



Cite this: *RSC Appl. Polym.*, 2024, **2**, 795

Received 7th March 2024,
Accepted 11th May 2024

DOI: 10.1039/d4lp00084f

rsc.li/rscappliedpolym

Perspective on the development and application of ionic polymer metal composites: from actuators to multifunctional sensors

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Ionic polymer metal composites (IPMCs) are a versatile class of ionic polymer-based transducers with excellent electromechanical properties and wide application potential in the fields of bionic robots, medical devices and wearable devices as actuators and sensors. In this perspective, we discuss the development of polymer matrices and applications of IPMCs, from actuating to multifunctional sensing. The latest ionic polymer matrixes for IPMCs show the trend toward being printable, programmable and degradable, which will facilitate multi-scenario applications of IPMCs. Regarding the applications, IPMCs have been widely used in various fields as actuators and multi-parameter sensors, and related devices will be further developed in the direction of miniaturization, integration and intelligence. Furthermore, some suggestions are put forward to facilitate the further development and wide application of IPMCs.

1. Introduction

IPMCs are a kind of polymer-based smart material with wide application potential in bionic robots, medical devices, wearable equipment and other fields, benefitting from light weight, low actuating voltage, and multifunctional sensing properties.^{1–5} A typical IPMC is composed of two electrodes and an ionomer to form a sandwich structure.⁶ As shown in Fig. 1, when a voltage (generally less than 5 Vpp) is applied to an IPMC on both sides, an electrostatic field will be generated between the electrodes. Microscopically, free cations will migrate to the cathode under the action of the electrostatic field, and the IPMC will bend towards the anode caused by uneven stress in the thickness direction.⁷ Conversely, in response to external stimuli, such as changes in humidity or deformation, internal cations inside the IPMC will migrate in a specific direction under the action of the stress or concentration gradient, resulting in a detectable electrical signal between the two electrodes.⁸ Owing to their ability to convert mechanical energy to electrical energy in both directions, IPMCs can be used as actuators and sensors.

Compared with other actuators like shape memory materials (SMS) and dielectric elastomers (DEs), IPMCs can produce larger deformations at lower driving voltages. Besides, compared with other sensors, such as resistive, capacitive and piezoelectric sensors, IPMCs rely on the migration of ions to achieve signal transmission, which is similar to the trans-

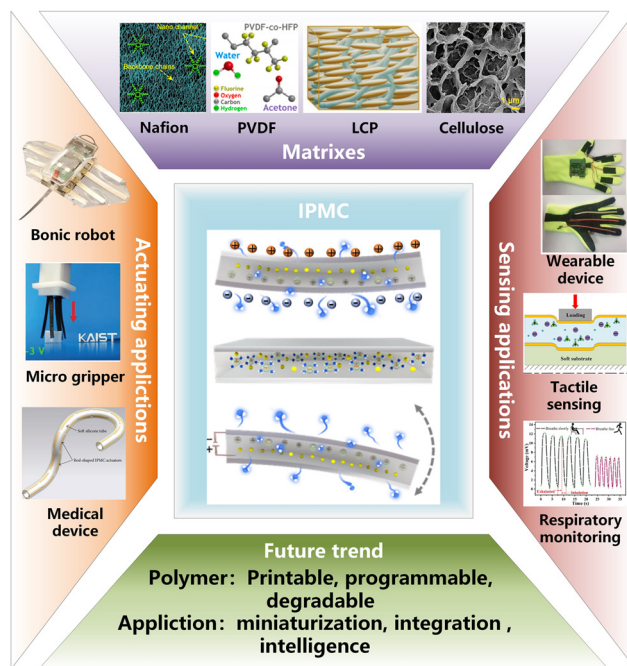


Fig. 1 Framework of this perspective. Nafion, free copyright 2018, Mu et al.¹⁷ PVDF, reproduced with permission.²¹ Copyright 2021, Elsevier. LCP, reproduced with permission.²⁶ Copyright 2019, Wiley. Cellulose, reproduced with permission.³³ Copyright 2021, Elsevier. Bionic robot, reproduced with permission.⁴⁰ Copyright 2011, Elsevier. Micro gripper, reproduced with permission.⁵⁶ Copyright 2017, Wiley. Wearable device, reproduced with permission.⁴ Copyright 2018, Wiley. Tactile sensing, reproduced with permission.⁷² Copyright 2022, IOP Publishing Ltd. Respiratory sensing, reproduced with permission.⁷⁷ Copyright 2023, American Chemical Society.

mission principle of biological signals and promotes the application of IPMCs in the field of biomimetics.

Since they were first proposed in 1992,⁹ the development of ionic polymer matrixes has been a focus of IPMC research. Because the actuating and sensing performance of IPMCs depends on the cation migration in the ionomer, the type of polymer matrix plays a decisive role in the performance of IPMCs. In addition to the most commonly used Nafion matrix, sulfonated poly(ether sulfone) (SPEES),¹⁰ sulfonated polyether ether ketone (SPEEK),¹¹ sulfonated styrenic pentablock copolymer ionomer (SSPB),¹² sulfonated polyvinyl alcohol (SPVA),¹³ and other ionic polymer matrixes have been developed for the preparation of IPMCs. These ionomers have advantages and disadvantages in terms of ion-exchange capacity (IEC) and water uptake (WU). However, in general, it is difficult for these polymers to replace Nafion for wide application because of its commercialization and accessibility. Herein, we focus on the latest ionomers for the IPMC, including polyvinylidene difluoride (PVDF), liquid crystalline polymers (LCPs) and cellulose, which represent three future trends of polymers for IPMCs, namely, printable, programmable and degradable.

Regarding the applications, IPMC actuators are mainly used in bionic robots, soft grippers, medical devices and other fields owing to their inherent characteristics such as low actuating voltage and large deformation. Meanwhile, IPMC sensors are used in the fields of human motion detection, tactile sensing and gas perception because of their multi-parameter sensitivity to deformation, pressure and gas. Herein, we provide an overview of the most recent application developments in the fields of actuation and sensing, as well as a discussion of future development trends including miniaturization, integration and intelligence.

In this perspective, we analyzed the ionomer development and applications of IPMCs as actuators and sensors; the outline is shown in Fig. 1. First, commonly used and recently developed polymer membranes for IPMC preparation are introduced, together with a discussion of their applications. Then, the typical application fields and application status of IPMCs as actuators and sensors are summarized. Finally, future development trends of IPMCs are discussed and some suggestions are put forward for their further development.

2. Potential IPMC matrices

2.1 Nafion

As a typical kind of perfluorosulfonic acid ion-exchange membrane, NafionTM is the most commonly used matrix membrane owing to its stable physical and chemical properties and high ion capacity.¹⁴ As shown in Fig. 2a, the main structure of Nafion is formed by the polymerization of the C–F long chain with the sulfonic acid group at the end of the side chain. The sulfonic acid group can be agglomerated to form a nanochannel, as shown in Fig. 2b, in which free cations and water molecules can migrate in a specific direction.^{15,16} Therefore, the interior of Nafion presents a microporous structure¹⁷ (Fig. 2c),

which also facilitates the rapid migration of ions. Because of the high commercialization, Nafion has been widely used in the preparation of IPMC actuators and sensors. To improve the stability and working time of Nafion-IPMC actuators, an ionic liquid is always used as the inner solvent, while for Nafion-IPMC sensors, water is generally used as the internal solvent because of its lower viscosity, which endows the sensors with a fast response speed and higher sensitivity. However, as a kind of cation exchange membrane, Nafion has low IEC, high elastic modulus and high cost. Therefore, some researchers have tried to develop new ionic polymers as an alternative to Nafion, and the earliest research included sulfonated SPEES, SPEEK, SSPB, and SPVA. Most of these polymers have been used to develop IPMC actuators, but the replacement of Nafion for large-scale applications has not been realized due to the complexity of the preparation process. In recent years, some new ionic polymer matrices have been developed for the preparation of IPMC actuators and sensors, such as PVDF,¹⁸ LCPs¹⁹ and cellulose.²⁰

2.2 PVDF

Like Nafion, PVDF is another fluoropolymer with excellent stability and mechanical strength, and its chemical formula is shown in Fig. 2d. When used to prepare ionic polymer matrices, PVDF is often copolymerized with hexafluoropropylene (HFP) to improve the robustness of the membrane.²¹ A typical PVDF-based IPMC is shown in Fig. 2e. PVDF is not hygroscopic, so generally two kinds of methods are used to endow it with ion-exchange capacity. One is to compound PVDF with a hydrophilic material and use water as the internal solvent, such as by attaching the hydrophilic functional groups to the PVDF backbone through radiation grafting.²² The other is to directly mix and copolymerize an IL with PVDF to form an ionic polymer matrix, which can be used to realize direct 3D printing of a matrix film (Fig. 2f).²³ Compared with Nafion, PVDF-based ionic membranes have higher IEC and lower elastic modulus, so they can be used to prepare high-performance IPMC actuators. However, the internal ion migration resistance of PVDF is usually high, so when used as a sensor, its response speed and sensing performance are inferior to those of Nafion-based IPMCs.²⁴

2.3 LCPs

LCPs are generally achieved by the alignment of liquid crystal mesogens in a polymer network.²⁵ Because the liquid crystal mesogens are not tightly bound together, there are inherent nanochannels in LCPs, and ionic LCPs can be obtained by introducing ILs into these nanochannels.²⁶ The electrodeformation of ionic LCP-based IPMCs depends on the anisotropic arrangement of the liquid crystal, mainly including planar, hybrid and homeotropic alignments, as shown in Fig. 2g. At present, ionic LCPs for IPMCs commonly use columnar liquid crystals, as shown in Fig. 2h, which can form a layered ordered structure by self-assembly method.²⁷ 1D and 3D ion channels can be effectively formed by different self-assembly methods, which can modify the performance of the matrix.^{28,29} It is worth mentioning that ionic LCPs are a kind of stretchable





Fig. 2 Ionic polymers matrices for IPMCs. a. Molecular formula of Nafion. b. Structure of Nafion. c. Nano channels of Nafion. Free copyright 2018, Mu *et al.*¹⁷ d. Molecular formula of PVDF. e. PVDF-based IPMC. Reproduced with permission.²¹ Copyright 2021, Elsevier. f. 3D printing of PVDF matrix film. Reproduced with permission.²³ Copyright 2021, Elsevier. g. Planar, hybrid and homeotropic alignments of LCPs. Reproduced with permission.²⁶ Copyright 2019, Wiley. h. Columnar liquid crystals. Reproduced with permission.²⁷ Copyright 2022, American Chemical Society. i. Stretchable LCP membrane. Reproduced with permission.²⁷ Copyright 2022, American Chemical Society. j. Molecular formula of cellulose. k. Porous structure of cellulose. Reproduced with permission.³³ Copyright 2021, Elsevier. l. Spiral winding of cellulose-based IPMC. Reproduced with permission.³⁶ Copyright 2021, Wiley.

membrane, as shown in Fig. 2i, which makes up for the poor stretchability of most IPMCs.²⁷ The stretchability will improve the matching and adaptability of IPMCs with other interfaces and help the development of high-performance IPMC sensors, although most of the ionic LCP-based IPMCs are used as actuators. In terms of sensing, Rajapaksha *et al.* preliminarily explored the relationship between their electrical response signals and deformation.³⁰ Further studies are needed to explore the potential of ionic liquid crystalline polymers as high-performance sensors.

2.4 Cellulose

With the development of green flexible electronics, there is increasing demand for environmentally friendly electronic devices, so some scholars have attempted to develop IPMCs using degradable ionic polymer membranes. Cellulose, the most abundant natural polymer, has attracted great attention owing to its high aspect ratio, good dispersion, and excellent

biodegradability and biocompatibility.^{31,32} The molecular formula of cellulose is shown in Fig. 2j, and there are a large number of $-OH$ groups on its polymer chain, which makes it easy to combine with electrolytes such as ILs. Besides, the interior of the cellulose polymer exhibits an intrinsic porous structure (Fig. 2k), which also provides a pathway for the directional migration of ions.³³ Compared with Nafion, it is easier to achieve ultra-thin structures using cellulose-based ionic polymers, and they have a lower elastic modulus, so the actuating voltage of cellulose-based IPMC is lower, potentially as low as 1 V or even 0.5 V.^{34,35} In addition, cellulose-based IPMCs have good plasticity, and some complex actions, such as spiral winding, can be achieved by heating and shaping (Fig. 2l), which is difficult to achieve with Nafion-based IPMCs.³⁶

2.5 Others

In addition to the above four matrixes, chitosan, a natural macromolecular polysaccharide, is another biodegradable



Table 1 Comparison of different matrixes for IPMCs

Matrixes	Properties			Advantages	Limitations	Ref.
	IEC (mMol g ⁻¹)	Conductivity (mS cm ⁻¹)	Bearing voltage (V)			
Nafion	0.91	23	2.5	1. High degree of commercialization 2. Good chemical stability	1. Low IEC 2. High cost	14
PVDF-PSSA	1	85	4	1. Printable 2. High conductivity	1. Require hydrophilization treatment	24
LCP	—	10	2	1. Programmable 2. Low voltage driving	1. Small deformation	26
Cellulose	1.95	0.06	4	1. Degradable 2. High IEC	1. Low conductivity	33

material that is commonly used in the development of ionic polymer matrixes. Similar to cellulose, chitosan also forms an ionic polymer matrix through copolymerization with an IL.³⁷ Chitosan-based ionomers can be used to prepare IPMC actuators and sensors. When used as a sensor, it shows a larger response voltage and better high-frequency response characteristics than the traditional Nafion matrix.³⁸ However, its performance is inferior to that of the traditional IPMCs when used as an actuator.³⁹

The comparison of different matrixes for IPMCs is summarized in Table 1, including advantages, limitations and properties like ion-exchange capacity (IEC), conductivity and bearing voltage.

3. Typical applications of IPMCs

3.1 IPMCs as actuators

3.1.1 Underwater robots. When used as an actuator, IPMCs can only be used to actuate microminiature devices owing to the limitation of small size and low output force. Because the actuating of IPMCs is strongly dependent on the internal water, underwater robots are one of the most important directions of IPMCs. Traditional underwater vehicles mostly rely on motors to drive the propeller to generate propulsion, which has the characteristics of high noise, disturbance and energy consumption. Because of the large deformation and sheet structure that can well simulate underwater propulsion action, such as fin swing, IPMCs have become one of the most important actuating materials for bionic underwater robots. In the actuating of bionic fishes, IPMCs can simulate the flapping of the pectoral fin and swinging of the caudal fin, as shown in Fig. 3a and b.^{40–42} In general, pectoral fin flapping has good motion stability, while caudal fin swinging has high propulsion speed. Therefore, these two methods can also be combined to achieve high mobility and high stability together, as shown in Fig. 3c.⁴³ In addition, IPMCs can be used to actuate other underwater bionic robots, such as bionic jellyfish and bionic turtles.^{44,45} For underwater robot actuating, because the gravity of the robot can be counteracted by buoyancy adjustment, the movement of the robot is mainly affected by the resistance of the water. To improve the motion performance of

the robot, the overall shape of the robot and structure of the IPMC actuator can be optimized to reduce the water resistance.

3.1.2 Crawling robots. In addition to underwater robots, IPMCs can be used to actuate some crawling robots. The most typical approach is to directly design the IPMC as the legs of the robot to construct a quadruped and multi-legged walking robot, as shown in Fig. 3d.^{46,47} By actuating these IPMC legs in a distributed way, the robot can move forward and turn. In addition, IPMCs can be used to mimic the crawling of some soft-bodied insects owing to their bend-actuating nature. For example, the bending deformation of an IPMC is directly used to simulate the “Ω” type motion of an inchworm, as shown in Fig. 3e.⁴⁸ As shown in Fig. 3f, the grasping and loosening of a caterpillar on a plant stem is simulated by the opening and closing of the semi-annular IPMC, and the straight movement is realized by the tensioning movement of a group of annular IPMCs.⁴⁹ Owing to low-frequency actuating characteristics, low stiffness and low output force of IPMCs, walking robots based on IPMCs usually have small size, weight and speed, which also makes it difficult to integrate the control module on the robot body. In view of the above problem, Hu *et al.* integrated an IPMC actuator into a bistable mechanism, as shown in Fig. 3g and used the IPMC to actuate the bistable structure to quickly release and drive the robot to move, which increased the driving power of the IPMC by 280 times.⁵⁰ This method provides a new idea for the wireless and full integration of small robots actuated by IPMCs.

3.1.3 Micro grippers. Benefitting from their large deformation at low voltages, IPMC actuators can also be used to develop miniature grippers for low voltage actuation. By changing the number of IPMC actuators, single-active-finger grippers and multi-active-finger grippers can be designed, and the number of active fingers can range from one to more than four.^{51–54} Typical two-finger and four-finger grasping processes are shown in Fig. 3h and i.^{55,56} With the increase of the number of fingers, the envelopment and stability of grasping will gradually increase. However, multi-finger grippers can only grasp blocky objects well, and it is difficult to grasp some columnar objects. To solve this problem, the IPMC can be programmed to simulate the spiral winding motion of a vine, as shown in Fig. 3j, so as



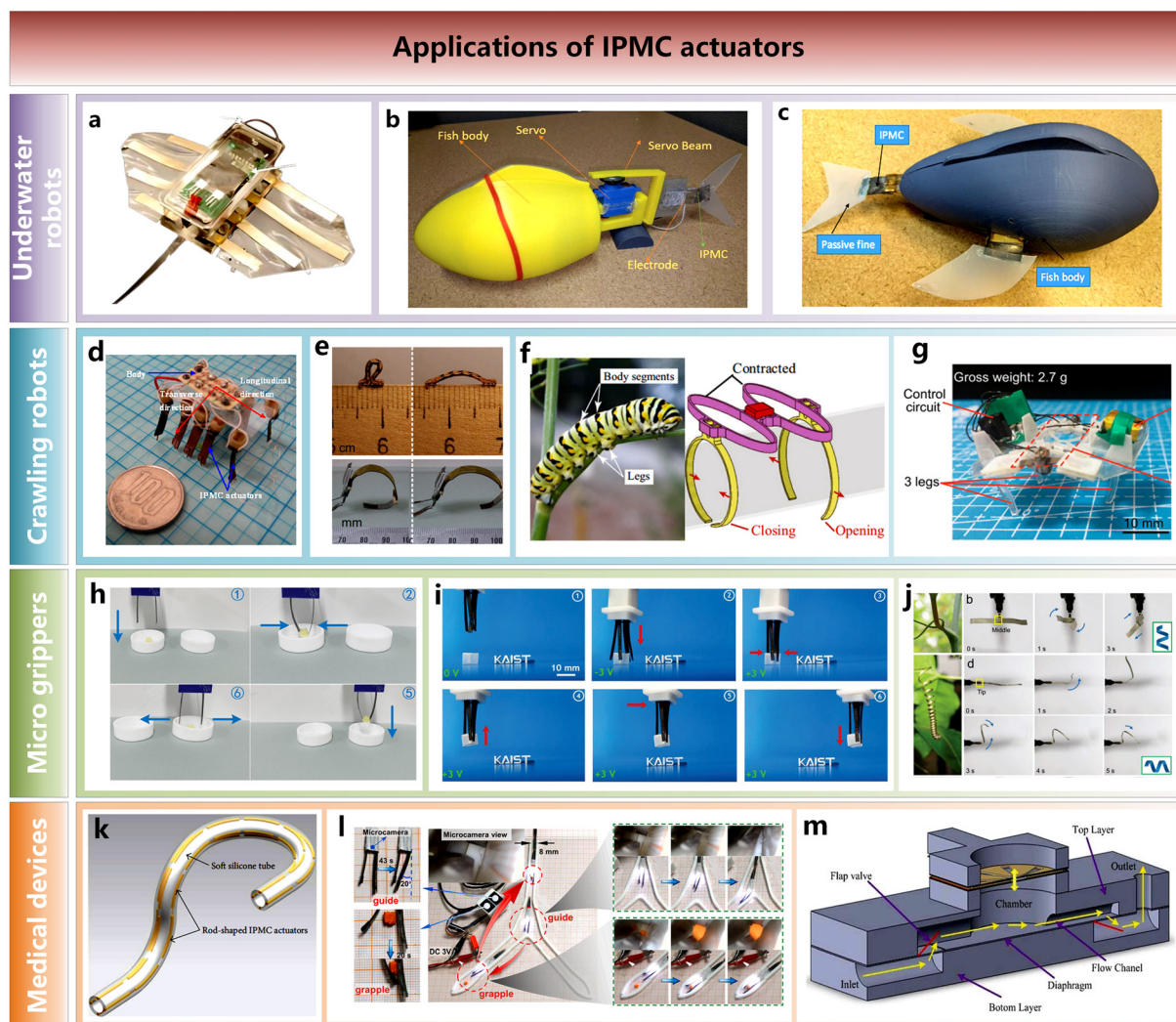


Fig. 3 Applications of IPMCs as actuators. a. Bionic fish actuated by IPMC pectoral fin. Reproduced with permission.⁴⁰ Copyright 2011, Elsevier. b. Bionic fish actuated by IPMC caudal fin. Reproduced with permission.⁴² Copyright 2019, ASME. c. Bionic fish actuated by both IPMC pectoral and caudal fins. Reproduced with permission.⁴³ Copyright 2017, Springer. d. Multi-legged walking robot. Reproduced with permission.⁴⁷ Copyright 2012, Elsevier. e. Crawling robot like inchworm. Reproduced with permission.⁴⁸ Copyright 2012, Wiley. f. Crawling robot mimicking the wriggling of a caterpillar. Free copyright 2019, Carrico et al.⁴⁹ g. Robot based on a bistable structure actuated IPMC. Free copyright 2019, Hu et al.⁵⁰ h. Two-finger grasping process. Reproduced with permission.⁵⁵ Copyright 2022, IOP Publishing Ltd. i. Four-finger grasping process. Reproduced with permission.⁵⁶ Copyright 2017, Wiley. j. Spiral winding grasping. Reproduced with permission.⁵⁷ Copyright 2023, Wiley. k. Tube with IPMC embedded. l. Surgical simulation of an IPMC end effector. Reproduced with permission.⁵⁸ Copyright 2023, Elsevier. m. Typical IPMC-based micropump.

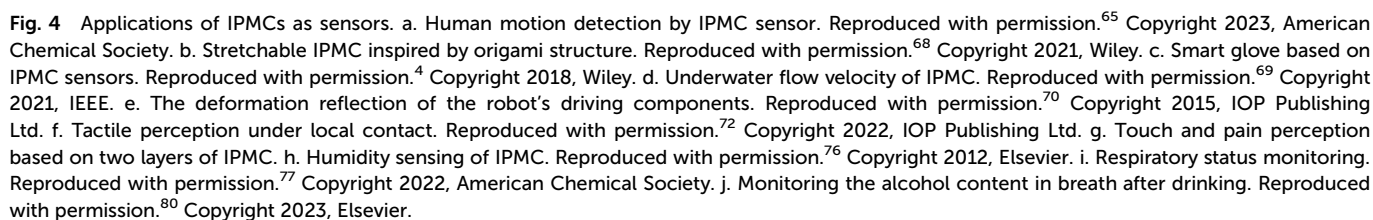
to grasp a columnar object.⁵⁷ Micro-grippers based on IPMC actuators usually have a very small mass and can only grasp very light objects. Therefore, it may have some potential applications in the field of laboratory fine operation and aerospace, especially in manned spaceflight, where the development of lightweight tools in a microgravity environment will become more and more important.

3.1.4 Medical devices. In addition, because of the low actuating voltage and good compliance, medical devices are another important application direction of IPMCs, especially in surgical operation and drug delivery. In terms of surgical operation devices, IPMC actuators can be used as medical catheters and end effectors.³ When applied in medical catheters, the

IPMC can be embedded in some tubes with low elastic modulus (Fig. 3k)⁵⁸ or directly adhered to the outer wall of the catheter,⁵⁹ and the movement of the catheter can be controlled by actuating the IPMC actuator. Some scholars have directly prepared a catheter using an IPMC,⁶⁰ which is more convenient to actuate, but it is still challenging to obtain large-sized devices. An end effector is generally installed at the end of the catheter, and when the catheter reaches the specified position, some surgical tasks can be conducted through the end effector.⁶¹ Fig. 3l shows the surgical simulation of an IPMC end effector, which can complete the task of removing foreign bodies by cooperating with the camera and other devices.⁶² In addition, drug delivery is a very important application direction for IPMCs, which can be



human motion detection and robot motion feedback. Because of their low elastic modulus and good flexibility, IPMCs can be easily attached to various parts of the human body, such as the wrist, knee, face, throat, and sole and further reflect the bending of joints, changes in facial expressions, vibration of vocal cords when speaking, and changes in gait when walking, as shown in Fig. 4a.^{65–67} Because IPMCs are not stretchable, they will produce an interface mismatch with the skin surface in the case of large deformation. To address this issue, Lee *et al.*⁶⁸ cropped an IPMC, inspired by the origami structure, to produce a strain of 80%, as shown in Fig. 4b. However, the stability is poor when the IPMC is directly attached to the skin and it may cause skin problems due to poor air permeability. Therefore, it is a better practice to make IPMCs into wearable



devices. Ming *et al.*⁴ integrated an IPMC sensor into a glove, which can not only recognize the gesture, but also control the movement of the robot hand through wireless signal transmission to realize remote human-computer interaction, as shown in Fig. 4c. When used as the sensing component of a robot, the IPMC can either sense the external environment (such as the flow velocity underwater, Fig. 4d⁶⁹) or reflect the deformation of the driving components of the robots (Fig. 4e⁷⁰) through its deformation.

3.2.2 Pressure sensing. In terms of pressure sensing, the application of IPMCs is mainly focused on tactile perception. In addition to being used for contact force perception,⁷¹ owing to the dynamics of ion migration inside the ionomer, it is also possible for IPMCs to distinguish more complex contact characteristics under local contact, including contact area and shape,^{72,73} as shown in Fig. 4f. This multi-modal tactile perception function may facilitate the application of IPMCs in the field of soft grasping. In addition, by laminating two pieces of IPMC and a layer of PDMS, different sensitivity and force sensing ranges can be achieved by the two IPMCs (Fig. 4g).⁷⁴ It can simulate human touch and pain perception, further enrich the function of artificial skin and help realize robots with human-like perception functions.

3.2.3 Gas sensing. In terms of gas sensing, IPMCs are mainly used for humidity (moisture) sensing.⁷⁵ As shown in Fig. 4h, when one side of the IPMC absorbs water molecules, the increase of solvent content (water) on this side will cause the decrease of cation concentration and increase the difference of cation concentration between this side with the other side, which leads to cation migration and the generation of an electrical signal.⁷⁶ Based on the response of IPMCs to humidity, they can be used to monitor human respiratory status, as shown in Fig. 4i.⁷⁷ Annabestani *et al.*⁷⁸ developed a wireless respiratory sensor based on an IPMC, which can monitor the respiratory status in real time and display it remotely on a mobile phone. In addition to moisture, IPMCs can detect gases such as amines, carboxylic acids, alcohols, and ketones.⁷⁹ Previously, our group used an IPMC to detect the ethanol content in a mixed solution of ethanol and water, and this function can also be used to monitor the alcohol content in breath after drinking, as shown in Fig. 4j.⁸⁰

The ability to respond to multiple physical parameters is an important advantage of IPMC sensors, which endows them with the potential for a wide variety of sensing applications.

Currently, most applications of IPMCs are in the laboratory stage. For applications in underwater vehicles, the main limitation is that the output force of IPMCs is small, so they cannot drive large-scale underwater devices. For non-underwater driving applications, including crawling robots, micro grippers and medical devices, the dehydration of IPMCs hinders their long-term operation, which greatly affects the durability of many devices. For real-world applications, an extremely sophisticated packaging process needs to be developed. In the applications of IPMC sensors, the challenge is the shift of the ionic signal, which brings great difficulties to deep learning and other technologies. Therefore, it is necessary to extract truly

effective signal features through time-frequency conversion and other methods.

4. Summary and perspective

In summary, we discussed the polymer development and applications of IPMCs as actuators and sensors in this perspective. Generally speaking, Nafion is the most widely used polymer membrane for IPMCs. However, many new polymer matrixes have been developed to make IPMCs applicable to more scenarios, where PVDF is suitable for high-performance IPMC actuators and is easy to realize 3D printing. LCPs can be used for stretchable sensors and can be programmable due to the self-assembly preparation method. Cellulose can be used to develop environmentally friendly IPMCs because of its degradable characteristics. It is not difficult to find that printability, programmability and degradability are three important trends of polymer development for IPMCs in the future.

The applications of IPMC actuators are mainly focused on bionic robots, micro grippers and medical devices because of their inherent characteristics of large deformation and low output force. Owing to the characteristics of multi-parameter sensitivity and ion dynamic migration, IPMC sensors have wide application potential in many fields, including human motion detection, environmental perception for robots, tactile sensing and detection of gas. In general, the applications of IPMC actuators are relatively mature, while those of IPMC sensors are still in the developmental stage. From the application status of IPMCs, miniaturization, integration and intelligence are three very important development trends.

To facilitate further development and wide application of IPMCs, we identified following challenges and outlined some potential solutions:

(1) Existing IPMC preparation methods, such as chemical methods, usually require a long time, and the performance of different batches of IPMCs is difficult to guarantee, which is not conducive to the large-scale application of IPMCs. Therefore, the most important thing at this stage is to develop a fast and stable IPMC preparation process. At present, 3D printing and printed electronic technology are two mature processing methods, which provide the possibility for the rapid and stable preparation of the matrix and electrode of IPMCs, respectively.

(2) Although centimeter-sized IPMCs with thickness in the range of tens of microns have been achieved, some demanding applications, such as nano-micro manipulation, may require devices as small as millimeter size. Therefore, the miniaturization of IPMC devices is crucial for expanding their applications, which requires the miniaturization of the IPMC itself, the associated circuitry, and electronic components. In this regard, micro-nano processing and MEMS technology may provide a certain degree of help.

(3) Although IPMCs have achieved wide applications as actuators and sensors, these applications are still in the laboratory stage and far away from commercialization. The main



reason is that these devices are not integrated and intelligent enough. Therefore, the next crucial step for IPMC prototypes is to integrate wireless transmission technology and achieve high-precision intelligence using deep learning and AI technology.

(4) Owing to the increasingly prominent problem of global electronic waste, waste IPMCs need to be recycled to avoid harm to the environment. In this regard, the physical preparation of IPMCs can effectively avoid the generation of chemical waste and has good environmental friendliness. Besides, developing IPMCs using some degradable substrates (*e.g.* cellulose) and degradable electrodes (*e.g.* squid ink) will effectively reduce the generation of hazardous waste and facilitate the recycling of IPMCs.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This research was supported by the Ministry of Education Joint Fund (8091B032250) and Fundamental Research Funds for the Central Universities (B240205045). The authors gratefully acknowledge the support.

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