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A comprehensive survey of ionic polymer metal composite transducers: preparation, performance optimization and applications

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Abstract
Ionic polymer metal composite (IPMC) transducers, as one of the typical electroactive polymers with excellent electromechanical coupling properties, have tremendous potential to achieve high-performance actuators and sensors for flexible electronic and soft robotics. In this survey, after briefly describing the energy conversion mechanism of IPMC, we divided the history of IPMC into three stages based on the published papers, and then introduced the preparation technologies of IPMC in detail, which mainly include the selection of ionomer membrane and formation of electrodes. From the point of view of optimization, we summarized and analyzed the performance improvement methods of IPMC and the problems when it is used as actuators and sensors, respectively. The latest and typical applications of IPMC are widely presented as actuators and sensors, such as actuation in robots, grippers, medical and wearable devices, underwater perception and energy harvesting. Moreover, the challenges and opportunities of IPMC were envisioned for future prosperity. This survey will provide an overall general outline for the categorization, mechanism, precursors, and preparation methods of IPMC, which is helpful in facilitating the rapid development and application of IPMC.

Keywords: IPMC, development, preparation, performance, optimization, applications

INTRODUCTION

Polymer-based smart materials not only have excellent flexibility but also can respond or perceive external stimuli[1-5], such as electricity, magnetism, light, mechanic, humidity, and so on[6-15], which have shown remarkable potential in soft robots, medical engineering, aerospace, deep-sea exploration and other fields[16-20]. With the development of material science and bionics, various polymer-based smart materials have been developed, including shape memory polymers (SMP)[21], dielectric elastomers (DE)[22], ionic polymer metal composites (IPMC)[23], *etc.* Due to the reversible energy conversion from electrical to mechanical, IPMC immediately attracted vast attention and has been considered as one of the most promising smart materials since proposed by Shahinpoor *et al.* in 1992, which has been widely studied and applied as actuators as well as sensors in fields of bionic robots, medical and wearable devices[24-28].

As shown in Figure 1, a typical IPMC is composed of an ionic polymer interlayer and two conductive electrode layers to form a sandwich structure. The interlayer mainly plays the role of ion transport medium, while the electrode layers are conductive materials acting as conductors of electrons[29]. When a voltage is applied, an electrostatic field will be generated between the electrodes that can be regarded as parallel plate capacitors. In the presence of the electrostatic field, the movable cations inside the ionomer will migrate from anode to cathode. Then the uneven stress distribution along the thickness direction will lead to IPMC bending toward the anode side[30]. On the contrary, when IPMC deforms under the action of external loads, the anisotropic strain in the ionomer forces movable ions to migrate to the direction with lower stress, and thus output voltage signals can be measured on both sides of IPMC[28].

According to the number of published papers related to IPMC [Figure 2] since 1992 (the generally accepted starting point of IPMC), we divided the history of IPMC research and development (R&D) into three stages, namely the root stage (1992-2004), the trunk stage (2005-2016) and the branch stage (2017 up to now). In the root stage, the R&D was mainly focused on the exploration of actuating and sensing mechanisms, together with the development of IPMC preparation methods, such as impregnation-reduction, sputtering, solution casting, and so on[31]. Although there had been several preparation methods developed up until 2005, IPMC still had small deformation and blocking force, and response relaxation when exposed to air. Therefore, mainly focusing on the parameters of preparation process, more researchers were involved in the performance optimization of IPMC from multiple aspects in the trunk stage, such as membrane type and size, electrode morphology of surface and interface, movable ion type, water uptake and thickness adjustment, *etc.*[32-35]. On the basis of optimized preparation methods, IPMC was investigated to use various actuators and sensors since 2017, mainly in fields of bionic robots, medical and wearable devices. However, in recent years, the applications of IPMC are still stuck in the laboratory and have not been extended to the commercial level, which is totally determined by the inability of large-scale preparation, low energy exchange efficiency and restricted performance of IPMC.

In order to clarify the dilemma faced by IPMC in the current stage, understand the factors that restrict its broad application, and provide suggestions and guidance for its development in the next stage, we performed a comprehensive survey on IPMC from the aspects of preparation, performance optimization and application, as shown in Figure 1. The remainder of this survey is organized as follows: Section "PREPARATION OF IPMC" introduces different preparation methods of IPMC, including the selection of ionomer and the formation of electrodes. In Section "PERFORMANCE OPTIMIZATION OF IPMC", the performance and working mechanisms of IPMC as an actuator and sensor are analyzed, and the corresponding optimization methods are concluded. Section "APPLICATIONS OF IPMC"reviews the latest applications of IPMC as actuators and sensors. Finally, some challenges and prospects for the development and application of IPMC are put forward.

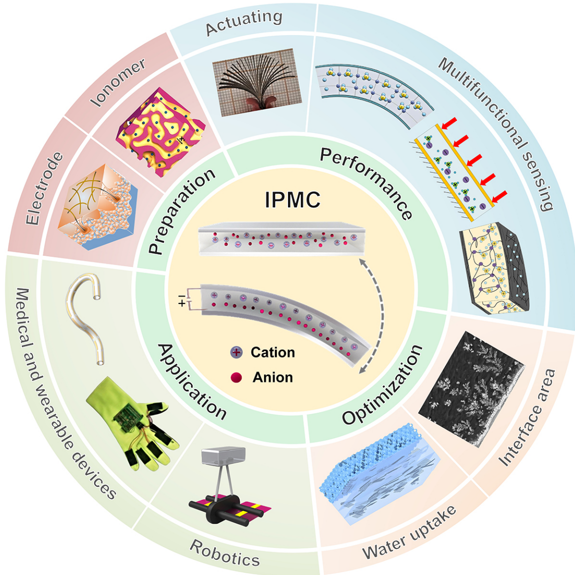

Figure 1. Schematic illustration showing the frame of this review. Mechanism of IPMC. Reproduced with permission from Ref.[44]. Copyright© 2021 The Korean Society of Industrial and Engineering Chemistry. Electrode. Reproduced with permission from Ref.[73]. Copyright© 2022 American Chemical Society. Ionomer. Reproduced with permission from Ref.[50]. Copyright© 2013 The Royal Society of Chemistry. Actuating. Reproduced with permission from Ref.[117]. Copyright© 2019 Elsevier B.V. Pressure sensing. Reproduced with permission from Ref.[137]. Copyright© 2022 IOP Publishing Ltd. Humidity sensing. Reproduced with permission from Ref.[32]. Copyright© 2021 Elsevier B.V. Water uptake. Reproduced with permission from Ref.[115]. Copyright© 2021 Elsevier B.V. Smart glove. Reproduced with permission from Ref.[189]. Copyright© 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. Robotics. Reproduced with permission from Ref.[194]. Copyright© 2015 IOP Publishing Ltd.

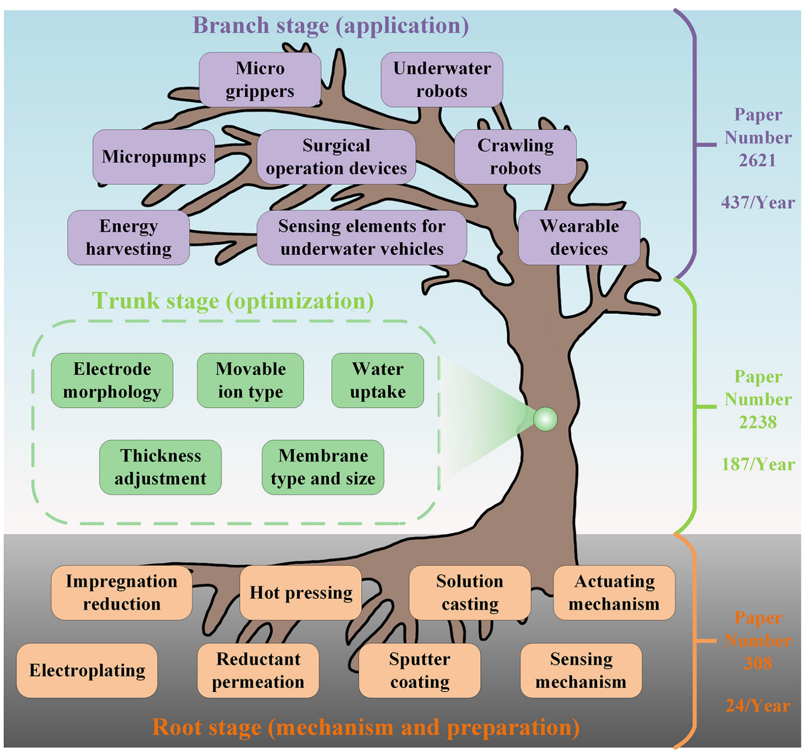

Figure 2. The development stage of IPMC in the past 30 years.

PREPARATION OF IPMC

A typical IPMC is composed of an ionic polymer matrix sandwiched with two conductive electrode layers. Therefore, the preparation of IPMC includes the selection of the ionomer and the formation of electrodes, both of which have a great influence on the performance of IPMC. This section mainly introduces the latest progress of IPMC in matrix membrane and electrode preparation process.

Selection of ionomer membrane

Commercial ionomer membranes

(1) Water based-Nafion

The selection of the ionomer is the first step of IPMC preparation, which has an important impact on the properties of IPMC, including the ion migration velocity and the overall elastic modulus. At present, the most commonly used matrix membrane for IPMC is perfluorinated sulfonic acid membrane (Nafion) because of its high ion capacity and stable chemical and mechanical performance[24]. The molecular formula and microstructure diagram of Nafion are shown in Figure 3A and B, respectively. The long carbon-fluorine chain forms the skeleton structure of Nafion, and the short side chain ends with a sulfonic acid group, which can bond with cations through ionic bonds and agglomerate to form nanochannels[36]. The water molecules contained in the nanochannel can dissociate the sulfonic acid group linked cations into hydrated cations and migrate in the nanochannel. Therefore, Nafion presents a solid-liquid two-phase structure at the micro-nano level. The solid phase serves as a skeleton to support the entire polymer structure, while the liquid phase accommodates free ions and acts as a route for ion migration[37]. The pure Nafion can absorb different solvents to form the liquid phase, by which the water phase is commonly used. In addition, the cation types can be exchanged into the Nafion by soaking different alkali solutions, including Li^+, Na^+, K^+, *etc*.[38-40]. When a voltage is applied, the cations inside clusters will migrate to the cathode side under the action of electrostatic forces, resulting in a sharp increase in the stress and ion concentration of the cathode. Then under the action of elastic and osmotic forces, the cation clusters will diffuse toward the anode again; meanwhile, the deformation of the IPMC will relax, which is known as the back relaxation phenomenon[41].

(2) Ionic liquid (IL)-Nafion

Due to the low viscous force of water, the back relaxation phenomenon of water-based Nafion is particularly obvious. Some researchers proposed to use IL with higher viscous force as the internal solvent of Nafion. Lee *et al.* studied the performance of IPMC with four different ILs as solvents, and showed that the performance of IL-IPMC mainly depends on the size of anions and ion mobility in ILs[42]. Due to the volatile property and low electrolytic voltage of water, it will constantly evaporate and be electrolyzed during the operation of water-IPMC, resulting in performance degradation over time[43]. On the contrary, ILs are not volatile as solvent, which can greatly improve the long-term working stability and compressive resistance of IPMC, as shown in Figure 3C[44]. However, IL-IPMC also has some disadvantages. For example, due to the high viscous force of ionic liquid, the resistance of ion migration is larger, which prolongs the response time of IPMC and even reduces the blocking force[45]. In order to solve the problem of long response time of IL-IPMC, Safari *et al.* tried to exchange lithium ions to ILs, aiming to enhance the ionic conductivity, capacitance characteristics, tip displacement and response speed[46]. The addition of lithium ions made the tip displacement of IPMC increase by 26 times and decreased the actuating attenuation with time. Wang *et al.* used methylammonium formate, a special IL with low viscosity, small ion size, high ionic conductivity and excellent electrochemical stability, as the solvent of IPMC, enabling it to achieve greater displacement, blocking force and response speed[44].

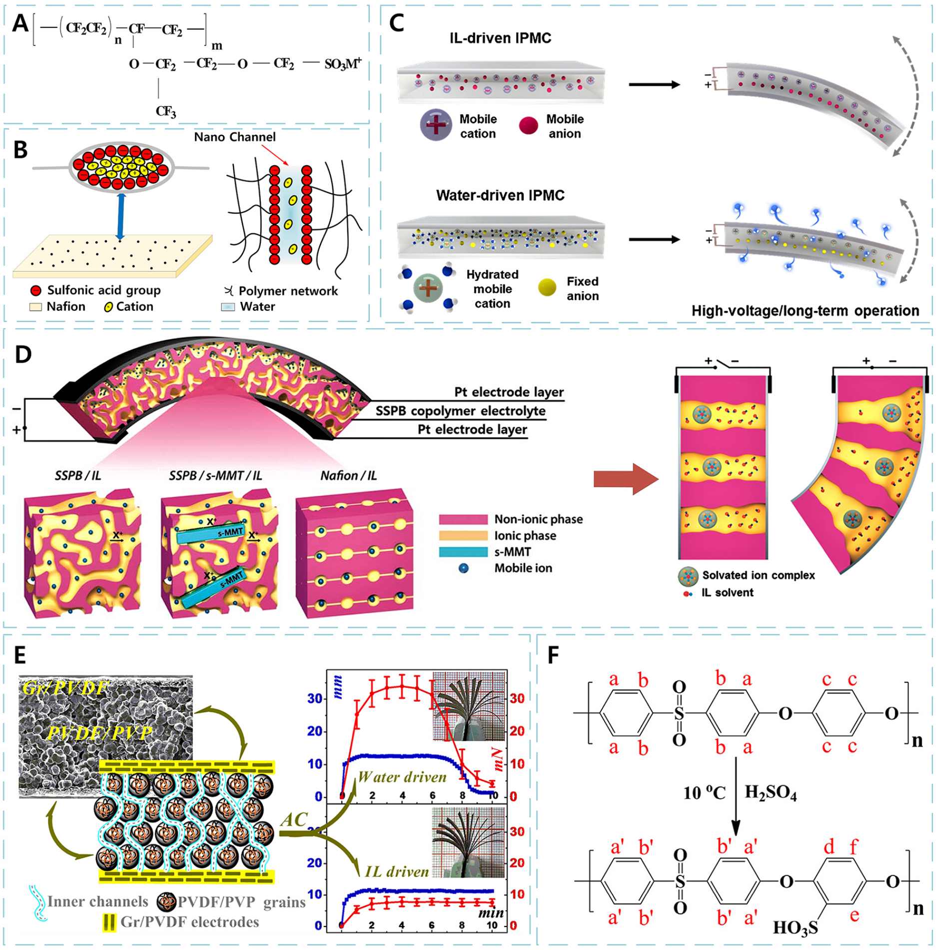

Figure 3. Ionomer of IPMC. (A) Chemical formula of Nafion. (B) Schematic diagram of Nafion microstructure. (C) Working performance of IL-driven IPMC and water-driven IPMC. Reproduced with permission from Ref.[44]. Copyright© 2021 The Korean Society of Industrial and Engineering Chemistry. (D). SSPB-based IPMC actuator. Reproduced with permission from Ref.[50]. Copyright© 2013 The Royal Society of Chemistry. (E) PVDF-based IPMC actuator. Reproduced with permission from Ref.[56]. Copyright© 2019 American Chemical Society. (F) Sulfonation reaction on PEES. Reproduced with permission from Ref.[57]. Copyright© 2015 Elsevier Ltd.

(3) Other commercial ionomers

In addition to Nafion, there are some other commercial perfluorinated olefin polymers that have been used as matrix membranes for IPMC, including perfluorinated carboxylic acid membrane Flemion[47] and Aciplex[48], perfluorinated sulfonate membrane Selemion[49]. For instance, Zhu *et al.* used Flemion as the matrix membrane to prepare an IPMC and tested its electromechanical response[47]. Compared with Nafion-IPMC, Flemion-IPMC has smaller relaxation deformation. Besides, the deformation rate of the IPMC prepared by the perfluorinated carboxylic acid film is slower, which is caused by the slow and continuous secondary dissociation of the weak acid group -COOH on the anode. The synthesis of perfluorinated carboxylic acid membranes is more complex compared with Nafion, which makes Nafion remain the most widely used commercial matrix film for IPMC preparation.

Novel ionomer membranes

Due to the drawbacks of commercial ionomers, such as high cost, small blocking force and back relaxation phenomenon, some researchers have tried to develop new ionic polymers for IPMC. Jo *et al.* summarized some novel ionic polymers developed prior to 2013 in Ref.[24], such as Sulfonated poly(ether ether ketone) (SPEEK), sulfonated poly(styrene-b-ethylene-co-butylene-b-styrene) (SSEBS), sulfonated poly(styrene-ran-ethylene) copolymer (SPSE), and sulfonated poly(arylenethioethersulfone) copolymer(SPTES). Therefore, in this review, we mainly describe the new ionic polymers after 2013, including sulfonated styrenic pentablock copolymer ionomer (SSPB), sulfonated polyvinyl alcohol (SPVA), poly(vinylidene fluoride) (PVDF), and so on.

(1) SSPB-based ionomer

SSPB is a kind of pentablock copolymer, which has been regarded as an alternative for Nafion in recent years. It can produce well-defined microphase-separated ionic domains on the scale of several tens of nanometers, which could work as more effective ion conduction channels than very small ionic clusters of Nafion on the scale of a few nanometers. Lee *et al*. prepared an SSPB/s-MMT-IL IPMC actuator with IL as the internal solvent, as shown in Figure 3D[50]. In detail, they added sulfonated montmorillonite (s-MMT) into SSPB copolymer dispersion, and the dispersion was cast onto a glass plate to obtain an SSPB/s-MMT membrane. Compared with Nafion-IL IPMC, it exhibits larger bending deformation and higher response speed, and also avoids the defect of back relaxation, because the sheet-like morphology of the inserted s-MMT particles promotes ion migration across adjacent ion channels. Subsequently, they introduced composite fillers with higher sulfonation degrees and aspect ratios to further improve the actuating performance. A novel SSPB/sGO-IL IPMC actuator was fabricated by embedding sulfonated graphene oxide (sGO) as a highly ionic conductive activated carbon filler in SSPB membrane, which has a better performance in bending strain, initial strain rate and charge-specific displacement[51].

(2) SPVA based ionomer

As a non-perfluorinated and sulfonated polymer, SPVA has various advantages, such as good forming capability, short processing time, low cost of fabrication, high thermo-mechanical stability, and excellent exchange and water-holding capacities. Inamuddin *et al*. prepared an SPVA/Py-Pt IPMC by casting the mixture of Polypyrrole (Py) and SPVA at 45 °C[52], which has high water-holding capacity under high applied voltage. The existence of Py coating with good water absorption capacity can prevent water molecules from leaking through cracks, so that the longest working time of the actuator at 5 V can reach 16 min. Besides, they also prepared an SPVA/PANI IPMC by replacing Py with polyaniline (PANI). The good conductivity and high redox cyclability of PANI not only improved the bending rate and displacement, but also minimized the back-relaxation phenomenon[53]. On the basis of SPVA/PANI copolymer, Luqman *et al*. further introduced silicotungstic acid (SA) to prepare a terpolymer, which enhanced the electrical conductivity and actuation ability of IPMC[54].

(3) PVDF base ionomer

Poly (vinylidene fluoride) (PVDF) has emerged as a promising candidate to replace Nafion, considering its excellent thermal stability, low chemical resistance, and moderate mechanical strength. It is worth noting that PVDF is one of the piezoelectric materials. However, the inherent hydrophobicity makes it difficult to absorb the liquid electrolyte. In addition, PVDF does not exhibit ion-exchange capacity (IEC) due to the absence of ionic and carboxyl plasma exchange groups on its side chains. Doping with hydrophilic materials may provide an opportunity for it to serve as a polymer matrix of IPMC. Mehraeen *et al*. prepared a high-performance PVDF/poly (styrene sulfonic acid) (PSSA) IPMC using a simple and novel method of radiation grafting PSSA, a well-known hydrophilic conductive functional group, onto PVDF, the fabrication of which is rapid and simple compared with the traditional Nafion-IPMC[55]. Guo *et al.* developed a polar PVDF/PVP film with a highly porous structure as an enhanced internal channel that supports significant ion migration by casting the mixture of the hydrophilic polymer polyvinyl pyrrolidone (PVP), PVDF and IL with a mass ratio of 2:1:1[56]. By introducing PVP, the composite membrane can adsorb different solvents to prepare water-IPMC or IL-IPMC actuator, as shown in Figure 3E[56].

(4) Others

In addition to the aforementioned materials, some other ionic polymers also have been developed for the preparation of IPMC. For instance, Xue *et al.* obtained a sulfonated poly(ether ether sulfone) (SPEES) membrane with high ion exchange capacity (IEC) and water uptake (WU) through a simple sulfonation reaction on commercial poly(ether ether sulfone) (PEES) membrane, as shown in Figure 3F[57]. The performance of the prepared SPEES IPMC is comparable to that of Nafion IPMC except for lower fabrication cost. Guo *et al.* synthesized a sulfonated diphenyl fluorene blocked polysulfone (SDPF-b-PSU) by varying sulfonic concentration via block copolycondensation reaction between dichlorodiphenyl sulfone (DCDPS) and Bis(4-hydroxyphenyl) fluorene (BHPF)[58]. When applying a voltage of 4 V, the blocking force of the prepared SDPF/PSU IPMC is 11 times that of the Nafion-IPMC under the same size. Besides, thorium (IV) phosphate (ThP), sulfonated poly(arylene ether ketone) (SPAEK), sulfonated polyether ether ketone (SPEEK), sulfonated polyvinyl chloride (SPVC) Kraton, *etc.*[59-64] are also candidates to substitute Nafion.

The performances of IPMC prepared by different ionic polymers are summarized and compared in Table 1. The strain ε can be expressed as $\varepsilon = 2ds/(s^2 + l^2)$, where l is the free length, d is the thickness and s is the tip displacement. It can be seen that IEC, WU and conductivity will all affect the performance of IPMC. At present, the development of ionomer membranes has achieved good results in terms of performance, and the research direction in the next stage should be focused on the mass preparation of new ionomer films commercially.

Formation of electrodes

The characteristics of the electrode, such as the conductivity, thickness, type and interface area, all have important effects on the performance of IPMC. Metal electrode materials, such as palladium, platinum, gold, *etc.*, were usually used to form electrodes for IPMC because of their stable electrochemical properties. Some researchers also tried to use non-metallic electrodes to replace metal electrodes for the purpose of improving the performance of IPMC, but there is no substantial advantage except for reducing the preparation price. At present, the electrode formation methods of IPMC can be divided into physical methods and chemical methods.

Physical methods

The physical method, also known as mechanical plating method, is the most direct and fast way to prepare electrodes for IPMC, which is cheap and convenient for large-scale preparation. The electrodes of IPMC prepared by physical method usually show high uniformity. At present, the widely used physical methods mainly include physical vapor deposition, solution casting, hot-pressing, direct assembly process, *etc.*

(1) Physical vapor deposition

Physical vapor deposition (PVD) refers to a technology that vaporizes the material source surface into gaseous atoms or molecules (or partially ionizes into ions) under vacuum conditions, and deposits a film

Table 1. Performance comparison of IPMC prepared by different ionic polymers

Ionomer	IEC (mMol/g)	WU (wt%)	Conductivity (mS/cm)	Maximum bearing voltage (V)	Maximum strain (%)	Maximum blocking force (mN)	Reference
Nafion	0.91	22	23	2.5	0.84	5	[23,64]
SSPB/S-MMT	1.5	150	42	3	0.8	3	[50,64]
SSPB/SGO		160	60	2	0.88		[51]
SPVA/Py	1.2	82.3	1.6	5.25	0.31	1.864	[52]
SPVA/PANI	1.6	425	1.7	5.25	0.52	0.08	[53]
SA/SPVA/PANI	1.6	245	1.9	3	0.36		[54]
PVDF/PSSA	1	62	85	4	0.33		[55]
PVDF/PVP	0.8	44.2	0.8	15	1.68	33.33	[56]
SPEES	2.1	58	65	3	0.40		[57]
SDPF	2.62	42.9	27	4	0.66	1.3	[58]
Thp/PANI	1.6	95.4	1.12	4.2	0.27	2.02	[59]
SPEEK/PANI	1.98	49	1.97	2	0.22	0.31	[61]
SPVC/PTA	1.98	57	1.6	4	0.42	0.242	[62]
Kraton	2.1	71.05	1.93	4	0.38	0.343	[63]

with a particular function on the substrate surface by low-pressure gas (or plasma). According to different working mechanisms, PVD can be classified as evaporation coating, sputter coating and ion coating[65], among which sputter coating is most commonly used in electrode formation of IPMC. Gudarzi *et al*. fabricated an IPMC sensor by sputtering a layer of gold electrodes on the surface of Nafion 117[66]. In order to improve electrode adhesion, sputter coating is usually combined with surface roughing techniques like plasma etching. For instance, Fu *et al*. developed an automatic equipment for large-scale preparation of IPMC by combining *in situ* plasma etching and magnetron sputtering technology[67]. Through parameter editing, the equipment can prepare IPMC sensors with silver electrode of different shapes. After plasma treatment on the matrix surface, Esmaeli *et al*. sputtered a layer of titanium or chromium between the gold electrode and Nafion matrix [Figure 4A], which further improved the adhesion between the electrode and the ionomer[68].

Sputter coating provides a controllable way for electrode formation of IPMC, as well as reducing the manufacturing time to less than 2 h (usually 12 h in chemical methods)[67]. Besides, this method can deposit a variety of different metals on the surface of substrate film, and the thickness of electrode is controllable and the uniformity is good. However, electrodes generated by sputter coating generally have poor adhesion and are prone to cracking or even peeling when used. Mousavi *et al*. compared the performance of IPMC actuators and found that the tip displacement of IPMC through sputtering was smaller than that by electroless plating[69]. In addition, obvious electrode cracking was observed in the experiment, which was caused by the poor adhesion between electrode layer and ionomer matrix[69].

(2) Solution casting

Solution casting is to put the ionic polymer solution into the mold and make it volatile and solidify by heating to get a certain shape of the film, which is generally used in the preparation of matrix film[70]. This section mainly discusses the research of solution casting methods used in electrode formation for IPMC. Generally, conductive particles are mixed with ionic polymer solution firstly, and then poured onto the surface of ionic polymer matrix film. After volatilization and curing, a layer of electrode will be formed. Chung *et al*. obtained electrodes with a thickness of 15 μm by dispersing silver nanoparticles into Nafion

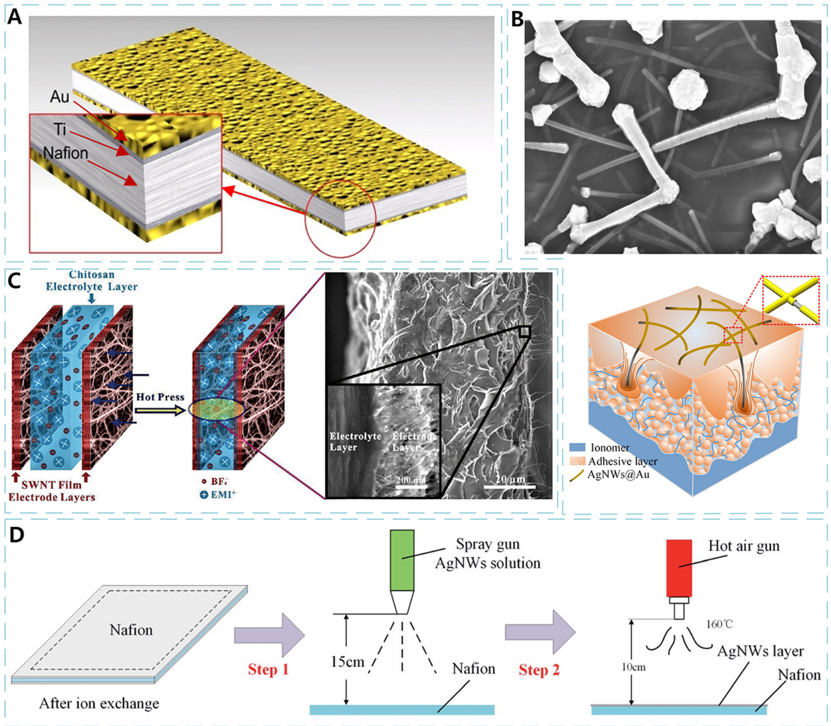

Figure 4. Physical methods of electrode formation. (A) IPMC sputtered with titanium or chromium between Au electrodes and the matrix. Reproduced with permission from Ref.[68]. Copyright© 2017 Elsevier B.V. (B) Electrodes with biological hair structure prepared by solution casting. Reproduced with permission from Ref.[73]. Copyright© 2022 American Chemical Society. (C) A novel actuator prepared by hot-pressing. Reproduced with permission from Ref.[75]. Copyright© 2011 American Chemical Society. (D) Method of spraying the silver nanowire solution on the surface of Nafion. Reproduced with permission from Ref.[35]. Copyright© 2020 The Royal Society of Chemistry.

solution and casting it on Nafion film[71]. It is worth noting that in order to improve the adhesion between the ionomer membrane and the electrode, the solution used to cast the electrode is generally the same type as the matrix. Khan *et al.* prepared a high-performance Kraton IPMC actuator by dispersing silver nanoparticles in Kraton solution and then pouring it onto Kraton matrix film[72]. The same approach was successful after replacing silver nanoparticles with platinum[63]. Electrodes prepared by solution casting generally have high resistance because the conductive particles are partially coated by polymers. Hence electroplating or similar techniques are usually required to further reduce the surface resistance. Zhao *et al.* sprayed a layer of Nafion solution on the surface of Nafion membrane as the connecting layer after heating and curing, and then sprayed a layer of ethanol dispersion of silver nanowires on the surface[73]. The silver nanowires were partially embedded in the connecting layer (Nafion solution layer after curing) by the alcoholization of Nafion, similar to biological hair structure, as shown in Figure 4B. Finally, the surface resistance was reduced from 10 Ohms to 1.5 Ohms by electroplating.

The solution casting method has the advantages of simple operation, fast preparation and low cost. Due to the same properties of the matrix film and the casting solution, the strong chemical bond between the electrode layer and the matrix results in a strong connection between them. However, the thickness of the electrode layer is difficult to control as a result of the volume reduction caused by solution volatilization during curing. In addition, during the casting process, the solution will spontaneously level under the action of gravity, so the slight tilt of the matrix will lead to severe uneven electrode thickness.

(3) Hot-pressing

Hot-pressing is a method to obtain IPMC with a certain thickness quickly. Firstly, all layers are put into the mold according to the order of electrode-matrix-electrode. Then the mold is placed on the hot press platform and pressurized under heating conditions. The edge of the laminated structure will be fused under high temperature and then resolidified as a whole under pressure. Some scholars combined hot-pressing with surface plating by embedding silver nanoparticles in Nafion membrane to prepare the electrodes of IPMC[74]. Nevertheless, this stacking method seems to be more suitable for the formation of non-metallic electrodes. Li *et al*. prepared a novel actuator by hot-pressing two SWNT films as electrode layers together with a chitosan matrix at 70 °C and 150 N for 2 min, as shown in Figure 4C[75]. It can be seen from the electron microscopy figure that there is no gap between the electrode and the matrix. Ru *et al*. developed a new Ionic polymer actuator by hot-pressing the self-made multi-space carbon nanotube film onto Nafion/IL hybrid membrane at a pressure of 16 kg cm^{-2} and the temperature of 180 °C for 30 min, which showed a good actuating performance[76].

Hot-pressing method has simple operation and good repeatability, and it is easier to control the overall thickness of the actuator compared with solution casting. However, it is difficult to control the separate thickness of core layer and electrode layer after hot-pressing. Besides, due to the high temperature and pressure during the preparation, the prepared IPMC generally has a large residual stress that needs to be eliminated by means of heating and washing, which may also lead to some changes in the shape of the material.

(4) Direct assembly process

Direct assembly process (DAP) is to form the electrodes by means of coating or spraying the electrode particle dispersion solution directly on the polymer surface. In order to improve the adhesion between electrode and matrix, the surface electrode can be further treated by hot-pressing. Akle *et al*. dispersed the conductive particles in the polymer solution and directly applied them on the surface of the ionic polymer matrix film to form electrodes, and then hot-pressed them to further improve the adhesion between the electrode and the matrix film[77]. The same way using carbon materials also achieved good results[78]. When preparing IPMC with gradient shape, Zhu *et al*. directly applied conductive silver paste on the surface of Nafion and pasted a layer of copper foil on it, then an electrode layer with good conductivity was formed[79]. In addition to coating, spraying is also an effective DAP method. Wang *et al*. directly sprayed the silver nanowire solution on the surface of Nafion through a spray gun [Figure 4D], forming a very uniform electrode[35].

When using DAP method, electrode composition can be selected freely and electrode thickness can be controlled. However, the large manual error reduces its repeatability. Besides, because the electrode solution is simply smeared on the matrix film, the adhesion between the electrode and the matrix is poor. Simple clamp holding or small tangential force may lead to electrode peeling. Meanwhile, the matrix film will absorb the water in the electrode solution and swell, which will lead to the folding of the surface electrode after drying.

Chemical methods

High interface area is a guarantee of the excellent performance of IPMC. However, the electrode prepared by most physical methods can only be attached to the ionomer surface (called surface electrode), which is difficult to grow into the ionomer to form a valuable interface electrode. Although some physical methods such as hot-pressing can also form certain interface electrodes to some degree, it exhibits structural characteristics due to the uniform distribution of electrode particles. While electrodes formed by chemical methods generally grow from the ionomer surface to the interior and the growing process is gradual, so the interface electrode exhibits natural and non-structural characteristics. This gradient distribution of interfacial electrode will play a good buffer role in the ion migration process, so chemical methods are the most commonly used methods to prepare high-performance IPMC. Currently, the most commonly used chemical methods mainly include electroless plating and electroplating.

(1) Electroless plating

The general mechanism of electroless plating is to reduce metal cations by reducing agents and depositing them on the inner and surface of the ionomer film to form metal electrodes. Takenaka and Millet reported two different electroless plating methods, respectively, known as reductant permeation (RP)[80] and impregnation-reduction (IR)[81]. In a typical RP process, the reducing agent and metal ions solutions are placed on both sides of the ionomer membrane, and the reducing agent penetrates into the membrane from one side and reacts with the metal ions permeating from the other side. This method can effectively obtain electrode layers with different kinds of metals. However, it has not been used on a large scale because the operation of placing two different solutions on both sides of the membrane is not convenient[80].

Compared to RP, the operation of IR is more convenient, and it is easier to form firm and durable metal electrodes. The operation steps of a typical IR process are shown in Figure 5A. Wang *et al.* combined this method with electroplating to prepare an IPMC with a palladium-gold electrode[28]. Firstly, the ionomer membrane was pretreated by ultrasonic cleaning, hydrochloric acid washing and deionizing water washing. The purpose of this step was to clean the surface and interior of the substrate film. Then, the pretreated membrane was immersed in the $Pd(NH_3)_4Cl_2$ alkaline solution of 0.01 mol/L, and the Pd ammonia ions were fully transported into the matrix membrane by shaking in water. Subsequently, the soaked membrane was put into the $NaBH_4$ alkaline solution of 0.02 mol/L, and the internal palladium cations were reduced to palladium electrode on the membrane surface, as shown in Figure 5B. Finally, the prepared palladium IPMC was coated with gold to reduce the resistance. The IR method can also be used to prepare IPMCs with metal electrodes of other types, and platinum electrode was plated on a Nafion membrane by this method, as reported by Millet *et al.*[81]. Besides, Byun and Yang *et al.* achieved the preparation of Cu-IPMC[82] and Au-IPMC[83] through IR method, respectively.

Contrary to the IR method, Chung *et al.* proposed a method called reverse electroless plating (REP)[84], which is to soak the ionomer in reducing agent and solution with ions in order to form the electrode. The metal particles formed by the REP method have a smaller diameter and a more uniform distribution in the membrane, and the overall surface roughness of the electrode is smaller.

The electroless plating method can produce firm and durable electrodes, and the electrode layer penetrates into the substrate film, which greatly improves the actuating and sensing performance of the IPMC due to the capacitance enhancement. However, electroless plating is time-consuming, expensive and complicated. In the preparation process, the amount of each reagent needs to be carefully controlled; even extremely slight errors may lead to electrode defects; therefore, the thickness and uniformity of the prepared electrode cannot be accurately controlled. In addition, the type of metal will have a great influence on the electrode morphology; there are mainly three types of electrodes, namely dendritic interfacial electrode (DIE), particle interfacial electrode and layered interfacial electrode represented by gold, palladium, and platinum, respectively [Figures 5C-E][85-87]. Among them, the IPMC with Au electrode shows the best performance due

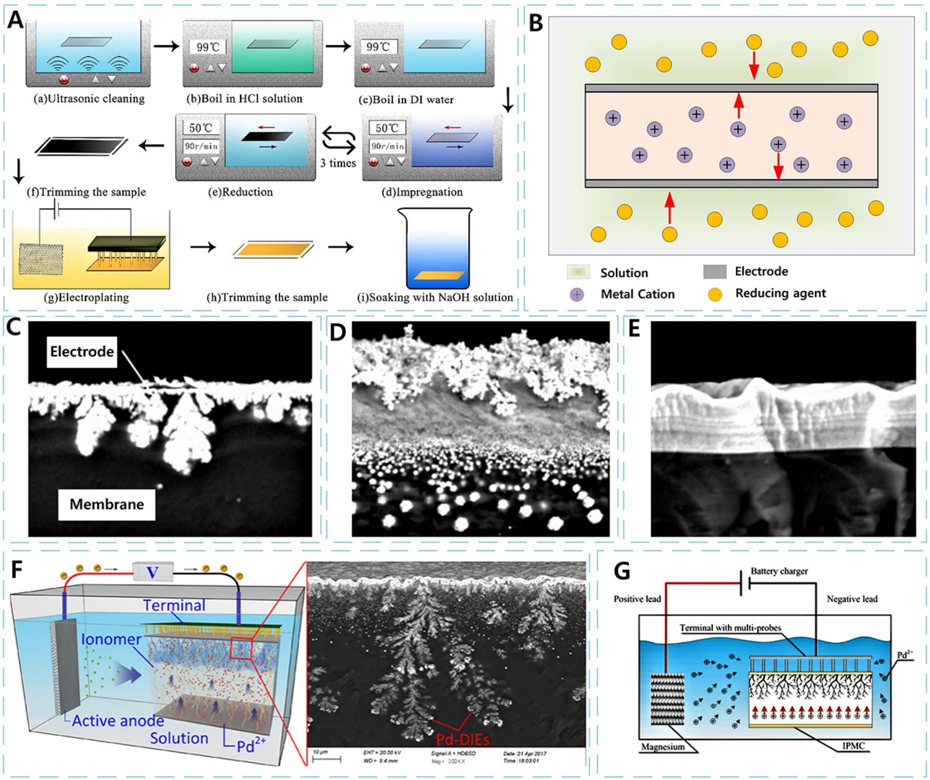

Figure 5. Chemical methods of electrode formation. (A) Typical process of IR. (B) Reduction of palladium cations to palladium electrode. (C) Dendritic interfacial electrode. (D) Particle interfacial electrode. (E) Layered interfacial electrode. (F) DIE formation through impregnation electroplating. Reproduced with permission from Ref.[34]. Copyright© 2017 American Chemical Society. (G) Set-up of electroplating. Reproduced with permission from Ref.[34]. Copyright© 2017 American Chemical Society.

to the high interface area of the DIE. However, the preparation of Au-DIE requires complex procedures and harsh conditions. In response to this problem, Wang *et al.* proposed a rapid, facile and efficient route to form the DIEs microstructure inside an ionomer, which was dominated by impregnation electroplating (IEP)[34]. Firstly, they used IR method to form an initial electrode layer on the surface of a Nafion film and immersed it in $Pd(NH_3)_4Cl_2$ solution sequentially before electroplating it at DC 5 V. Palladium ions migrated to the cathode inside the film and were reduced to form a DIE layer, as shown in Figure 5F. Formation of DIEs has also been successfully achieved in the validation of other metals (e.g., Pt, Ag) and other matrix films (e.g., Aciplex).

(2) Electroplating

Apart from obtaining DIE, electroplating is generally used for rapid reduction of surface resistance, the principle of which is to deposit metal ions on the membrane of electrolytic cell cathode by electrochemical reaction. Generally, the metal solution to be plated is used as the electrolytic cell, the anode of the power supply is connected with the titanium mesh and the cathode is connected with the ionomer membrane to be coated [Figure 5G][34]. The ions to be plated will be deposited on the surface of the ionomer cathode under the electrochemical action. Taking electroplating Au as an example, Au ions are reduced to form a layer of gold electrode at the cathode, while hydroxide ions are oxidized to oxygen and water on the surface of the titanium mesh at the anode.

Since electroplating requires an electrochemical reaction, it is necessary to deposit a layer of electrode on the outside of the ionomer membrane by other methods to make it conductive, so the electroplating method is usually used in conjunction with solution casting, hot-pressing, electroless plating, and so on[28,73,74].

The advantages and disadvantages of different electrode forming methods are concluded and listed in Table 2. All in all, the physical methods have a faster preparation speed, but the adhesion between the electrode layer and the matrix is poor. While the IPMC prepared by chemical method has better performance, the cycle time is long and the cost is expensive. Therefore, how to select the preparation process needs to be evaluated in combination with specific application scenarios.

PERFORMANCE OPTIMIZATION OF IPMC

Performance of IPMC as actuators

Effects of applied voltage

When used as an actuator, the deformation of IPMC generally increases with the increase of the applied voltage, and there is an approximate quadratic relationship between them. However, when the voltage is greater than 3 V, the water inside IPMC will be electrolyzed, which will affect the service life of IPMC, so the applied voltage is generally set as 0-3 V. Besides, the frequency of applied voltage also affects the deformation of IPMC. Jung *et al*. tested the tip displacement of IPMC at voltages of different frequencies and found that the tip displacement of IPMC only decreased with the increase of frequency[88]. Kobayashi *et al.* found a different rule that the tip displacement decreases from 0.1 Hz to 2 Hz and then increases from 2 Hz to 4 Hz. When the frequency is higher than 4 Hz, the tip displacement decreases continuously[89]. The appearance of the two different rules may be attributed to the difference between the preparation process and the internal parameters. For the current general IPMC, the latter rule seems to be the more accurate embodiment.

Effects of electrode characteristics

(1) Morphology of the electrode

The electrode morphology has an important effect on the actuating performance of IPMC, especially the roughness of the electrode. Aureli *et al*. analyzed the effect of electrode surface roughness on the electrical impedance of IPMC using the linearized Poisson-Nernst-Planck model[90]. They found that the increase in the surface area of the effective electrode greatly enhanced the charge storage of the IPMC. In general, the roughness of the electrode is usually determined by surface roughening in the fabrication process. Jin *et al*. studied the influence of different roughening methods on the actuating performance of IPMC and concluded that IPMC roughened by manual grinding has better performance compared with sanding machine polishing[91]. However, manual grinding has large errors and low repeatability, so how to improve the effect of mechanical roughening is the key to improving the performance of IPMC. Wang *et al.* further quantified the relationship between the surface topography of the matrix and the electromechanical properties of IPMC with the help of sandblasting[92]. Sandblasting can effectively form a controllable rough surface on the ionomer matrix, the principle of which is shown in Figure 6A. The longer the sandblasting time and the larger the gravel radius, the roughness of the surface after roughening will be higher and the penetration depth of the electrode will be greater. The experimental results of Liu *et al*. show that the impact level of different factors in sandblasting on the electrode roughness is that sand-size > sandblast pressure = sandblast time[93]. From the perspective of multi-physics model, they also revealed that the rough landscape of the interface can contribute to the formation of a thicker potential boundary layer, which can lead to larger charge accumulation and better water transport, hence larger deformation[94].

Table 2. Advantages and disadvantages of different electrode forming methods

Preparation methods		Advantages	Disadvantages
Physical methods	Physical vapor deposition	1. Controllable electrode thickness 2. Good electrode uniformity 3. Fast preparation speed 4. Suitable for large-scale preparation	1. Expensive cost 2. Poor electrode adhesion 3. Electrode prone to cracking
	Solution casting	1. Simple operation and low price 2. Relatively good electrode adhesion	1. The electrode thickness is difficult to control 2. Easy to cause uneven electrode thickness 3. High surface roughness of electrode
	Hot-pressing	1. The overall thickness of the driver is controllable 2. Good repeatability	1. High temperature and pressure conditions are required 2. The electrode layer thickness is not controllable 3. Large residual stress and easy to deform
	Direct assembly process	1. Simple operation and low price 2. Controllable electrode thickness	1. Poor electrode adhesion 2. The electrode is prone to crimp 3. Poor capacity to withstand tangential forces
Chemical methods	Reductant permeation	1. Effectively obtain electrode layers based on different metals	1. Harsh experimental conditions 2. Complex operation
	Impregnation-reduction	1. Firm and durable electrode 2. High performance in actuating 3. The interface electrode can be formed effectively	1. Long preparation time 2. The process is tedious and delicate 3. The thickness and uniformity of the electrode are difficult to be controlled
	Reverse electroless plating	1. The electrode particle diameter is small 2. Low surface roughness	1. Similar to impregnation-reduction
	Electroplating	1. The operation is simple and fast 2. Effectively reduce the surface resistance	1. It needs to be used in conjunction with other methods

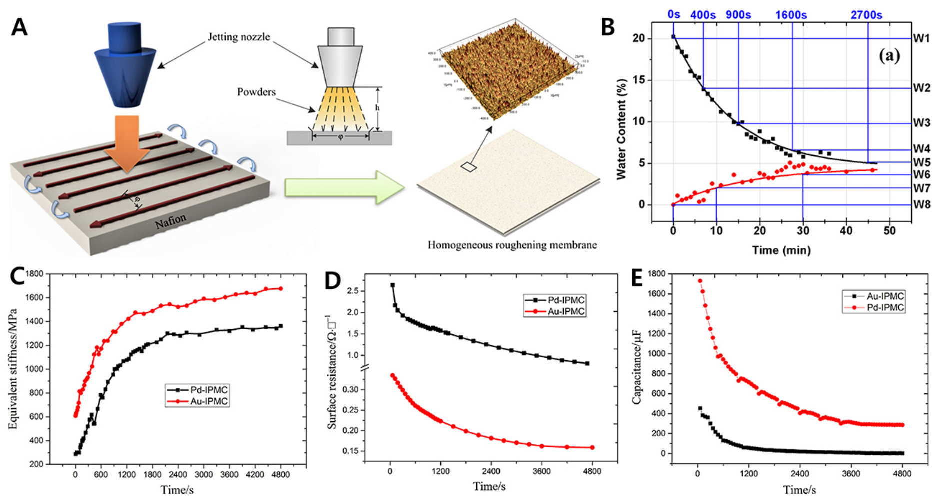

Figure 6. Performance of IPMC as actuators. (A) The principle of sandblasting. Reproduced with permission from Ref.[92]. Copyright© 2016 IOP Publishing Ltd. (B) Water content of IPMC during adsorption and desorption. Reproduced with permission from Ref.[47]. Copyright© 2014 AIP Publishing LLC. (C) The relationship between the stiffness and water content. Reproduced with permission from Ref.[98]. Copyright© 2014 Elsevier Ltd. (D) The relationship between surface resistance and water content. Reproduced with permission from Ref.[98]. Copyright© 2014 Elsevier Ltd. (E) The relationship between the capacitance and water content. Reproduced with permission from Ref.[98]. Copyright© 2014 Elsevier Ltd.

(2) Resistance and thickness of the electrode

In addition to roughness, characteristics such as resistance and thickness of electrode will also affect the actuating performance of IPMC. Shahinpoor *et al.* found that the reduction of surface electrode resistance can improve the actuating performance of IPMC[95]. They deposited a thin layer of silver (or copper) on top of the platinum electrode through electroplating to reduce the surface electrode resistance and tested the performance of IPMC actuator at low voltage. The results indicated that a lower surface electrode resistance will result in higher actuating capacity of IPMC. Punning *et al*. further revealed the influence of surface resistance on IPMC actuating performance[96]. The surface resistance of IPMC is highly correlated with the material curvature, and its curvature at a given point depends on the surface resistance, which in turn affects the further bending of the IPMC. In terms of the effects of electrode thickness, the actuating performance of IPMC has a nonlinear relationship with the electrode thickness, which increases first and then decreases with the increase of electrode thickness[33]. The reason lies in that the increase of electrode thickness will lead to the increase of spatial density of electric flux on the one hand and the increase of the IPMC stiffness on the other hand. While the optimization in terms of excitation voltages and electrode thickness is the result of the trade-off between electrical force acting on cations and stiffness of actuators.

Effects of the matrix

The ionomer matrix contains the media needed for IPMC actuating, including water and cations, *etc*. Therefore, the characteristics of the matrix, including water content, cation type and the size of the matrix film, will all affect the actuating performance of IPMC.

(1) Water content

The water content of IPMC can be controlled through the water exchange between IPMC and the environment [Figure 6B][47]. According to the experimental results of Zhu *et al*., with the decrease in water content, the back relaxation phenomenon will gradually disappear and the deformation of IPMC will become stable, but the stable value of tip displacement will also decrease with the decrease of water content[97]. The water content will affect the deformation behavior of IPMC by affecting the osmotic pressure and total and capillary pressure of the matrix. The back relaxation of IPMC is the result of reverse migration of free water. There is an optimal water content for IPMC to achieve maximum tip displacement. When the water content exceeds this optimal value, the reverse migration of the excess free water in the IPMC reduces the amount of cation migration at the cathode and leads to a small anode deformation. When the water content is below the optimal value, the lack of free water will reduce the mobility of water and cations, which results in a smaller and slower anode deformation. Besides, mechanical and physicochemical parameters of IPMC, such as stiffness, surface resistance and capacitance of IPMC, are closely related to water content [Figures 6C-E], which increased or decreased by more than three times during dehydration[98]. These factors play a leading role in different stages, affecting the deformation and response speed of IPMC. When operating in an open environment, the stable water content of IPMC is determined by the relative humidity of the environment, and increasing the ambient humidity to more than 50% can help reduce the corrosive effect of IPMC and improve its working time in an open environment[99].

(2) Cation types and thickness

Cations are the power source of IPMC deformation, hence the type of which has an important influence on the driving performance of IPMC. Shahinpoor *et al*. tested the blocking force of IPMCs exchanging different cations including Na^+, Li^+, K^+, H^+, Ca^{2+}, Mg^{2+}, Ba^{2+}, as well as larger hydrophobic cations such as TBA^+ and TMA^+, among them IPMC with Li^+ had the maximum blocking force[100]. While in terms of the tip displacement, IPMC exchanging TMA^+ had the maximum tip displacement, for that TMA^+ can effectively inhibit water electrolysis[41]. In general, the bending response of the IPMC actuator increases with the ionic radius because a larger ionic radius leads to a larger volumetric strain gradient between electrodes. In addition to the water content and cation type in the matrix, the thickness of the matrix itself will also affect the actuating performance of IPMC, which has been explored by Oh *et al*.[101]. They concluded that the blocking force increases while the tip displacement decreases with the increase of the thickness, which is related to the stiffness change caused by thickness[101].

Effects of other factors

Aside from intrinsic parameters, external conditions such as temperature and clamping force also have influences on the actuating performance of IPMC. For Nafion-IPMC, the temperature change will affect its recovery speed rather than deformation amplitude. The test results of Kobayashi *et al*. show that with the increase in temperature (253-393 K), the recovery speed of IPMC will be faster[89]. While for IL-IPMC, temperature affects not only the response speed, but also the deformation amplitude of IPMC. According to the research results of Almomani *et al*., the increase in temperature (25-70 °C) will lead to a decrease in deformation[102]. The relationship between cationic curvature and temperature is nearly linear, while the relationship between anionic curvature and temperature is nonlinear, reaching a minimum at 60 °C. When used as an actuator, one end of the IPMC is usually clamped by a clamping device to form a cantilever structure, and the clamping force also affects the actuating performance of the IPMC, which was studied by Moeinkhah *et al*.[103]. During the clamping process, the IPMC is squeezed along the thickness of the clamping area, which results in a change in its mechanical stiffness. Furthermore, the electrical contact resistance between the IPMC and the two electrodes of the clamping device will be greatly altered depending on the clamping pressure. There is an optimum clamping pressure for getting the best actuation performance of the IPMC actuator because of a balance between mechanical and electrical effects.

Optimization methods of IPMC actuators

Under the guidance of the influence rule of the above factors, scholars have proposed various methods to optimize the actuating performance of IPMC, mainly reflected in two aspects, including electrode and matrix.

Increasing interface area

Higher electrode interface area is helpful in improving the actuating performance of IPMC, including tip displacement and blocking force. Therefore, some scholars tried to improve the electrode interface area of IPMC by various methods. Roughening is the most direct and effective way to obtain high interface area. Chang *et al*. proposed that microneedle roller roughening (MNRR) could be used to improve interface area of IPMC[104]. They obtained deep needle-like penetrated electrode after the first impregnation process by roughening the membrane using microneedle roller. This method is direct, convenient and cost-effective, which increased the tip displacement of IPMC by 300%. Apart from roughening, there are some other simple physical methods to obtain high interface area. For instance, Ma *et al*. proposed a method of using isopropyl alcohol (IPA) assisted electroless plating to improve the interface area of IPMC[23]. The principle is shown in Figure 7A; they put Nafion containing platinum ions into IPA solution to make it fully swollen to obtain a larger volume, and then carried out reduction plating steps. Due to the higher surface area of the matrix, electrodes with higher interface area were obtained after reduction plating [Figure 7B]. The IPMC actuator prepared by this method has a tip displacement of 35.3 mm and a blocking force of 33 mN. In a similar way, Liang *et al*. prepared an IPMC with Pd-Pt composite electrodes[105], which has a tip displacement of 36.5 mm. Jung *et al*. obtained Pt electrodes with higher interface area by coating a layer of PVA outside Nafion membrane instead of roughening[106], which increased the blocking force of IPMC by 100% but resulted in a 26% reduction in tip displacement. Effective interfacial electrodes can also be obtained by compositing with non-metallic electrodes. For example, Ru *et al*. have formed an interface layer of IPMC through hot-pressing sulfonated multi-walled carbon nanotube (SCNT)/Nafion hybrid layer, and formed a layer of Pt surface electrode through electroless plating[107]. The large thickness of the interface layer increased the tip displacement of IPMC by 150% and the blocking force by 285%. Tian *et al*. increased the interface area of the electrode by adding silica and graphene oxide (GO) in Nafion[108], which increased

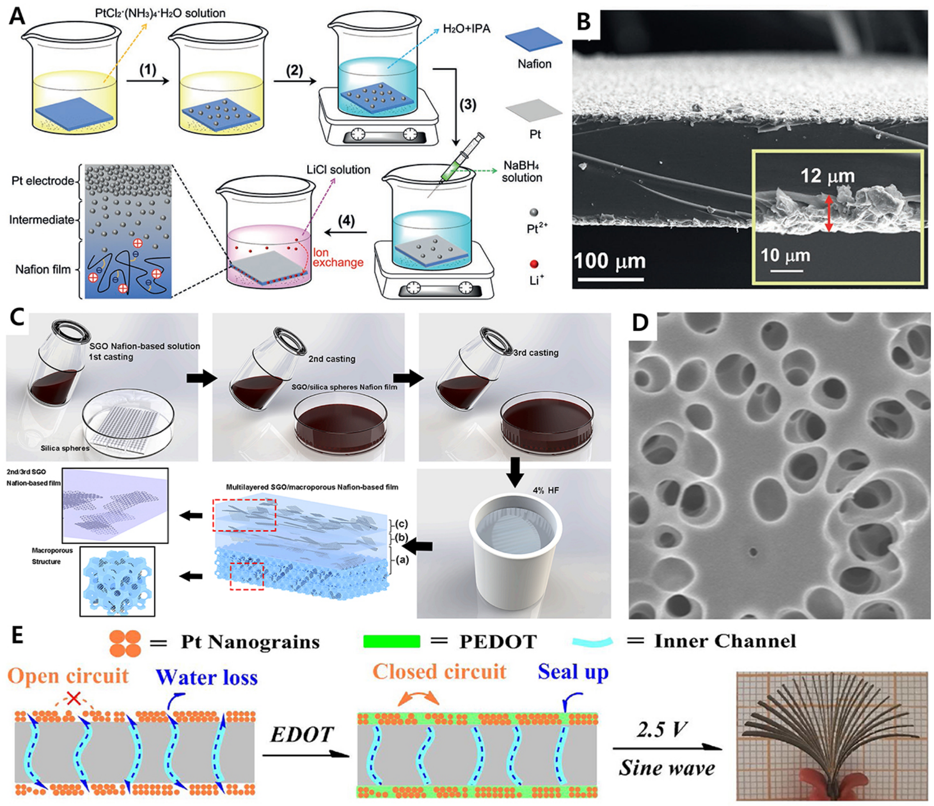

Figure 7. Optimization of IPMC actuator by increasing interface area and WU. (A) The principle of IPA assisted electroless plating. Reproduced with permission from Ref.[23]. Copyright© 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) The electrode of IPMC using IPA assisted electroless plating. Reproduced with permission from Ref.[23]. Copyright© 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (C) Process of preparing a macroporous multilayer SGO/Nafion polymer. Reproduced with permission from Ref.[114]. Copyright© 2021 Elsevier B.V. (D) A porous structure of SGO/Nafion polymer. Reproduced with permission from Ref.[114]. Copyright© 2021 Elsevier B.V. (E) Encapsulation of IPMC using PEDOT. Reproduced with permission from Ref.[116]. Copyright© 2019 Elsevier B.V.

the tip displacement and driving force of IPMC by 110% and 200%, respectively. Besides, some chemical methods can also effectively improve the interface area of IPMC. For instance, Wang *et al*. formed the DIE inside IPMC by impregnation electroplating, which greatly increased the interface area of IPMC and the tip displacement by 366%[34]. However, the existence of DIE will hinder the ion conduction to a certain extent, resulting in the reduction of the response speed.

Increasing WU capacity

The water content and ion type of matrix both affect the actuating performance of IPMC. Among them, increasing the WU of the matrix is the most commonly used optimization method, which mainly includes improving the porosity and doping moisture-sensitive materials. It is hard for pure Nafion to get a porous structure. Zhao *et al*. developed a freeze-drying process to endow Nafion membrane with a porous structure with a porosity of nearly 40.45%[109]. The WU of the prepared IPMC increased to 40.45% and the tip displacement was increased by 4963.6% under a voltage of 2 V. But the blocking force is reduced by 72.1% due to the decrease of elastic modulus caused by porous structure. Microcellular foaming process can also be used for the preparation of porous Nafion[110]. The pores are generated by volatilization of gases dissolved in the polymer, resulting in a porous structure. The WU of the prepared IPMC was increased to 40.45%, and the tip displacement and blocking force were increased by 50% and 100%, respectively. Compared with improving porosity, doping moisture-sensitive materials is a simpler and more effective way to improve the WU of IPMC. For example, doping polyethylene oxide (PEO)[111] and sulfonated graphene oxide (SGO)[112] with Nafion can increase the WU of IPMC to 40% and 60%, respectively. Bian *et al.* doped $BaTiO_3$ nanoparticles into Nafion, which did not improve the WU but improved the mechanical and electrical performance of IPMC, and the tip displacement and blocking force were increased by 250% and 375%, respectively[113]. Some researchers have combined the above two methods to prepare doped porous Nafion matrix. For example, Zhang *et al.* prepared a macroporous multilayer SGO/Nafion polymer using SGO, the process of which is shown in Figure 7C and the porous structure is shown in Figure 7D[114]. The experimental results showed that the tip displacement and blocking force of the prepared IPMC increased by 150% and 377%, respectively. Furthermore, a macroporous multilayer multiwall carbon nanotubes (MCNTs)/Nafion polymer was obtained by replacing SGO with MCNTs[115]. Although the improvement of WU can enhance the tip displacement and blocking force of IPMC, it will still exchange water with the environment. In order to improve the working time of IPMC, encapsulation is a good optimization method. Guo *et al.* used PEDOT layer to prevent the water exchange between IPMC and the external environment and improved the working time of IPMC[116], as shown in Figure 7E. Besides, the materials used for encapsulation of IPMC include parylene[117], PDMS[118], PVP[119] and liquid metal[120].

Increasing the thickness of IPMC

As mentioned in Section "Effects of the matrix", the thickness increase of IPMC can also effectively improve the blocking force despite the decrease in tip displacement. Therefore, some researchers tried to optimize the performance of IPMC actuator from the perspective of matrix thickness. For example, Park *et al.* enhanced the generating force of IPMC through thickness manipulation[121]. They formed multilayer Nafion membrane into a whole by hot-pressing a plurality of Nafion 117 membranes into a whole and prepared an IPMC on the basis, which produced a blocking force of 250 mN. Then they further combined it with IPA assisted electroless plating [Figure 8A], increasing the blocking force to 550 mN and even increasing the tip displacement by 4 times[122]. Swarrup *et al.* stacked the prepared IPMC from the width direction to the thickness direction to improve the generating force of IPMC because stacking leads to an increase in stiffness[123]. In other words, stiffness increase is an effective method to improve the blocking force. They found that the force improvement efficiency was higher when stacked IPMC from the width direction. Bian *et al.* stacked IPMC in three ways, namely width stacking, RSSS and USSS of thickness stacking [Figure 8B][124]. In the stacking state of 5 IPMCs, the three stacking modes increased the blocking force of IPMC by 400%, 240% and 373%, respectively, but the tip displacement did not increase significantly. Lee *et al.* prepared a three-layer polyelectrolyte film by continuous solution casting [Figure 8C][125], which is composed of a Nafion layer containing an amphiphilic organic molecule (10-camphorsulfonic acid; CSA) in the middle section and two Nafion/modified inorganic composite layers in the outer sections. The middle layer is used for fast and large ions migrating, while the outer layer is used for large accumulation/retention of mobile ions. Compared with the traditional IPMC, its end displacement was increased by 60%.

Other methods

Aside from the optimization methods for matrix and electrode mentioned above, there are some other methods to improve the actuating performance of IPMC. For example, Boldini *et al.* enhanced the deformation range of IPMC through electrostatic actuation[126]. As shown in Figure 8D, they placed two parallel metal plates on both sides of the IPMC, and the electrostatic field generated by the applied voltage could enhance the deformation of the IPMC [Figure 8E], the tip displacement of which was increased by 80% under the electrostatic field of 4 kV. Ru *et al.* proposed a method to control IPMC by moisture coupled with electricity[127], which can help eliminate the unstable factors in the actuating process of IPMC, such as back relaxation. Lu *et al.* used ethanol immersion to reduce the crystallinity of Nafion[128], obtained a matrix

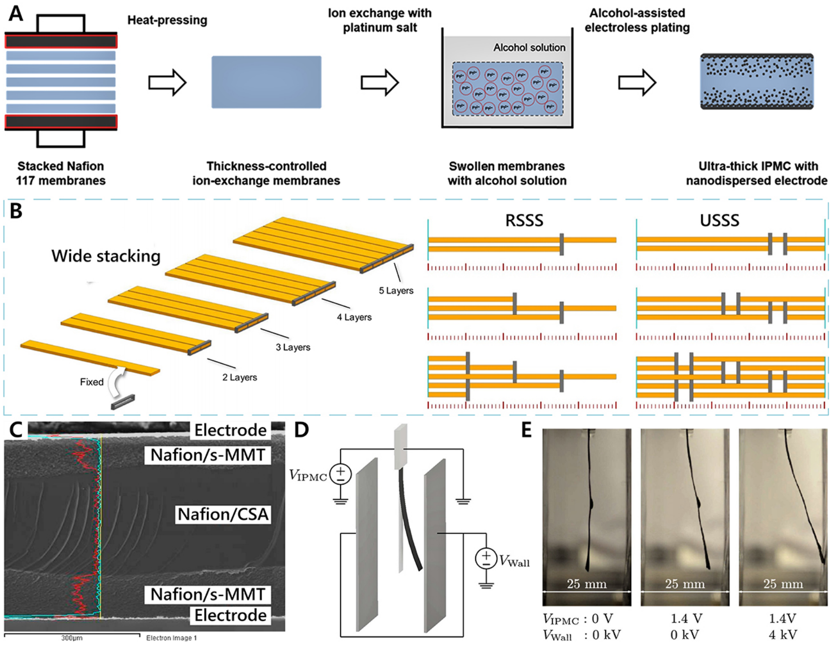

Figure 8. Optimization of IPMC actuator by increasing thickness. (A) IPMC prepared through hot-pressing multilayer Nafion 117 and IPA assisted electroless plating. Reproduced with permission from Ref.[122]. Copyright© 2017 American Chemical Society. (B) Different stacking ways of IPMC in width and thickness direction. Reproduced with permission from Ref.[124]. Copyright© 2021 IOP Publishing Ltd. (C) Three-layer polyelectrolyte film prepared by continuous solution casting. Reproduced with permission from Ref.[125]. Copyright© 2014 American Chemical Society. (D) The mechanism of enhancing the deformation of IPMC by electrostatic field. Reproduced with permission from Ref.[126]. Copyright© 2018 AIP Publishing. (E) The deformation of IPMC under different electrostatic fields under the applied voltage of 1.4 V. Reproduced with permission from Ref.[126]. Copyright© 2018 AIP Publishing.

with a lower elastic modulus, and the tip displacement of the prepared IPMC was increased by 500%. Xu *et al*. increased the bending angle of IPMC by changing the electrode separation[129]. When the electrode spacing increased from 3 mm to 7 mm, the bending angle of IPMC increased from 5° to 35°.

The effects of various optimization methods for IPMC actuators are summarized and listed in Table 3. In general, many methods can improve the actuating performance of IPMC, and even improve the tip displacement and blocking force at the same time. However, there is an upper limit to the actuating performance of IPMC; for example, the blocking force can only be up to 700 mN. Therefore, the choice of IPMC should be based on the application scenario rather than the pursuit of performance improvement.

Performance of IPMC as sensors

When IPMC is used as a sensor, the ions in which will migrate directionally under different external stimuli, such as bending, pressure and humidity. The ion migration of bending sensing originates from the uneven stress distribution in the thickness direction caused by the overall deformation. The ion migration of the pressure sensing comes from the coupling effect of shock load and local deformation. While used as a humidity sensor, after one side of IPMC adsorbing water, the ion concentration will decrease. In the meantime, there will be a concentration difference between the two sides of IPMC, which will cause the internal cations to migrate.

Bending sensing performance

The bending sensing principle of IPMC is shown in Figure 9A[25]. When the whole IPMC is bent, one side is stretched, and the other side is compressed. The hydrated cations will migrate directionally along the

Table 3. The effects of various optimization methods for IPMC actuators

Optimization mechanisms	Specific methods	Increase in tip displacement	Increase in blocking force	Reference
Increasing interface area	microneedle roller roughening	300%	-	[104]
	IPA assisted electroless plating	200%	65%	[23]
	PVA instead of roughening	100%	-26%	[106]
	Compositing non-metallic electrode	150%	285%	[107]
	Impregnation electroplating	360%	-	[34]
Increasing WU capacity	Forming porous structure	50%	100%	[110]
	Doping PEO	116%	26.7%	[111]
	Doping SGO	700%	-	[112]
	Doping MCNTs to form porous structure	99.4%	102.3%	[115]
Increasing thickness	Hot-pressing multilayer Nafion	400%	2000%	[122]
	Stacking multi-chip IPMC	-	400%	[124]
	Casting multilayer polyelectrolytes	60%	-	[125]
Others	Electrostatic field coupling	80%	-	[126]
	Soaking ethanol to reduce crystallinity	500%	-	[128]
	Changing the electrode separation	600%	-	[139]

thickness direction under the action of the stress gradient, thus outputting a voltage signal between the two electrodes. The reason why IPMC can be used as a bending sensor is that there is a quantitative relationship between the output voltage of IPMC and the bending curvature. Song *et al*. showed an approximately linear relationship between the output voltage of the IPMC sensor and the bending curvature by controlling the bending deformation of the whole IPMC[130]. In addition, the output voltage of the IPMC is also related to its size, and they found that with the same curvature, the output voltage of the IPMC increases with the increase of width, while it is almost not affected by the length and thickness. Wang *et al*. studied the relationship between the output voltage and the size of IPMC with the tip displacement as a fixed quantity[28]. Their experimental results showed that the output voltage of IPMC increases with the increase of thickness and decrease of length but is not affected by the width. Besides, the frequency of bending also affects the sensing voltage of the IPMC, and Dominik *et al*. found that the sensitivity of the IPMC sensor increases with frequency when the frequency is less than 1 Hz[131]. However, due to the low-speed characteristics of ion migration, high working frequency will affect the performance of IPMC sensor. Therefore, IPMC is generally used for a low frequency bending sensor. Because the voltage of IPMC sensor comes from the migration of hydrated cations, the internal water content will also affect its sensing behavior, which is mainly determined by the ambient humidity. Brunetto *et al*. first studied the effect of relative humidity on the sensing performance of IPMC[132]. The results show that the output voltage of IPMC sensor is strongly dependent on the environmental humidity, and the voltage amplitude of the IPMC sensor increases with the increase of humidity. This is because the increase of internal water content reduces the resistance of ion migration, resulting in the increase of the ions number and speed of ion migration. But on the other hand, the increase in humidity results in an increase in the signal-to-noise ratio of the output signal. Zhu *et al*. have done further in-depth research on this matter[38,39,133]. They found that the bending sensing ability of IPMC increases first and then decreases with the increase of environmental humidity, which reaches the peak when the relative humidity is 90%. When the humidity is further increased, the sensing ability is gradually weakened due to the effect of back relaxation. Besides, the cation types also affect the bending sensing ability of the IPMC. With the decrease in the cation radius, the sensing ability of IPMC is gradually enhanced, because the cations with smaller radius have a stronger ability to bind water and are easier to migrate in the nanochannel.

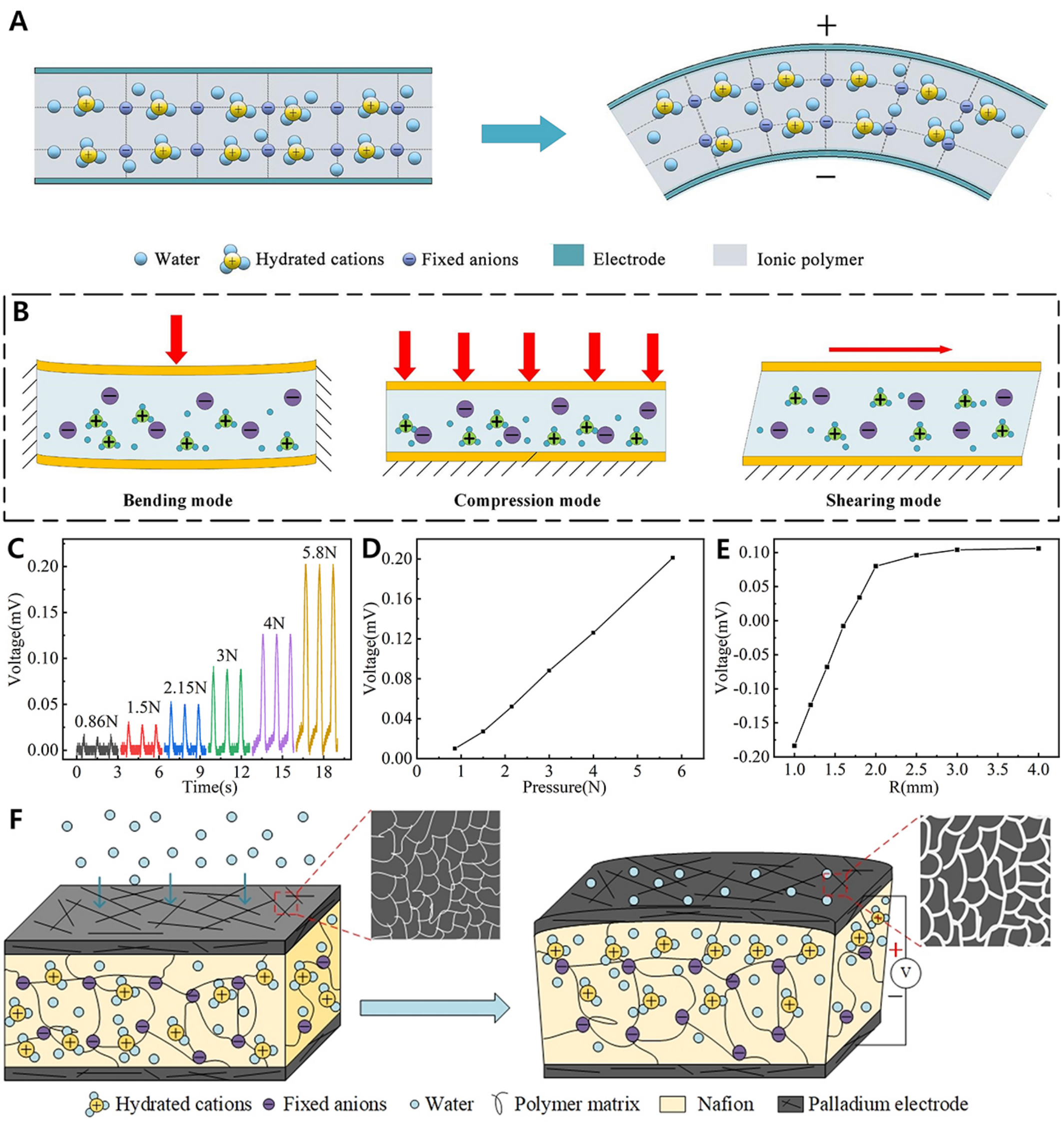

Figure 9. Performance of IPMC sensor. (A) The bending sensing principle of IPMC. (B) Three working modes of IPMC pressure sensor. Reproduced with permission from Ref.[136]. Copyright© 2022 IOP Publishing Ltd. (C) The output voltage of IPMC under a dynamic force with different peaks. Reproduced with permission from Ref.[136]. Copyright© 2022 IOP Publishing Ltd. (D) The evolution of voltage amplitude of IPMC as the peak of the applied pressure increases. Reproduced with permission from Ref.[136]. Copyright© 2022 IOP Publishing Ltd. (E) The evolution of the output voltage of IPMC as the radius of the contact circle increases. Reproduced with permission from Ref.[136]. Copyright© 2022 IOP Publishing Ltd. (F) The humidity sensing principle of IPMC. Reproduced with permission from Ref.[32]. Copyright© 2021 Elsevier B.V.

Pressure sensing performance

The pressure sensing mechanism of IPMC is similar to that of bending sensing, which mainly originates from the ion migration caused by deformation together with the ion migration caused by shock loading. Gudarzi *et al.* divided the working modes of the IPMC pressure sensor into three types, namely bending mode, compression mode and shearing mode[134,135], as shown in Figure 9B[136]. The bending mode is to fix the edge of IPMC slice, and the load exerted on the surface can be measured through the voltage caused by the deformation of IPMC. This mode can only measure relatively concentrated small loads because there is no support behind the IPMC. While the shearing mode is to fix one surface of the IPMC and then the friction on the other surface can be measured according to the generated voltage. However, it is necessary to understand the friction coefficient when measuring the pressure, and the result of the measurement needs to be converted. Comparatively speaking, the compression mode is the most direct and effective method to measure the pressure, but the output voltage of which is rather low compared with the bending mode. Similar to bending sensing, there is also a certain correspondence between the output voltage of the IPMC and the applied pressure when it is used as a pressure sensor. According to the experimental results of Wang *et al*., when applying a dynamic force with a period of 1 Hz, there is an approximately linear relationship between the output voltage of IPMC pressure sensor and the applied pressure, as shown in Figure 9C and D[136]. In addition, they also studied the relationship between the output voltage of the IPMC pressure sensor and the contact area. As shown in Figure 9E, when the radius increases from 1 mm to 2 mm, the output voltage quickly changes and undergoes a change from negative to positive. When the radius is between 2 mm and 3 mm, the growth rate of the voltage is obviously reduced. When the radius is larger than 3 mm, the voltage remains constant. However, if the characteristics of the applied force change, the electrical response will change accordingly. For example, if a static state is applied, the output voltage will drop slowly after reaching a peak, which is also caused by back relaxation. We have added the above analysis in the revised version.

Humidity sensing performance

When used as a humidity sensor, IPMC can not only generate a voltage upon the change of humidity, but also its capacitance and surface resistance will change with the humidity, the humidity sensing principle of which is shown in Figure 9F[32]. After absorbing water, the physical properties of IPMC show an obvious uneven swelling. Firstly, the swelling of Nafion layer after absorbing water leads to a change in the capacitance of the double layer. It indicates that this film can be potentially used as a capacitance-humidity sensor. Secondly, due to the loose characteristics of the surface electrode, the resistance of the electrode layer increases accordingly together with the volume change caused by IPMC water absorption, with which IPMC can be potentially used as a resistance-humidity sensor. Finally, internal ions migrate in a directional way under the action of uneven swelling of IPMC, and ionic electrical response can be detected between the electrode layers. It means that IPMC can be potentially used as a voltage-humidity sensor. Esmaeli *et al*. first studied the humidity-capacitance sensing performance of IPMC[68]. They prepared an IPMC humidity sensor by sputtering a layer of Cr and Ti on the surface of Nafion, which can detect humidity in the range of 40% RH to 90% RH with an average sensitivity of 140 nF. The linearity of the prepared sensor is poor, and the sensitivity increases with the increase of humidity, which is caused by the influence of water content inside the IPMC. Beigi *et al*. investigated the humidity sensing capability of IPMC doped by double hydroxide by taking voltage as the measurand parameter[137]. They realized the humidity detection in the range of 30% RH to 95% RH. In addition, they found that the humidity-voltage sensing ability of IPMC was related to its bending angle. The IPMC with a larger bending angle has a stronger humidity sensing ability, which is attributed to the fact that the bending state increases the initial stress inside the IPMC to facilitate ion migration. Wang *et al*. conducted a comprehensive study on the capacitance, resistance, and voltage humidity sensing performance of IPMC prepared by IRP methods, on the occasion of considering the change of the thickness, cations type and IRP times[32]. It was found that the capacitance and resistance can respond to the static humidity, while the voltage can only be generated from dynamic humidity. The humidity response ability of IPMC is closely related to its water absorption ability; less IRP times, smaller thickness and cation radius will lead to stronger water absorption ability. In addition, in terms of sensing range, the capacitance response can only sense humidity in the range of 22% RH to 100% RH, while the resistance and voltage response can achieve full humidity range sensing. In terms of response time, the response time of capacitance and voltage is as long as several hours, while the response time of voltage is only about 0.5 s.

Optimization of IPMC sensors

Bending sensing optimization

The optimization of IPMC sensor can also be carried out by modifying electrode and matrix. In particular, researchers have done a lot of work to optimize the bending sensing performance of IPMC. In terms of electrodes, Fu *et al.* increased the bending sensing voltage of IPMC to 200 mV by sputtering silver electrodes, which is about 20 times higher than that of ordinary IPMC[67]. The method of increasing the interface area also has a good effect on optimizing the sensing performance of IPMC. For example, Palmre *et al.* developed a kind of Pd/Pt-IPMC with large surface area electrodes and thick ionomer membrane[138]. Compared with the pure Pt-IPMC, the response voltage is improved by 200%. Hong *et al.* varied the morphology of the metal composite component (conductive network composite (CNC)) of IPMC through physical casting[139], the sensitivity of which is three times higher than that of ordinary IPMC. In terms of the optimization methods for the matrix, Chang *et al.* introduced a biological template method to fabricate IPMC strain sensor with bionic hierarchical structure[140]. As shown in Figure 10A, they cast a Nafion membrane with hierarchical structure using reed leaves as a substrate, and prepared an IPMC on the subatrate. The prepared IPMC obtained a 300% increase in sensitivity and was able to respond to subtle changes in facial expression. By replacing the electrolyte inside IPMC with ionic liquid and replacing the metal electrode with graphene electrode, He *et al.* increased the output voltage by 2.74 times[141]. Furthermore, the output voltage of the IPMC is very stable and does not decay over time due to the non-volatile nature of the ILs. However, the maximum voltage of Nafion-IPMC bending sensor can only reach millivolt level, and the type of ionomer needs to be changed to obtain higher output voltage. A novel strategy including the introduction of poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) into a poly(vinylidene-fluoride-trifluoroethylene-chlorotrifluoroethylene) (P(VDF-TRFE-CTFE)/polyvinylpyrrolidone blend was proposed by Panwar *et al.* to develop a new IPMC, which exhibited a high output voltage of up to 27 V in IL condition[142]. In addition to optimizing the intrinsic parameters of IPMC, some scholars have tried to optimize the bending sensing performance of IPMC from the perspective of external working conditions. For instance, Qaviandam *et al.* proposed a new approach to improve the sensing performance of IPMC through sensor biasing[143]. By applying an appropriate electrical bias signal, the sensitivity of IPMC can be improved by 446.5 times. Tang *et al.* proposed a novel strategy to enhance the bending sensing voltage of IPMC through magnetoelectricity[144]. As shown in Figure 10B, they placed two parallel plate permanent magnets on both sides of the IPMC. When the IPMC bends in the magnetic field, in addition to the voltage generated by ion migration, the metal electrode cutting the magnetic induction line will also generate an induced electromotive force. The coupling effect realizes the enhancement of the output voltage. The voltage enhancement of IPMC increases with the increase of bending frequency and magnetic field intensity, which reached 158% when the frequency is 8 Hz and the magnetic field intensity is 100 mT.

Pressure sensing optimization

Because the mechanism of pressure sensing is similar to that of bending sensing, the optimization methods mentioned above for intrinsic characteristics of IPMC are also effective in improving its pressure sensing performance. Besides, there are some other methods that can be used to optimize the performance of IPMC pressure sensors. For example, Histed *et al.* reported the development of tailored 3D-structured polymer-metal interfaces to create enhanced IPMC sensors towards high sensitivity pressure sensor applications[145]. They introduced a novel advanced additive manufacturing approach to tailoring the morphology of the polymer-electrode interfaces via inkjet-printed polymer microscale features. Zhu *et al.* tried to improve the pressure sensing performance of IPMC by using gradient shape, as shown in Figure 10C[79]. Due to the asymmetry of the upper and lower surface electrodes, the stress gradient of the IPMC under pressure is more uneven, the sensitivity of which will increase with the increase of the area ratio and reach saturation gradually. When the area ratio is 7, it generates an output voltage of 46 mV under a load of 5 N, which is

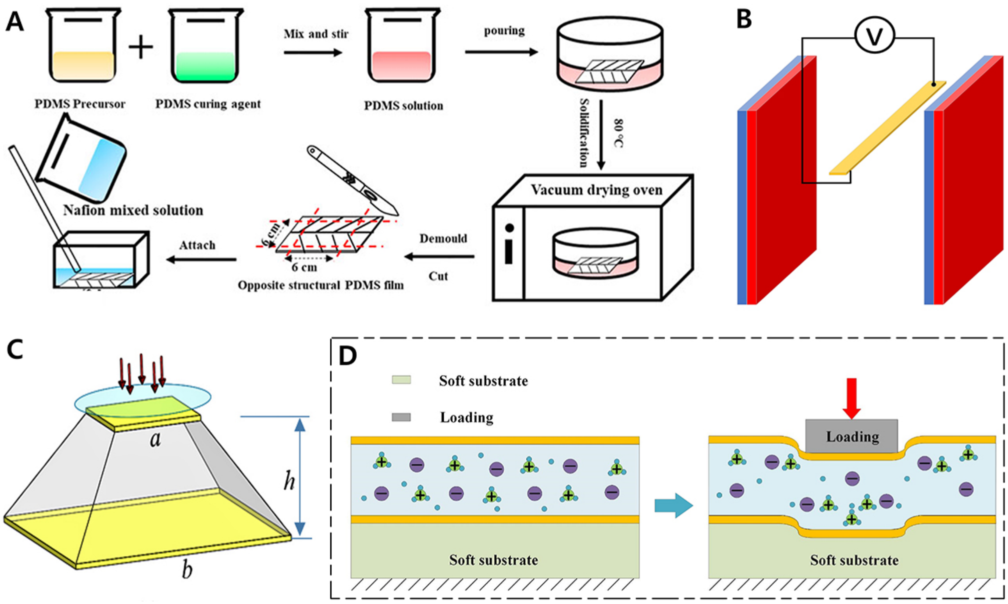

Figure 10. Optimization of IPMC sensor. (A) Preparation process of Nafion membrane with hierarchical structure. (B) The mechanism of enhancing the bending sensing voltage of IPMC through magnetoelectricity. Reproduced with permission from Ref.[144]. Copyright© 2021 IOP Publishing Ltd. (C) An IPMC sensor with tailored 3D-structured polymer-metal interfaces. Reproduced with permission from Ref.[79]. Copyright© 2019 AIP Publishing. (D) Amplifying the deformation by placing a soft substrate under the IPMC pressure sensor. Reproduced with permission from Ref.[136]. Copyright© 2022 IOP Publishing Ltd.

about 460 times higher than that of a common sheet-like IPMC. By placing a soft substrate under the IPMC pressure sensor to amplify its deformation [Figure 10D], Wang *et al*. realized the improvement of the pressure sensing signal[136]. Compared with working on a rigid substrate, the output voltage of IPMC with a soft substrate has been improved by 100%.

Humidity sensing optimization

The optimization of IPMC humidity sensing performance can also start from both the substrate and the electrode. According to the research results of Wang *et al*., the humidity sensing performance of IPMC is related to the thickness, cation types and IRP times[32]. By selecting thinner thickness, cations with stronger absorption and reducing the IRP times, the humidity response voltage of IPMC can be increased by 150%, 1133% and 200%, respectively. It can be seen that improving the water absorption of the matrix has the best optimization effect. Beigi *et al*. deposited double hydroxide with good hydrophilicity in Nafion to improve the water absorption of IPMC[137], which increased the output voltage of IPMC humidity sensor by 67%. Furthermore, they found that increasing the bending angle of IPMC can also improve the humidity sensing voltage of IPMC, which was increased by 92% when the bending angle reached 90°. Wang *et al*. reduced the density of the electrode by spraying silver nanowires to form electrodes, which made it easier for IPMC to exchange humidity with the environment. Under 100% RH condition, its output voltage reached 8.5 mV, which was 1316% higher than that of IPMC prepared by electroless plating[35].

The effects of various optimization methods for IPMC sensors are summarized and listed in Table 4. It can be seen from the above that IPMC has the potential formulti-functional sensing. That is to say, IPMC can work on its own functioning as a variety of sensors on some occasions, which provides an attractive prospect for the application of IPMC sensors.

Table 4. The effects of various optimization methods for IPMC sensors

Optimization aspects	Specific methods	Increase in tip displacement	Reference
Bending sensing	Sputtering silver electrode	2,000%	[67]
	Forming Pd-Pt electrode with high interface area	200%	[138]
	Varying the morphology of CNC	300%	[139]
	Casting hierarchical structure	300%	[140]
	Replacing the electrolyte with IL	274%	[141]
	Changing the type of ionomer	270,000%	[142]
	Sensor biasing	44,650%	[143]
	Applying magnetic field	156%	[144]
Pressure sensing	Tailored 3D-structured polymer-metal interfaces	130%	[145]
	Gradient shape	46,000%	[79]
	Placing a soft substrate	100%	[136]
Humidity sensing	Reducing the thickness	150%	[32]
	Selecting cations with stronger absorption	1,133%	[32]
	Reducing the IRP times	200%	[32]
	Depositing double hydroxide	67%	[137]
	Forming electrodes by spraying silver nanowires	1,316%	[35]

APPLICATIONS OF IPMC

IPMC can be used in many fields because of its actuating and sensing functions. For example, as an actuator, IPMC can be used in the fields of bionic robots, micro-grippers and medical devices due to the material characteristics of biological muscles. While as a sensor, it is usually used as artificial skin in wearable sensing devices, energy harvesting and other fields because of its good biological skin-like sensing characteristics.

Applications of IPMC as actuators

Bionic robots

(1) Underwater robots

Bionic robots are an important application area of the IPMC actuator because of its large deformation and designable shape. Due to the strong dependence of IPMC actuator on water, the underwater environment provides a natural advantage for its actuating, so the application of IPMC robot is mainly underwater robots, such as bionic fish, bionic jellyfish and bionic turtle. In the application of biomimetic fish, IPMC is usually used for the design of actuating components such as caudal fin, pectoral fin, and so on. Yi and Sunkara *et al.* have carried out a series of studies on the application of IPMC in caudal fin propelled fish[146,147]. By controlling the motion mode of IPMC, the robotic fish can realize swimming forward and turning motion. IPMC can also be used in the pectoral fins of bionic fish, which have more diverse movement patterns compared with caudal fin propulsion. Shen *et al.* designed a robotic fish with multiple symmetrical pectoral fins, as shown in Figure 11A[148]. By regulating the movement time of each IPMC, a smooth movement like that of a manta ray can be produced [Figure 11B]. Compared with the caudal fin propulsion, the pectoral fin propulsion of the bionic fish is more stable, but the existence of multi-piece IPMC makes it more difficult to control. There are also scholars trying to combine pectoral fin propulsion with caudal fin propulsion to control the movement of robotic fish, which is the same as the movement pattern of most fish. Li *et al.* mounted both caudal and pectoral fins on a capsule-to-like underwater robot, where the caudal fin was used for forward propulsion and the combination of pectoral and caudal fins could be used to control steering[149]. In addition to robotic fish, IPMC can also be used in other bionic underwater robots. For example, Wang *et al.* designed a jellyfish-inspired robot by combining the IPMC with the DE

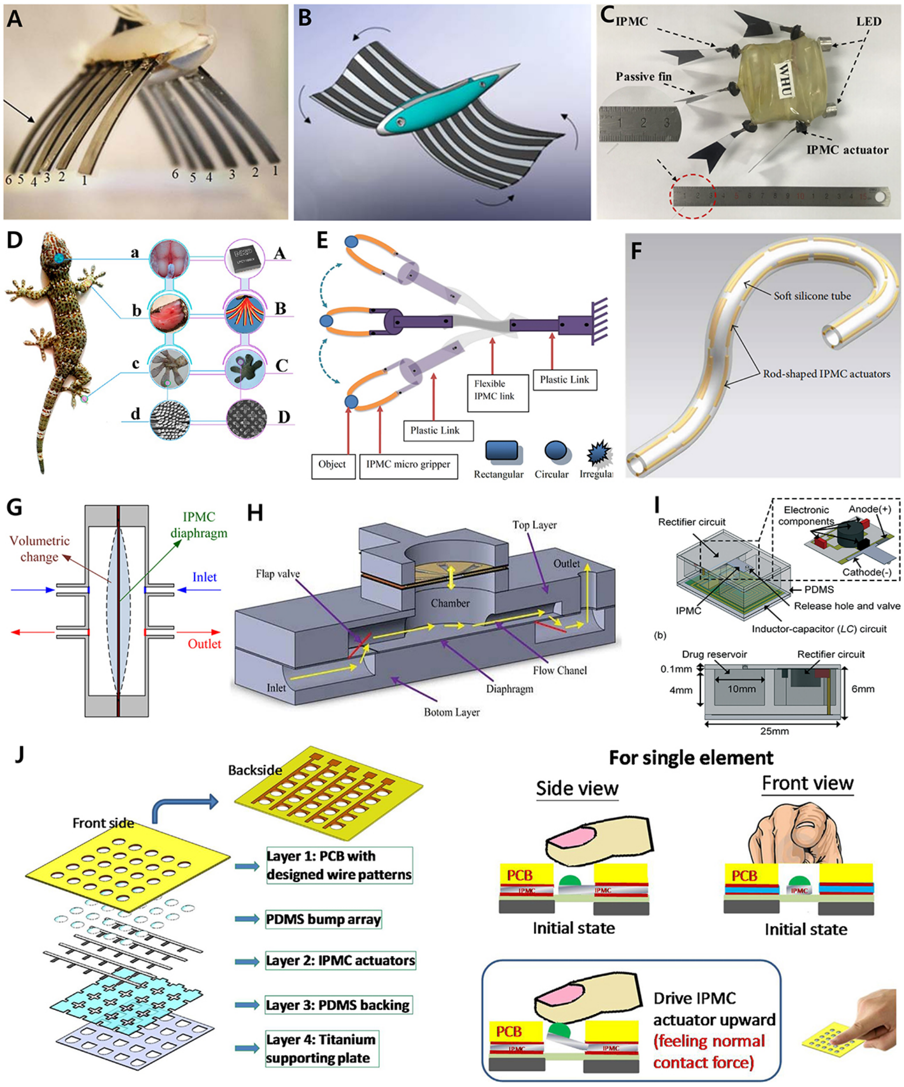

Figure 11. Applications of IPMC actuators. (A) A robotic fish with multiple symmetrical IPMC pectoral fins. Reproduced with permission from Ref.[148]. Copyright© 2020 IOP Publishing Ltd. (B) Smooth movement like manta. Reproduced with permission from Ref.[148]. Copyright© 2020 IOP Publishing Ltd. (C) A robotic tortoise with 5 IPMC actuators. (D) An actuator with adhesion function inspired by gecko. Reproduced with permission from Ref.[154]. Copyright© 2015 American Chemical Society. (E) Double active finger gripper. Reproduced with permission from Ref.[161]. Copyright© 2018 Society of Plastics Engineers. (F) Rod-shaped IPMC actuators embedded in the soft silicone tube. (G) Schematic diagram of a typical IPMC micropump. Reproduced with permission from Ref.[174]. Copyright© 2012 Elsevier B.V. (H) Micropump with an inner petal-shaped IPMC as an actuating diaphragm. (I) A wirelessly activated device with an integrated IPMC cantilever valve for targeted drug delivery. Reproduced with permission from Ref.[180]. Copyright© 2018 The Royal Society of Chemistry. (J) An innovative digitally controllable tactile bump array. Reproduced with permission from Ref.[182]. Copyright© 2018 Elsevier B.V.

actuator[150]. The DE material is used to provide power for the robot, while the deflection of the IPMC can change the direction of the robot. Sun *et al.* attempted to develop a robotic tortoise, in which five IPMC actuators were used to simulate the tortoise's limbs and tail, as shown in Figure 11C[151]. The collaborative control of multiple IPMCs can help the robot tortoise to realize the functions of swimming and steering. All the underwater robots mentioned above use IPMC as the actuator rather than the body structure. Because the driving force of the IPMC is small, the body structure needs to have a small mass to have a reasonable speed for the robot. In addition, the assembly of the actuator and the body also brings some challenges to the design of the robot, so some scholars try to use monolithic IPMC thin film to develop the underwater robot. For example, Kodaira *et al*. developed underwater robots of different shapes using the multilayer casting process, the underwater velocity of which was 2.3 mm/s[70].

(2) Other robots

In the case of solving the problem of water evaporation, IPMC can also be applied to other robots. For example, Nguyen *et al*. developed a quadruped walking robot based on 2-DOF IPMC legs, which can walk on a flat surface at a speed of 0.4 mm/s by carrying a PCB control board[152]. Inspired by Venus flytrap, Li *et al*. designed a soft robot by casting IPMC in PDMS matrix[153]. Through size optimization, the performance of the Venus flytrap robot could be close to the biological Venus flytrap and capture a small ball with a mass of 0.3 g firmly. Inspired by the adsorption mechanism of gecko, Guo *et al*. developed an actuator with adhesion function by connecting poly (methylvinylsiloxane) (PMVS) micropillars to IPMC, as shown in Figure 11D[154]. This novel gecko-inspired synthetic adhesive exhibited fascinating reversible adhesion and might be used in unprecedented devices, such as a spiderman-like wall-climbing device. Ishiki *et al*. prepared a novel PF-IPMC with paper and fabric as the substrate, which has good foldability[155]. On this basis, they developed a 3D crafted robot, which showed good driving repeatability.

(3) Robotic grippers

The microgripper for flexible grasping technology is also a very important application direction of IPMC due to its high flexibility. According to the number of IPMC actuators, the IPMC based microgripper can be divided into single active finger gripper, double active finger gripper, and so on. A typical single active finger gripper consists of an IPMC finger and an auxiliary rigid finger[156]. In order to improve the manual maneuverability of the single active finger gripper, Jain *et al*. employed electromyographic (EMG) signals generated by movement of index finger muscles through long tendons to control the deformation of the IPMC finger, which exhibited excellent flexibility and compliance in grasping[157,158]. Some scholars replaced the rigid finger of single active finger gripper with IPMC sensor to enhance the perception and control of the grasping process[159]. In order to improve the applicability of the single active finger gripper, Cheong *et al*. proposed a wireless powered single active finger microgripper that is operated using external radio-frequency magnetic field for biological cell manipulation application[160]. The double active finger gripper, as its name implies, is a gripper that uses two IPMC actuators, as shown in Figure 11E[161]. IPMCs with different ionomers have been used to develop double active finger gripper, such as Nafion[162], PVDF[163] and SPVA[161]. Yang *et al* demonstrated a self-powered actuation technique based on the IPMC-TENG conjunct system; the transferred charges from TENG can be utilized for driving and controlling the deformation performance of double active finger gripper based on IPMC[164]. Compared with double active finger gripper, single active finger gripper performs better[165]. By increasing the number of IPMC actuators, triple or even quadruple active finger gripper[53,72] can also be developed. With the increase of active fingers, the wrapping and stability in the grasping process will be enhanced, but the flexibility of grasping will be reduced.

Medical treatment devices

Because of the elastic modulus of IPMC close to that of human body and good biocompatibility, IPMC actuators also have broad prospects in the field of biomedicine, especially medical devices in surgical operation and drug delivery.

(1) Surgical operation devices

In the applications of surgical operation, IPMC actuator can be used to drive surgery end effector as well as surgical guide device. Feng *et al*. proposed a method to fabricate microfeature sized IPMC with arbitrary shapes, which can be used as tiny accurately controlled surgical devices[166]. The fabricated prototype was experimentally demonstrated to have potential application as an electronically activated microgripper in robotic laparoscopic surgical systems. Aw *et al*. designed a robotic surgical end effector with an embedded force feedback sensor implemented by using an IPMC actuator for actuating[167]. McDaid *et al*. developed a compliant surgical robotic instrument by integrating actuating and sensing function of IPMC[168]. Compared with conventional devices, the presence of IPMC enhances the natural compliance of the joint and ensures the ease of miniaturization for invasive surgery. In the application of medical catheters, the IPMC needs to be designed as a cylinder[169] or a square rod[170], and equipped with four electrodes to ensure the omnidirectional movement. For example, He *et al*. developed an interventional catheter with active guiding function based on the square rod-shaped IPMC, and the human aortic vascular model was built to complete the *in vitro* interventional simulation experiment[170]. Wang *et al*. embedded the rod-shaped IPMC actuators in the soft silicone tube to implement more complex actions [Figure 11F][171]. By incorporating optical fibers, IPMC catheters can also be further used in medical endoscopic devices[172,173]. Feng *et al*. encapsulated an optical fiber on a 4-electrode cylindrical IPMC actuator, allowing electronic directional control of the conducted laser light. The laser beam can be moved in multiple directions by controlling the deformation of the IPMC actuator, which is expected to be used in areas such as microendoscopic eye surgery[173].

(2) Micropumps

In terms of drug delivery, IPMC micropumps are commonly used to pump drugs. A typical micropump driven by IPMC is shown in Figure 11G, where a single piece of IPMC clamped at all edges is adopted as the pumping diaphragm[174]. The bidirectional deformation of the IPMC diaphragm under applied voltage leads to the variation in pump volume and the pumping activity. The IPMC diaphragms in original micropumps were generally disk-like[175,176]. However, the overall deformation of the disk-shaped IPMC is very small, resulting in poor performance. Nam *et al*. proposed a novel design for IPMC diaphragm which can help to produce larger deformation and increase the durability of IPMC actuator[174]. Unlike the conventional design of using a piece of disk-like IPMC clamped at all edges, they used several IPMC actuators (clamped at single edge) to drive elastic thin films. By eliminating the edge constraints for IPMC actuators, the new diaphragm can provide significantly larger deformation. Wang *et al*. proposed a double-chamber valveless pump actuated by an IPMC cantilever[177]. A cantilever IPMC is used as a driver to increase the deformation of the valve, thereby improving the performance of the micropump. Wang *et al*. integrated an inner petal-shaped IPMC as an actuating diaphragm into micropump [Figure 11H], improving the performance of micropump[178]. Besides, some other configurations for micropumps have also been proposed. For instance, Sideris *et al*. proposed a linear peristaltic microfluidic pump, which provides ideas for the design of drug pumps with new configurations[179]. Cheong *et al*. designed a wirelessly activated device with an integrated IPMC cantilever valve for targeted drug delivery, as shown Figure 11I[180]. The IPMC cantilever was integrated with a planar LC receiver circuit consisting of a resonator and load loops, which served as the actuator to control the opening gap of the release orifice using input power from the transmitter.

(3) Other medical devices

In addition to surgical operation and drug delivery, IPMC has several other applications in the field of medical devices. Horiuchi *et al*. designed a voltage-controlled accommodating intraocular lens using an

IPMC actuator to change focus[181]. A soft, clear material is placed in the device as the intraocular lens, which can be deformed by the IPMC actuator. Feng *et al*. presented an innovative digitally controllable tactile bump array driven by IPMC actuators for a refreshable braille display application [Figure 11J][182]. The device is composed of IPMC actuator arrays with a PDMS bump on them. When a voltage is applied, the target bump will be pushed through the printed circuit board by the IPMC actuator, and then pressed against the finger in contact with the device, providing tactile feedback.

Applications of IPMC as sensors

Wearable devices

Because the sensing mechanism of IPMC is similar to that of biological skin, i.e., the transmission of signals is realized by ion migration, IPMC is often used as "artificial skin", which can monitor the life activities of the human body by directly attach to the body surface or prepared into wearable devices. Lee *et al*. attached an IPMC bending sensor to the larynx to monitor human respiration[183]. Through deep learning technology, it can distinguish coughing, humming, nodding, swallowing and other behaviors [Figure 12A]. Chang *et al*. attached an IPMC bending sensor to the human face, which can recognize the micro-expression changes of the face, such as smiling, frowning, *etc*. [Figure 12B][140]. However, this way of attaching directly to the skin is not practical. On the one hand, the way of bonding is easy to cause the sensor to fall off. On the other hand, the wired measurement method makes it impossible to carry. Therefore, in order to monitor human life activities more conveniently, it is a reliable way to design IPMC sensors into wearable devices. For example, Chattaraj *et al*. designed a wearable sensor to record natural auscultations of the wrist artery based on IPMC, which can read the periodic pulse signal, but the wearing comfort can be further improved[184]. Annabestani *et al*. designed a non-rigid air flow active sensor (NAFAS) to detect respiratory patterns, which can also transmit the exhalation signal to the mobile phone in real time through wireless transmission[185]. It can accurately detect weak breathing patterns and has promising features to develop low-cost, wearable and ubiquitous spirometers. Ming *et al*. designed a smart glove based on the IPMC bending sensor, as shown in Figure 12C[186]. The voltage signal is processed and sent remotely by a PCB board, which realizes the wireless of the wearable device. The glove can not only monitor the pulse and identify Braille, but also control the robot hand through remote human-computer interaction [Figure 12D]. Besides, IPMC sensors can also be used in the field of medical surgery, such as biomedical probe[187]. Brunetto *et al*. designed a vibrating tactile probe using an IPMC sensor and an IPMC actuator to realize the recognition of different materials[188]. The actuator forces the system into resonant conditions that change with the mechanical properties of the contacted material. The output signal produced by the IPMC sensor contains the information required for the estimation of the contacting material.

Sensing elements for underwater vehicles

In underwater areas, IPMC sensors can be used for underwater robotic sensing, flowmeters, and so on. Inspired by fish canal lateral line, IPMC can be developed as a differential pressure sensor and used in underwater robots for target detection, angle measurement and source localization. Sharif *et al*. proposed an IPMC differential pressure sensor and integrated it into a robotic fish[189]. The output voltage of the sensor was experimentally characterized as the fish-like body is rotated with respect to a dipole source, which confirmed that the sensor is capable of capturing the pressure difference between the two pores. Jiang *et al*. developed an artificial lateral line system integrated with IPMC pressure sensors and flow velocity sensors to realize underwater source localization of robotic fish, the structure of which is shown in Figure 12E[190]. Shen *et al*. integrated IPMC bending sensors into a biomimetic underwater vehicle [Figure 12F], which can monitor the deformation and oscillation frequency of the wave-driven propeller[191]. Pasquale *et al*. utilized an IPMC sensor to develop a flowmeter based on the vortex-shedding phenomenon[192]. The signal produced by the IPMC sensor can be further manipulated by standard electronics to extract the frequency of the vortex shedding.

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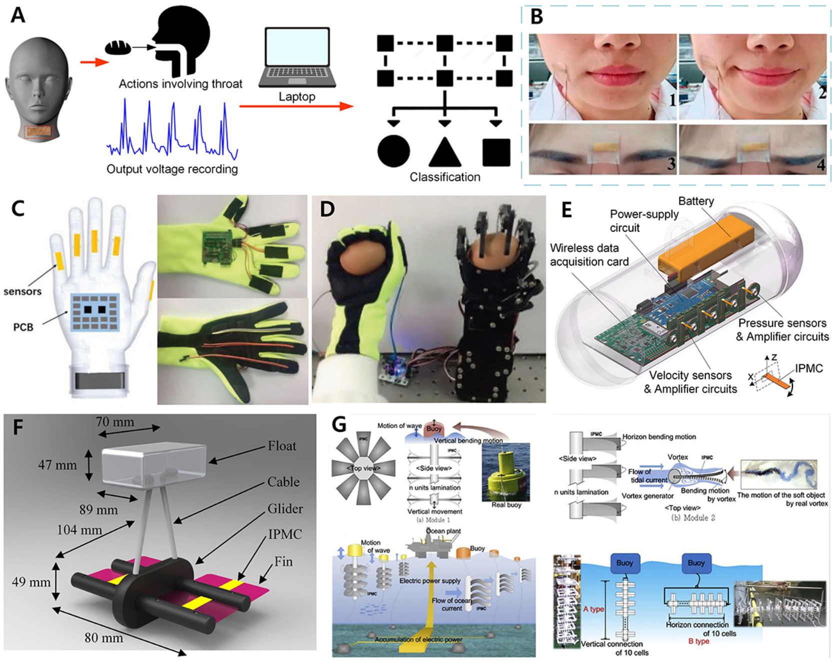

Figure 12. Applications of IPMC as sensors. (A) Monitoring human respiration by attaching an IPMC bending sensor to the larynx. (B) Recognizing the micro-expression changes of the face using an IPMC bending sensor. (C) A smart glove based on the IPMC bending sensors. Reproduced with permission from Ref.[186]. Copyright© 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (D) Control the robot hand through remote human-computer interaction. Reproduced with permission from Ref.[186]. Copyright© 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (E) An artificial lateral line system integrated with IPMC pressure sensors and flow velocity sensors. Reproduced with permission from Ref.[190]. Copyright© 2022, IEEE (F) A biomimetic underwater vehicle with IPMC bending sensors. Reproduced with permission from Ref.[191]. Copyright© 2015 IOP Publishing Ltd. (G) an ocean-based electricity generating system. Reproduced with permission from Ref.[196]. Copyright© 2017 Elsevier B.V.

Energy harvesting

Due to the ability to convert mechanical energy into electrical energy, IPMC also shows good application potential in the field of energy harvesting. For example, Cha *et al*. fixed the IPMC on the tail of the robotic fish to collect the energy generated when the tail swings[193]. Cellini *et al*. fixed the IPMC on the turbine to collect the energy from fluid flow[194]. Patel and Mukherjee installed an IPMC energy harvester on the shoes to collect energy generated by walking[195]. Based on IPMC, Vinh and Kim developed an ocean-based electricity generating system to absorb the kinetic energy of the ocean waves and generate electricity[196]. As shown in Figure 12G, the system consists of a large number of IPMCs in an array. The recorded data of 296 days showed that the average power density generated per day remained stable at about 245 $\mu W/m^2$, and the decrease in the electrical performance of the IPMC was insignificant in the long-term working. They also built a simulated model for the system to protect the IPMC from damage[197].

Generally speaking, IPMC has a variety of applications in both actuating and sensing aspects. However, most of the related applications are still in the laboratory stage, and there is still a certain distance to commercial devices, which may be related to the preparation and performance of IPMC itself.

CONCLUSION AND OUTLOOK

In this review, we conduct a comprehensive survey on the R&D of IPMC in recent years from the aspects of preparation, performance, optimization and application. In terms of preparation, Nafion is still the most commonly used matrix material because of its high level of commercialization and stability, and the emergence of some new ionomers also provides hope for the development of high-performance IPMC. The formation of the electrode can be achieved by both physical and chemical methods. The physical methods have a faster preparation speed, while the chemical methods can form strong and durable electrodes. In terms of performance, the performance of IPMC for actuators and sensors will both be affected by the size, matrix state and electrode morphology. Taking advantage of the influence rule of these factors, the performance of IPMC can be optimized by increasing the interface area and WU, optimizing the external working environment, *etc*. Finally, some applications of IPMC were introduced. IPMC actuators are generally used in the fields of medical surgery and robotics, while IPMC sensors show good potential in the fields of wearable devices and energy harvesting. However, there are still many difficulties and challenges in the development of IPMC. The challenges in the current stage and accordingly prospects are put forward as the following.

1. The preparation process of IPMC has become mature in both physical and chemical aspects, but there is still a lack of large-scale preparation methods, which is one of the important factors limiting the wide application of IPMC. At present, the commonly used matrix Nafion has a high degree of commercialization, so it is urgent to solve the problem of large-scale formation of electrodes. Although chemical methods can be used to prepare high-performance IPMC, the scale of preparation is severely limited due to complex process and difficulty in standardization, while physical methods seem to be expected to open the way for large-scale preparation of IPMC because of their simple processes and good repeatability. In particular, the electron sputtering method for large-scale preparation of IPMC has been preliminarily verified. A standard preparation process and production equipment with low price, high stability and efficiency should be developed in the next stage.

2. Compared with the emerging new smart materials, the performance of IPMC is not outstanding in all aspects, and there are many substitutes, which also makes it difficult for scholars in other disciplines to generate research interest. However, after several years of exploration, we found that multi-functional sensing seems to be a great advantage of IPMC, which can replace a variety of sensors on some occasions and provides an attractive prospect for the application of IPMC sensors. In the next stage of development of IPMC sensors, a key breakthrough direction is the coupling mechanism of multi-sensing functions and the signal decoupling method.

3. Although many scholars are committed to optimizing the actuating performance of IPMC, it is not difficult to find that there is an upper limit, such as its blocking force can only reach the level of 100 millinewtons, which still cannot meet the driving requirements of many robots. In particular, it is difficult to optimize the blocking force and deformation at the same time, and the improvement of one often means the weakening of the other. Therefore, we believe that the appropriate IPMC should be selected for the application scenario; for example, the IPMC with high driving force can be selected for the robot actuator, while the IPMC with larger deformation needs to be selected when used as a medical catheter.

4. At present, the application of IPMC mostly stays in the laboratory stage, and there is still a certain distance to commercial devices, which is related to the difficulty of precise control of IPMC performance. However, the emergence of deep learning and other technologies seems to provide a solution for this, and we have seen that some wireless devices based on IPMC have shown good commercial prospects. Therefore, through the combination of deep learning, artificial intelligence and other technologies, it is expected to develop more stable and practical intelligent devices, which will pave the way for the development of IPMC commercial devices.

5. Due to the increasingly prominent problem of global electronic waste, the environmental friendliness of electronic devices has been paid more and more attention. Because the substrate material of IPMC is non-degradable, the waste IPMC needs to be recycled and treated in a specific way to avoid harm to the environment. In the meantime, the development of degradable substrate materials is also an important research direction for IPMC in the future. Furthermore, in the chemical preparation process of IPMC, a large number of chemical reagents are involved, such as hydrochloric acid, Pd salts, and so on. The waste produced by these chemical reagents has environmental threats and even biological toxicity, and thus complex means are needed to deal with it. In comparison, the physical preparation of IPMC can effectively avoid the generation of chemical waste, so it has better environmental friendliness. In addition, the biocompatibility of IPMC with heavy metals as electrodes is poor, while the safety of IPMC applied in human body can be greatly improved by using non-metallic materials such as graphene as electrodes.

To conclude, ionic actuating and sensing is a novel mechanism that is closest to biological mechanisms. Although there are some drawbacks and problems to be solved at present, we believe that IPMC will certainly show its advantages and play a great role in the future. We also call on more researchers from other disciplines to join us to overcome the key scientific and technical problems that hinder the commercial application of IPMC.

DECLARATIONS

Authors' contributions

Conception and design of the study, data analysis and interpretation: Tang G, Wang Y
Administrative, technical, and material support: Mei D, Zhao X, Zhao C
Language modifying: Li L

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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