafion-Pt	Increase in tip displacement by 8 times, 52% and 64.3% increase in IEC and WUP	Significant increase in capacitance and PC	Leila Naji, <i>et al.</i>
61	sPS-Nafion-Pt	Tip displacement reaches 45.1 mm, blocking force reaches 26.5 mN	Significant reduction in reverse relaxation phenomenon	Mohammad Luqman, <i>et al.</i>
65	GO-Nafion-Pt	Increase in tip displacement and blocking by 3.2 times and 2 times	Significant improvement in WU and IEC	Aifen Tian, <i>et al.</i>
66	SiO ₂ -GO-Nafion-Pt	Increase in PC, IEC, and WUC	Good stability	Mohammad Luqman, <i>et al.</i>
69	Kraton-Pt	Increase in PC, WUC, and IEC	Good stability	Mohammad Luqman, <i>et al.</i>
70	SA-SPVA-PANI-Pt	Performance comparable to Nafion	Good WU and IEC	Mohammad Luqman, <i>et al.</i>

5.2.2. IPMCs based on electrode materials. The performance of an IPMC depends not only on the development and modification of the base membrane material but also on the selection of electrode material. However, precious metals such as Pt and Au widely used as electrode materials significantly increase the manufacturing costs and restrict their applications. Therefore, many researchers have been devoted to discovering alternative and cost-effective electrode materials. Studies have indicated that the selection of electrode materials has a considerable influence on the electrochemical and mechanical characteristics of the IPMC actuators.^{73,74}

Electrode materials can be categorized into different groups, including metals, carbon nanotubes⁷⁵, graphene,⁷⁶ and conductive polymers,⁷⁷ Although metal electrodes have high conductivity and mechanical strength, they are susceptible to oxidation and corrosion. In contrast, carbon nanotube electrodes display outstanding chemical stability and corrosion resistance, but they suffer from low conductivity. In comparison, conductive polymer electrodes exhibit both good flexibility and high conductivity. However, their mechanical strength is roughly lower than that of metals and carbon nanotube electrodes.

KSK Chaitanya *et al.*⁷⁸ proposed a strategy to enhance the performance of Nafion-based IPMC actuators using carbon nanotubes and graphene as electrodes. The optimum performance of the IPMC actuator is obtained at a ratio of GO (7%) to CNT (6%). Inspired by this study, Zhou *et al.*⁷⁹ developed an innovative IPMC actuator based on a PVDF membrane coated with polypyrrole electrodes. This IPMC actuator permits large operating voltages and can operate continuously for up to two months.

Ajahir Khan *et al.*⁸⁰ developed a soft actuator that employs GO/Ag/Pani composite electrodes. The electrode material consists of a silver electrode coated with polyaniline. Compared to various conventional IPMC membranes, this novel IPMC actuator demonstrates notably higher values of WU, IEC, and PC. Moreover, Guo Dongjie *et al.*⁸¹ proposed a distinctive hydrophilic PVDF-based composite membrane composed of layered micro/nanostructures coated with flexible GR/PVDF electrodes. The base membrane displayed a porosity of 15.8% and a high WU of 44.2% was obtained due to that the three-dimensional interconnected channels inside the IPMC actuator promoted ion migration. As a result, the output force tripled, and the tip displacement nearly doubled when compared to Nafion membranes.

Table 2 summarizes the comparison between new electrode materials and traditional Nafion-based IPMC actuators using platinum electrodes. The selection of suitable electrode materials should correspond to specific requirements and the overall performance of the IPMC actuators.

6. Macroscopic and microscopic features of electrodes and their influence on IPMC

Dimension and configuration of surface electrodes coated on the base membrane are prominent macroscopic features that significantly affect the response rate and sensitivity of the IPMC

Table 2 New electrode materials developed for IPMC actuators in recent years and their key performance benefits and characteristics

Reference	Base membrane	Electrode material	Performance benefits	Other characteristics	Authors
26	SCNT–Nafion	Pd	Improved response rate and output force by about 3 times	Higher working density by 10 times	Jie Ru, <i>et al.</i>
27	Nafion	MWCNT	Increase in strain and stress	More than 20 000 cycles	Jie Ru, <i>et al.</i>
38	Nafion	Ni	Large tip displacement and output force	Good conductivity	M. Siripong, <i>et al.</i>
50	SPVA	PPy and Pt	High stability and IEC, but low WL and PC	Low cost and good flexibility	Inamuddin, <i>et al.</i>
54	CNT–Nafion	Au	Capacitance increase by 2.2 times	Tip displacement of 1.3–2.3 times that of conventional IPMC	Horiuchi Tetsuya, <i>et al.</i>
57	Nafion	Pd–Pt	Improved output force by more than 2 orders	Blocking force of 3.4 N, improved PC	Palmre, Kim, <i>et al.</i>
59	Nafion	Ag	Increase in blocking force and tip deflection by 70% and 360%, respectively	—	Xi Liang Chang, <i>et al.</i>
75	Nafion	Ag and nano-filler	Increase in WU	Increase in tip displacement	Yesaswi Ch Sridhar, <i>et al.</i>
79	PVDF	PPy	Increase in cycle-life	Withstand voltage increase	Dezhi Zhou, <i>et al.</i>
81	PVDF–PVP, PVDF	GR–PVDF	Increase in output force and tip displacement by 3 and 2.2 times, respectively	44.2% higher WU, 15.8% higher porosity	Dongjie Guo, <i>et al.</i>

actuators. Reducing the electrode area improves the response rate and sensitivity of the IPMC, but also decreases its ability to withstand voltage. Moreover, the electrode preparation process and surface treatment also influence the performance of the IPMC actuator.

6.1. Influence of macroscopic features

The mechanical properties of an IPMC actuator are influenced by various factors such as the thickness of the electrodes, surface roughness, and selection of the coating material. The thin thickness of the electrodes of an IPMC can break down due to a high loading voltage. Omiya Masaki *et al.*⁸² studied a hydrogen-driven IPMC actuator based on the water decomposition process. This study revealed that the thickness of the palladium (Pd) electrode significantly impacted the mechanical properties of the PVC–Pd actuator. In addition, as shown in Fig. 7, to address the issue of short stable working time caused by electrode fatigue and high water loss rate in IPMC, Guo Dongjie *et al.*⁹ proposed a simple and effective repair method, which includes electrochemical grafting of a conductive flexible polymer, poly(ethylene dioxythiophene) (PEDOT), on the IPMC electrode surface.

Yang *et al.*⁸³ investigated the microstructure of the IPMC electrodes by analyzing metal particle deposition and infiltration. They found that packing density, overlap ratio, electrode thickness, and particle radius significantly affect the electric resistance of IPMC. Appropriate control of these parameters enables design of the electrical properties of the IPMC actuator, resulting in its superior comprehensive performance. For example, Mohsen Shahinpoor⁸⁴ conducted research examining the influence of surface electrode resistance on the performance of IPMC artificial muscles. This study demonstrated that reducing surface electrode resistance could enhance the drive capability of the IPMC actuator. Moreover, Zhao Jintao *et al.*^{85,86} coated graphite and carbon nanotube electrodes onto platinum-plated IPMCs, leading to a maximum output tip displacement of 4.9 mm and a maximum output force of 39 mN. These values exceeded those of a typical Pt-electrode-based IPMC (3.18 mm and 31 mN), which verified that using this approach, not only was cost reduced, but also both maximum tip displacement and output force were increased.

6.2. Influence of microscopic features

Apart from the macroscopic features, the microstructure of the electrodes also influences the performance of the IPMC actuators. Aifen *et al.*⁸⁷ investigated the influence of dendritic interface electrodes (DIEs) on the microstructure and electromechanical response characteristics of IPMCs. Using chemical deposition, Jiahua *et al.*⁸⁸ created an IPMC with the composite electrodes of Pt and Cu to analyze the effects of DIEs on IPMCs. They evaluated the WUP, surface resistivity, and electromechanical driving performance, and determined the surface roughness of the electrodes using Matlab. These experiments demonstrated that introducing Cu²⁺ can repair cracked electrodes on the surface of the IPMC actuator, resulting in improved driving performance while reducing surface electrode resistance.



on the manufacturing process, electrode surface features, and actuation performance of Pt-electrode-based IPMCs to determine the suitable IPMC manufacturing process for driving bionic tulips. They identified the optimized numbers of IPMC sheets and driving voltage by measuring the flowering angle of tulips and the energy density of the IPMC strain. They established the groundwork for integrating IPMC actuators in bionic robotics through the successful blooming of bionic tulips. Inspired by this research, Jiahua Li *et al.*⁹² designed a Venus flytrap robot that closely imitates its biological counterpart. The Venus flytrap robot was mostly made of polydimethylsiloxane (PDMS) membrane-based IPMC actuators. The optimized performance of the soft robot was obtained with a length, width, and driving voltage of 3 cm, 1 cm, and 5.5 V, respectively, resulting in a very close biomimetic behavior to that of the real Venus flytrap.

7.2. Biomedical devices

IPMCs can be used in applications of medical devices such as artificial muscles and blood vessels to address the physical disabilities of human bodies. Moreover, the sensitive actuation and large displacement of the IPMC actuators permit the creation of medical devices such as the artificial cochlea, which can restore hearing for individuals with hearing loss. Eva A. Sideris *et al.*⁹⁴ have developed the first IPMC actuator for a linear peristaltic pump that is commonly used in pharmaceuticals, chemicals, food, healthcare, and blood handling. Additionally, Cheong *et al.*⁹⁵ reported a novel wirelessly powered IPMC soft actuator, whose driving mechanism is based on an external radio frequency (RF) magnetic field. The soft actuator is designed for precise targeted drug delivery. Wireless control of the actuator is achieved by embedding an LC resonant circuit in a 183 μm thick IPMC cantilever valve and adjusting the magnetic field frequency to a resonant frequency of approximately 25 MHz, and the experimental results show that wirelessly activated soft IPMC cantilever valves are a proof of concept for mobile, reliable and safe targeted drug delivery and can effectively reduce HeLa cell viability. As shown in Fig. 9, the application of IPMCs can be observed more intuitively.⁹⁵

Due to their bionic features,^{96,97} IPMCs have gained significant attention in robotics applications, such as urethral sphincter and esophageal swallowing devices. Xi Liang Chang *et al.* demonstrated the use of an IPMC actuator for radio frequency control in a water environment. They achieved activation of the actuator by tuning the frequency of the external magnetic field to match the resonant frequency (~ 13.6 MHz) of the receiver's antenna through magnetic resonance coupling. Additionally, they developed an IPMC-based actuator for drug reservoir devices that can wirelessly activate the opening of a fluid reservoir seal, thereby releasing the drug into the water environment.⁹⁸ Hosseini Sara Sadat *et al.*⁹⁹ introduced a nano-composite IPMC actuator that employs non-metallic composite material electrodes composed of bovine serum albumin (BSA) and microcrystalline cellulose (MCC) polymers and CNT nanofillers, in conjunction with conventional Au and Bt based IPMC electrodes. This IPMC actuator exhibits exceptional properties such as large displacement and low mechanical stress

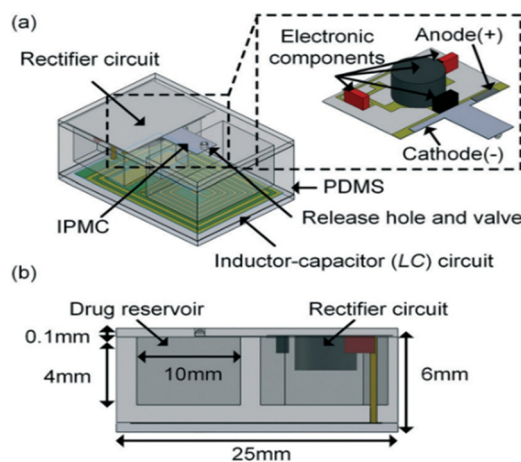


Fig. 9 IPMC-based cantilever valve for drug delivery. The body of the device is made of biocompatible polydimethylsiloxane (PDMS) and has external dimensions of 15 mm \times 25 mm \times 6 mm, including drug storage with a maximum loading capacity of 520 μL . (a) Schematic illustration of the drug delivery device; (b) side view of the drug delivery device. Reproduced with permission.⁹⁵ Copyright 2018 Royal Society of Chemistry.

damage, making it a promising candidate for restoring eyelid movement for patients with ptosis.

7.3. Underwater actuators

Due to the high flexibility, strong affinity for aqueous environments, low driving voltage (< 5 V), large displacement, miniaturization capabilities, and plasticity of IPMC materials, they have promising potential as a driving/motion device for underwater robots.¹⁰⁰ Researchers have reported experimental models of IPMCs for underwater robots, such as Gupta Ankur *et al.*¹⁰¹ and Safari Yousef *et al.*¹⁰² These models used IPMC artificial muscle bundles to achieve precise and continuous movement. Traver José Emilio *et al.*¹⁰³ analyzed the bending of IPMC segments in a swimming robot driven by artificial eukaryotic flagella (AEF) to investigate the generation of planar wave motion through distributed driving. He Qing-Song *et al.*¹⁰⁴ prepared a multi-layer Nafion structure with a continuous casting process. It can be used to drive robot fish, imitate gecko toe movement, drive false eyes, and many other parts. In addition, Li, Hongkai *et al.*¹⁰⁵ designed a capsule-like robot with pectoral and caudal fins made of IPMCs, as shown in Fig. 10. By analyzing the displacement response characteristics of square waves under different frequencies and low-level voltages, and using these characteristics to control the movement of capsule robotic fish, forward swimming, turning, and positioning movements have been achieved. IPMCs have the advantages of large bending displacement, low driving voltage, and flexibility, and have a wide range of applications in the field of underwater robots. However, challenges in durability, cost, and manufacturing technique remain to be solved. Therefore, when selecting materials, it is important to comprehensively consider specific application requirements and material characteristics to achieve the high performance and reliability of underwater robots.



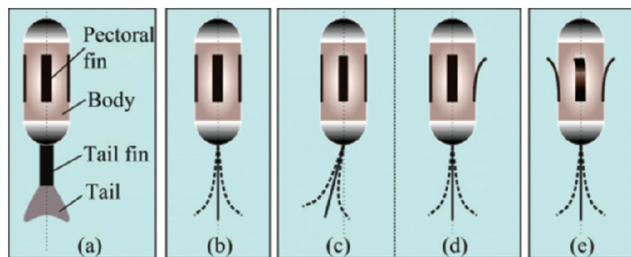


Fig. 10 The mechanical structure of a capsule-like robot and motion control principle. (a) Mechanical structure; (b) swim forward; (c) turning by asymmetric swing of caudal fin; (d) turning by opening of pectoral fin; (e) positioning. Reproduced with permission.¹⁰⁵ Copyright 2020 Springer.

8. Summary and outlook

8.1. Summary

This paper offers a comprehensive overview of IPMC actuators, covering their fundamental concepts such as deformation mechanisms, preparation processes, performance characteristics, and factors affecting their performance. It mainly focuses on the recent developments in flexible and composite materials of the IPMC actuators, highlighting the impact of electrode materials and macroscopic and microscopic features of the electrodes of IPMCs on their overall performance. Besides, it briefly provides hot application spots of the IPMC actuators. The article aims to present a clear and concise comprehension of the IPMC actuator, which is useful for more researchers and practitioners working and discovering in this field.

IPMC actuators have several advantages, including their small size, light weight, low actuation voltage, large displacement, simple actuation configuration, good biocompatibility, and environmental friendliness, resulting in their broad application prospects in the fields of biomedical science, robotics technology, and intelligent materials.

8.2. Outlook

In the past two decades, significant progress has been made in various aspects of IPMC materials; however, the ultimate industrial and commercial application objectives remain elusive due to persistent challenges in IPMC research. These challenges include several key aspects.

Firstly, regarding preparation, current IPMC driving materials suffer from two major limitations: low force output and short effective actuation time. Recent research efforts have concentrated on addressing these shortcomings through enhancements such as substrate film improvement, surface treatment, electrode optimization, and the exploration of new electroactive polymers, resulting in notable advancements in the driving performance of IPMCs. Nevertheless, these advancements need to be further improved. For instance, the integration of carbon nanotubes into the matrix film and the use of chemical oxidation for surface functionalization unavoidably introduce structural defects on the carbon nanotube surface, while the utilization of gold electrodes further increases the preparation cost, hindering mass production feasibility. Consequently,

future research endeavors may focus on mitigating structural defects, possibly through the addition of surfactants.

Secondly, in terms of electrodes, although current metal electrodes exhibit high conductivity and dendritic structures conducive to large interface areas, substrate penetration, and high capacitance, repeated driving of IPMCs leads to the formation of numerous cracks, resulting in leakage of working media. Additionally, prolonged exposure causes nano platinum metal particles to oxidize, diminishing electrode stability and substantially reducing IPMC driving efficiency. Hence, future research should prioritize rectifying existing electrode deficiencies and exploring novel electrode types to simultaneously reduce costs and enhance performance.

Lastly, concerning the preparation process, many methods are available for IPMC fabrication, each with its advantages and drawbacks. Thus, the optimal preparation method should be selected based on factors such as IPMC performance, preparation cost, complexity, and application environment, with ongoing optimization and adjustment throughout the experimental phase to meet performance requirements. For instance, a combination of solution pouring and hot pressing techniques can enhance the performance of the IPMC actuators. Future research should aim to optimize overall IPMC performance, with continuous exploration of IPMC actuators based on newly developed composite materials committed to promoting their applications in different fields.

Conflicts of interest

There are no conflicts to declare.

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