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REVIEW ARTICLE

Review on Improvement, Modeling, and Application of Ionic Polymer Metal Composite Artificial Muscle

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Abstract
Recently, researchers have concentrated on studying ionic polymer metal composite (IPMC) artificial muscle, which has numerous advantages including a relatively large strain under low input voltage, flexibility, high response, low noise, light weight, and high driving energy density. This paper reports recent developments in IPMC artificial muscle, including improvement methods, modeling, and applications. Different types of IPMCs are described, along with various methods for overcoming some shortcomings, including improvement of Nafion matrix membranes, surface preparation of Nafion membranes, the choice of high-performing electrodes, and new electro-active polymers for enhancing the properties of IPMCs. IPMC models are also reviewed, providing theoretical guidance for studying the performance and applications of IPMCs. Successful applications such as bio-inspired robots, opto-mechatronic systems, and medical engineering are discussed.

Keywords Ionic polymer metal composite · Artificial muscle · Improvement · Model · Bio-inspired application

1 Introduction

An artificial muscle is defined as a material or device that can change shape after being subjected to external physical or chemical stimulation. Artificial muscles include not only new intelligent shape memory materials that mimic the actual animal muscle structure through biotechnology, but also actuators powered by electricity, magnetic energy, or chemical energy [1–3]. Compared with traditional motor driving, artificial muscles have many advantages, such as versatility, a high power-to-weight ratio, and a high stress-to-weight ratio, without the need for complicated connection devices [4–7]. Over the past 30 years, artificial muscle has shown great potential in applications such as bionic robots, robotic prostheses, exoskeletons, medical robots, and soft robots [8–10].

Ionic electro-active polymers (EAPs), such as carbon polymer composites and ionic polymer metal composite (IPMCs), have recently drawn much attention because these polymers exhibit a low actuation electrical voltage, large strain, light weight, noiseless operation, and flexibility similar to that of natural muscle [11, 12]. An IPMC is composed of an ionic exchange polymer chemically plated by a noble metal, such as palladium (Pd), platinum (Pt), or gold (Au) [13–15]. The strain generated by an IPMC is attributed to the migration of hydrated cations in the ionic exchange film under the input potential [10, 16]. Due to the above stated advantages, IPMCs can be applied in bio-inspired/soft robotic actuators, artificial muscles, and sensors [17–20].

In this paper, to provide a more comprehensive description of the research status of IPMC flexible drivers, improvements in IPMC performance, various IPMC models, and IPMC applications in bio-inspired robots, opto-mechatronic systems, and medical engineering are reviewed.

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The second section of this article primarily describes methods for improving the IPMC artificial muscle performance. Section 2.1 introduces a modification process for the Nafion matrix membrane to improve the comprehensive driving performance of IPMC artificial muscle from the perspective of internal mechanisms and material structure. In Sect. 2.2, a surface roughening process for the Nafion matrix film is introduced, which further improves the driving performance of IPMCs by changing the external influential factors of the Nafion film. Section 2.3 primarily focuses on improvements to the IPMC electrode, because the electrode structure, electrode morphology, deposition method, and metal electrode type strongly influence the driving and sensing performance of IPMCs. This section also analyzes current work on electrode optimization for IPMCs from the perspective of IPMC electrodes. Section 2.4 briefly introduces the current research and status of composite electrodes and novel ionic actuators.

The third section gives an overview of some IPMC models, including the black box model, gray box model, and white box model, which are constructed from macroscopic and microscopic perspectives, respectively.

The fourth section introduces the application and exploration of IPMC artificial muscle in the world. This section summarizes current applications and explorations from the perspective of bionic robots, photoelectric systems, and biomedical fields.

At the conclusion of this paper, the research status of IPMCs is summarized, and future prospects are described. Future research directions and difficulties for IPMCs are also discussed. We hope that this paper will provide readers with a comprehensive understanding of the research status of IPMCs. This paper can also provide reference value for future investigations and applications of IPMCs.

2 Current Research on Improving IPMC Performance

IPMC applications have been limited due to the low output force (several mN) and short effective air lifetime of IPMCs; therefore, many investigators have sought to enhance the output force and air lifetime of IPMCs. The driving performance of IPMCs is related to numerous parameters, such as the physical dimensions of the IPMC [21, 22], distribution of metal electrodes [10, 23], cation type and concentration [24], water content [25], surface preparation of the Nafion membrane [26], interface area between the metal electrodes and polymer matrix [27], and electrolyte solvent [28, 29].

2.1 Improvement of the Nafion Matrix Membrane

The Nafion matrix plays a significant role in IPMC actuators. The electromechanical performance of IPMC actuators is influenced by the elastic modulus, conductivity, capacitance, water content, migration channel, and cationic degree of hydration of the Nafion matrix membrane. Therefore, it is necessary to improve the performance of the Nafion matrix, and consequently, increasingly more researchers have focused on this area.

The most commonly used Nafion membrane is the commercial thin (50–200 μm) membrane prepared by the Dupont Company in the United States. Practical applications of this membrane have been limited by a low output force (only several mN). Kim and Shahinpoor fabricated Nafion membranes with different thicknesses by casting a polymer solution to obtain IPMCs of different thicknesses. The IPMC output force increased dramatically with increasing thickness, but the output displacement was reduced [21]. Lee et al. [30] and Bonomo et al. [31] also reported Nafion membranes with varying thicknesses, obtained by hot pressing a number of commercial Nafion membranes; their results were similar to those of Kim and Shahinpoor. He et al. studied the modulus and current characteristics of Nafion matrices with different thicknesses [32]. Zhao et al. improved IPMCs by applying a thickness gradient [22].

The Kanno model indicated that an IPMC under an input voltage generates electricity, causing internal stress, which results in a volume change and bending deformation of the IPMC [33]. Electricity plays a critical role in IPMC actuators; therefore, it is essential to simultaneously enhance the electricity, output force, and displacement or to seek a balance among these factors to boost the comprehensive performance of IPMCs. Nanoparticles can be introduced to the polymer to improve the mechanical properties, thermal stability, conductivity, and versatility of the matrix [11]. To prepare IPMC actuators with high performance, nanoparticles, such as carbon nanotubes [34–36], fullerenes [37, 38], silver nanoparticles [39], silicate [40, 41], oxidized graphite [42], graphene [43], polypyrrole/aluminum oxide [25], or carbon nanocages [17], have been introduced into the Nafion matrix membrane to enhance the membrane's physical and chemical properties. One-dimensional carbon nanotubes have received much attention owing to their light weight, perfect hexagonal structure, and excellent electrical, mechanical, and chemical properties. Gojny et al. found that Multi-walled Carbon Nanotubes (MWCNTs) have greater potential than Single-walled Carbon Nanotubes (SWCNTs) for enhancing electrical conductivity, because MWCNTs can be more easily dispersed in the polymer matrix [44]. Consequently,

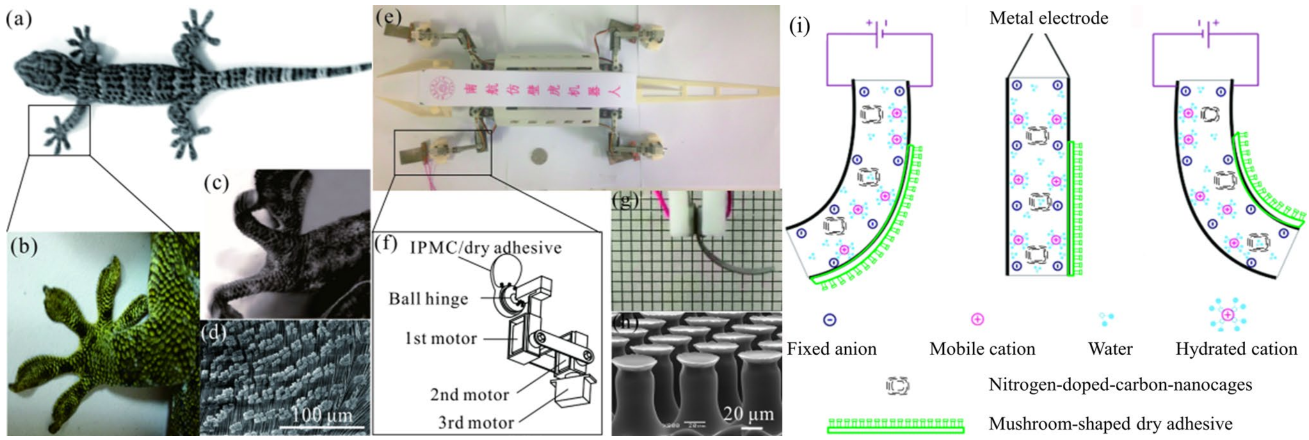

Fig. 1 The IPMC-based electro-active adhesive inspired by the gecko toe by He et al. [17]. **a**, **b** Gecko's toes. **c**, **d** Bristle structures on the toes of geckos. **e** Gecko-like robot with IPMC as toes. **f** Structure diagram of the gecko-like robot's leg. **g** IPMC actuator based on nitrogen-doped carbon nanocages enhancement. **h** Adhesive material similar to gecko bristle structure. **i** Schematic diagram of IPMC driver with dry adhesive material

MWCNT–polymer composites have been widely used in different technical fields [34, 45–47]. Strong van der Waals forces are present in carbon nanotubes, resulting in nanotube agglomeration. Methods for avoiding agglomeration include ultrasonic treatment, rapid mixing, and surface functionalization of the carbon nanotubes (covalent and non-covalent functionalization) [48–50]. Lee et al. used MWCNTs as an additive to prepare IPMCs. Carbon nanotubes were boiled in a mixture of concentrated sulfuric acid and nitric acid, providing a better dispersion in water or ethanol [51]. Lian et al. examined methods for preparing functional carbon–Nafion complex actuators and the resultant electrical and mechanical properties. Carbon nanotubes were oxidized and then esterified by polyethylene glycol [52]. The above studies suggest that carbon nanotubes enhance the performance of IPMCs, but functionalizing the carbon nanotube surface via chemical oxidation inevitably produces defects, degrades the electrical and mechanical properties, and inhibits the development of IPMCs [53]. He et al. investigated the use of surfactant-treated MWCNTs in Nafion, which preserved the excellent properties of MWCNTs and improved the actuation performance of IPMCs compared with acid-treated MWCNTs [36]. He et al. studied the effects of the addition of nitrogen-doped carbon nanocages on IPMC actuation properties and further adopted this approach to develop an IPMC-based electro-active adhesive inspired by a gecko toe (Fig. 1) [17].

Nguyen et al. developed a multilayer Nafion membrane by applying continuous casting on the basis of a nanoparticle-reinforced matrix polymer comprised of a silica-enhanced Nafion matrix sandwiched between two montmorillonite and silver nanopowder-modified Nafion matrices. More metal cations were introduced by the silica and montmorillonite, and the conductivity of the silver was six-fold higher than that of platinum. The surface conductivity of the above IPMC was greatly improved by the higher conductivity of silver, generating a high current and energy storage capacity. The IPMC provided an output force of 6 or 9 gf under a direct current (DC) voltage of 2 or 3 V, respectively. The IPMC also exhibited a better output displacement and response time than an IPMC composed of a pure Nafion film, with no relaxation phenomena [54]. He et al. successfully prepared a multilayered Nafion structure via a consecutive casting method. The structure consisted of a primary Nafion/Tetraethyl Orthosilicate (TEOS) layer sandwiched between two Nafion/MWCNT layers [55]. Lee et al. fabricated a Nafion membrane with three layers of functionalized nanomaterials by applying a continuous casting method. The layers on each side were Nafion membranes enhanced with polypyrrole/alumina, and camphor sulfonic acid was introduced in the middle Nafion membrane. The redox reaction of polypyrrole and the hygroscopicity of alumina absorbent allow more hydrated ions to migrate. Compared with a traditional IPMC composed of a pure Nafion membrane, this new type of IPMC showed a higher response speed, and the output displacement and output force of the new IPMC increased by 42% and 50 ~ 74%, respectively, under a DC voltage of 3 V [56]. Lee and Yoo prepared a multilayered actuator composed of a Nafion/MWCNT layer (as a supporting electrode) and a Nafion-117 membrane by hot pressing and electric spinning. In addition, an Au electrode was sputtered on the surface of the Nafion/MWCNT supporting electrode to reduce the surface resistance. The strain energy efficiency was ten-fold higher than that of a traditional IPMC. Because of the relatively flexible support electrode, fewer cracks arose on the surface of the metal electrode after repeated bending deformation. The remarkable performance

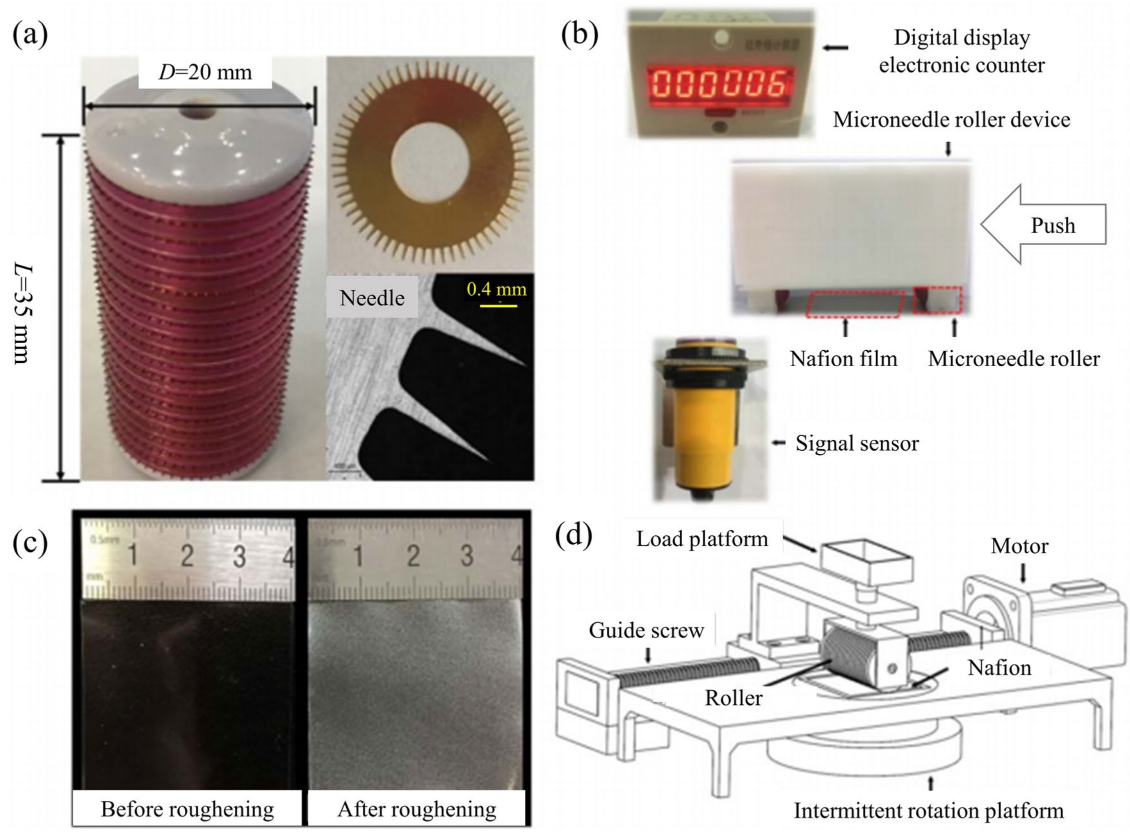

Fig. 2 Images of the microneedle roller device for surface roughening by Chang et al. [74]. **a** Coarsening device based on microneedle roller. **b** The experimental results of roughening device based on the microneedle roller. **c** Effect comparison of Nafion matrix membrane before and after coarsening. **d** Schematic diagram of the microneedle roller coarsening device used to coarsen the matrix membrane of Nafion

of this IPMC actuator was ascribed to the effective quantum chemistry and double electrostatic effect of charge injection, which was attributed to the uniform MWCNT dispersion obtained by electric spinning [57]. Liu et al. fabricated a new type of IPMC actuator consisting of a Nafion membrane enhanced by vertically aligned carbon nanotubes and a pure Nafion membrane, produced by Nafion solution adhesion and hot pressing. An Au electrode was sputtered on the surface of the Nafion membrane with vertically aligned carbon nanotubes. This actuator exhibited continuous migration channels, low resistance, and anisotropic elastic deformation due to the inclusion of vertically aligned carbon nanotubes [58]. Some researchers have used new ion polymer membranes instead of Nafion membranes, such as sulfonated block copolymers with nanostructures and ion channels [59–63].

2.2 Surface Preparation of Nafion Membranes

The performance of IPMCs is closely related to the interface between the matrix membrane and the electrode. The interface structure should be designed to facilitate ion migration as well as platinum ammonia ion penetration and reduction [64, 65]. One effective method is to increase the surface area of the matrix membrane [66–68]. To increase the surface area density of the matrix polymer, Palmre et al. roughened the Nafion membrane surface by sandblasting and sandpapering the surface [10]. Choi et al. prepared an ionic polymer with a large surface area by applying plasma etching to enhance the displacement and response speed of the actuator [69]. Noh et al. prepared an actuator with a large interface area using replicating technology and studied the effect of an increased interface surface area on deformation [70]. He et al. altered the polymer surface with hierarchical microstructures and a large interfacial surface area by applying both polishing and replication. They obtained a high-performing metal electrode with a hierarchical surface texture [71]. Kim et al. [72] and Saher et al. [73] prepared an ion polymer surface with an acicular microstructure via plasma O_2 etching and evaluated the prepared actuator performance. Chang et al. developed a microneedle roller device to roughen the polymer, which produced needle-like electrodes (Fig. 2) and resulted in large deformation of the IPMC at high frequencies [74].

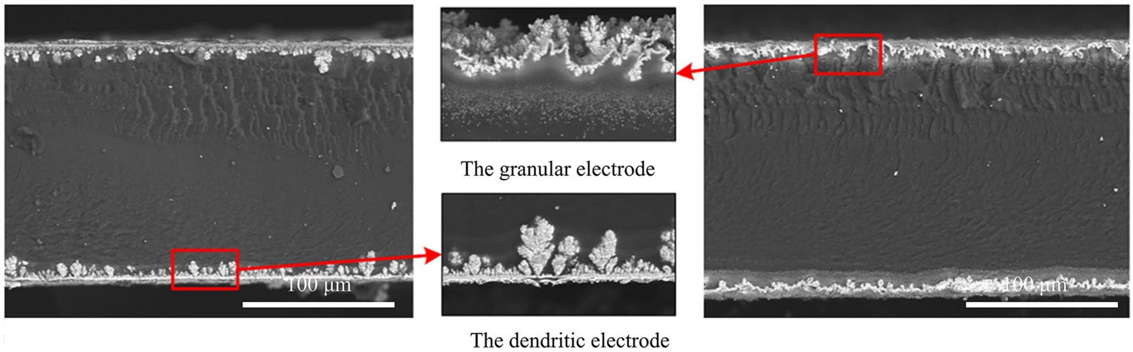

Fig. 3 The SEM image of the Au electrode with dendritic structure prepared by Wang et al. [78]

2.3 High-Performance Electrodes

It is important to produce a stable electrode on the effective surface construction of the Nafion membrane for IPMC actuation. Numerous studies have reported modified electrodes with characteristics such as high conductivity (low surface resistance), high capacitance, large rough interfaces between the matrix and electrode, good metal particle penetration, and excellent stability.

Chung et al. [39] adopted an Ag nanopowder and deposited Ag by electroless plating. Shahinpoor et al. [23] sputtered Ag and Cu on a platinum electrode layer to produce composite electrodes to achieve high conductivity. However, these electrodes had low electrochemical stability. Yang et al. used MWCNTs and graphene to form an electrode on the Nafion surface by spraying and baking [75]. Chen et al. fabricated an actuator by hot pressing carbon nanotubes and a Nafion membrane doped with ionic liquid [76]. Onishi et al. prepared an Au electrode with a dendritic structure in an ionic polymer via continuous adsorption/reduction-electroless plating to increase the surface area between the ion polymer and the electrodes, which induced the electric double-layer effect and increased the capacitance and electrodynamic output performance of the IPMC [77]. Wang et al. performed similar research, as shown in Fig. 3 [78]. Wallmersberger et al. reported that an electrode with a large interface surface area has a large dielectric coefficient, which leads to an increased charge [79].

Although repeated electroless plating causes a Pt electrode to form a dendritic electrode, which enlarges the interface surface area of the electrode, this method increases the rigidity of the electrodes and the IPMC, which hinders IPMC actuation. Park et al. prepared an IPMC actuator with Pt, Au, or Pd metal electrodes and found that a Nafion membrane with Pd particles had the greatest penetration depth [80]. Kim et al. plated Pd metal particles as a buffer layer before performing electroless plating of Pt. Energy-dispersive spectrometry (EDS) analysis revealed that the Pt particles in the IPMC with a Pt electrode rarely penetrated in the Nafion membrane, which had a structure similar to that of a plate capacitor. Pd metal particles in an IPMC with a Pd/Pt composite electrode penetrated 30 µm in the polymer, resulting in an electrical double-layer capacitance [68]. Palmre et al. prepared a Pd/Pt electrode with a large surface area and a thick Nafion membrane to improve the electromechanical properties of the IPMC. EDS analysis displayed that the penetration depth of Pd particles reached 100–200 µm in the Nafion membrane [81]. However, Lu et al. reported that deposition of a Pt layer on the surface of an IPMC actuator produces micro/nanometer cracks during the water evaporation process [82]. Lee et al. [57] and Park et al. [83] observed that the surface of an IPMC actuator had a fragmented fissured electrode surface after several actuation processes. We also observed these fractures, as shown in Fig. 4. These cracks on the electrode cause water molecules to leak out of the electrodes, which affects the IPMC hydration degree, causes energy loss, significantly reduces the conductivity of the electrode, and degrades the IPMC actuation performance [84]. Moreover, platinum nanoparticles are unstable and become active catalysts, which is attributed to the change in atomic configuration and electron spin caused by the activity of atoms on the nanosurface [85]. The platinum nanograins oxidize and clearly increase the surface resistance. Therefore, cracks on metal electrodes and nanoparticle oxidation are two causes of increased surface resistance in IPMCs. As shown in Fig. 5, He et al. deposited MWCNTs on a metal electrode via an electrophoresis method to fill the cracks and reduce the platinum nanoparticle oxidation, thus retaining the stability of the electrode [14].

2.4 New Ionic Electro-Active Polymer

Recently, ionic EAPs, such as IPMCs, conductive polymers, and nanocarbon composite materials, have drawn widespread attention because they exhibit high flexibility, large displacement, and light weight advantages similar to those

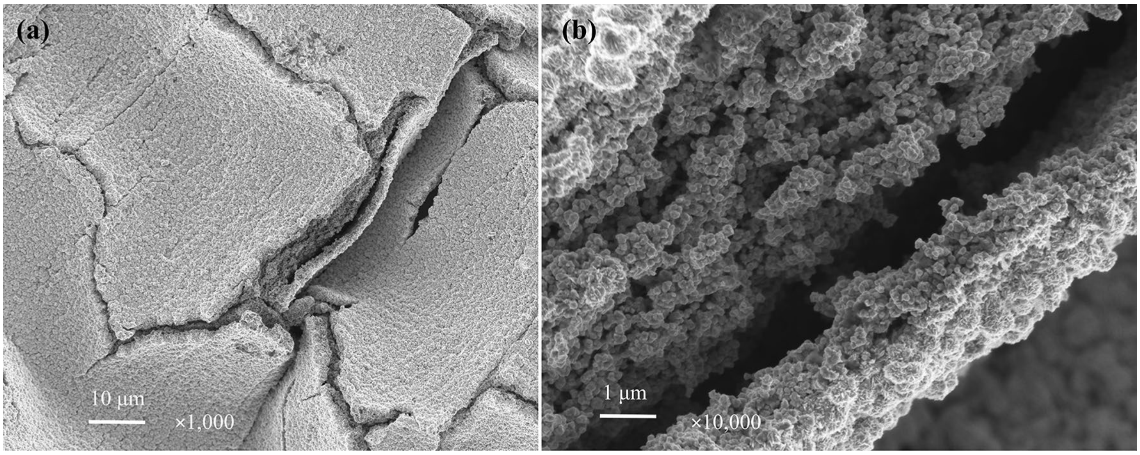

Fig. 4 Surface SEM image of the surface of IPMC [84]. **a** Existing micro cracks on the electrode surface of IPMC, which would aggravate the water leakage and decrease the driving performance of IPMC. **b** Magnified cracks

of natural muscle under low voltage [86–88]. An IPMC is typically composed of an ionic electrolyte thin membrane sandwiched between two metal electrodes consisting of gold, platinum, or palladium [89–93]. As stated above, after several actuation processes, the metal electrodes generate numerous cracks. Thus, a non-fissured electrode with excellent electrical conductivity is essential for achieving a high-performing actuator.

SWCNTs, which exhibit excellent electrical conductivity and high specific capacitance and can be utilized in quantum

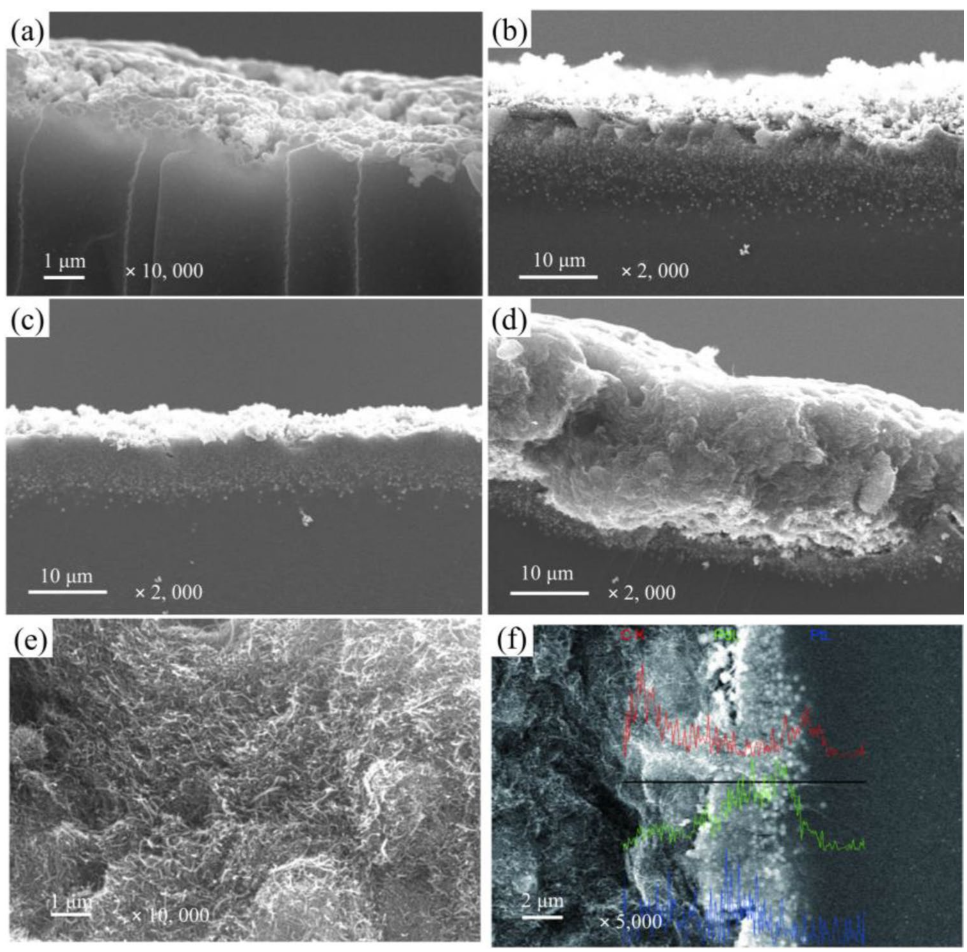

Fig. 5 Cross-sectional SEM images of the IPMC with various electroless plating and electrophoresis deposition cycles [14]. **a** First plating with Pd; **b** Second plating with Pd; **c** Third plating with Pt; **d** Effects of electrophoretic deposition of MWCNTs; **e** Deposited MWCNTs observed at ×10,000 magnification; **f** EDS analysis of C (Red Line), Pd (Green Line) and Pt (Blue Line) content test in IPMC actuator

mechanics applications beyond metallic electrodes, can be used as electrode materials for actuators and super-capacitors [94]. Mukai et al. reported two-generation SWCNT actuators that were superior to previously reported dry-state EAP actuators. These SWCNT actuators were superior because millimeter-long SWCNTs and ionic liquids were closely linked together and formed a thin membrane with ultra-high conductivity (169 S/cm) [12]. Palmre et al. studied the effect of incorporating SWCNTs on the carbon electrode of carbide derivatives. SWCNTs can introduce more mesopores into carbon-based membranes of carbide derivatives, which promotes mass transfer (diffusion) of the electrolyte in the electrode layers and provide more pores and cavities for rapid ion migration [95]. However, SWCNTs are currently rare, expensive, and difficult to disperse evenly.

Graphene membranes exhibit extraordinary electrical, thermal, and mechanical properties resulting from their unique structure. There is an abundance of inexpensive graphite in nature for producing graphene; thus, graphene is an inexpensive alternative to replace SWCNTs [96]. Yang et al. successfully prepared graphene membranes with layered structures via a chemical conversion method [97]. These graphene gel membranes have a self-adaptive and metastable porous structure and can be irreversibly compressed by capillary pressure. By controllably removing volatile solvent in the gel, the packing density can be increased. Additionally, the graphene stacks face to face in the membranes; thus, the packing density reached 1.33 g/cm^3, almost twice the density of traditional active porous carbon (0.5–0.7 g/cm^3). More importantly, graphene membranes containing liquid electrolytes provide a continuous ion channel network and achieve a high capacitance and energy density. Hwang et al. developed a new permanent floating ion polymer–graphene complex actuator. In this work, hydrophobic graphene was etched by an asymmetric laser. The actuator utilized water and ionic liquids as an electrolyte and showed an exceptionally persistent drive with no obvious attenuation. More importantly, the surface of thin graphene membranes was modified by laser etching to increase the binding force between the thin graphene membrane electrode and the Nafion membranes [98]. Lu et al. introduced a new electromechanical actuator based on reduced graphene oxide/carbon nanotube electrodes (135 S/cm), which showed a large strain, wide frequency response, and stable actuation [99]. In addition to human-related applications, degradable and biocompatible ion-exchange polymers are needed for intelligent electronic fabrics, surgical tools, and disposable medical devices. In recent years, chitosan, which exhibits biodegradability, biocompatibility, and biological activity, has attracted increasing attention from researchers and has played an important role in these fields. Thus far, studies have shown that ionic liquids at the molecular level are compatible with biopolymer chitosan, and their complexes can be used as biosensors [100, 101]. Li et al. reported a new electrochemical actuator based on SWCNTs, comprising a thin SWCNT membrane electrode layer on two sides and a middle chitosan electrolyte/ionic liquid layer [102]. Lu et al. introduced a type of actuator based on a chitosan/ionic liquid electrolyte and a chitosan/MWCNT electrode [101]. Sun et al. [103] and Zhao et al. [104] developed a chitosan polymer gel actuator with an MWCNT electrode. He et al. investigated a novel ionic actuator, composed of two graphene films and a basement film. In this actuator, the bending strain ranged from 0.032 to 0.1% under various voltages, as shown in Fig. 6. The graphene membrane electrode exhibited a low sheet resistance of 10 Ω·sq^{-1} and excellent adhesion to the ionic electrolyte polymer membrane, forming an electric double layer at the interface between the ionic electrolyte membrane and

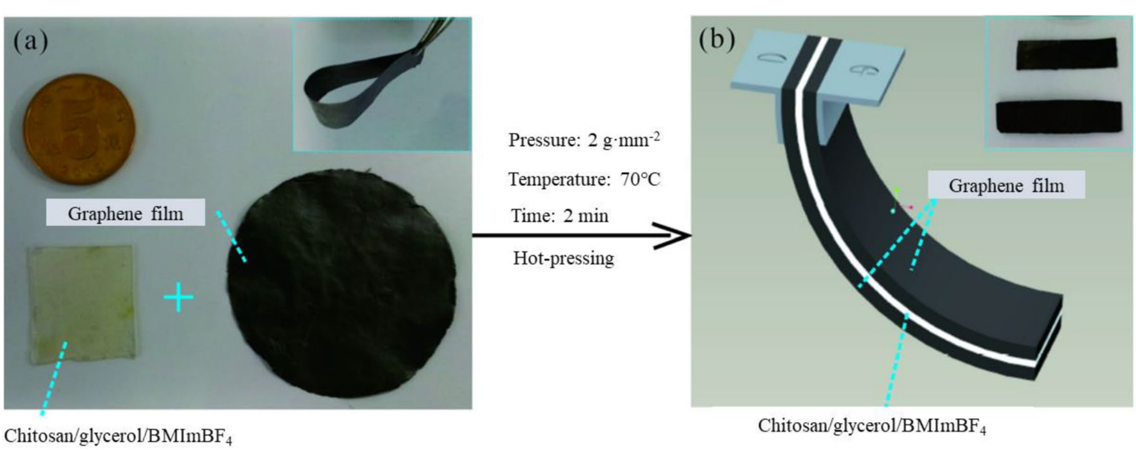

Fig. 6 Schematic of preparation of the newly ionic actuator by He et al. [84]. **a** Graphene film and chitosan/glycerol/$BMImBF_4$ film, the inset is a tailored flexible graphene film; **b** The schematic diagram of the structure of the ionic actuator made by hot pressing, the inset is the prepared sample

Table 1 Performance comparison for different manufacture methods

Improving method	Method	Interlayer material	Electrode	Displacement (mm)	Blocking force (mN)	References
Material modification	Solution casting	Nafion film	Pt/Au	~4	~196	[21]
Material modification	Solution casting	Nafion film	Pt	~40	~21.56	[22]
Material modification	Hot pressing	Three layers Nafion-117	Pt	~1.5	~18.62	[30]
Material modification	Hot pressing	Five layers Nafion-117	Pt	~0.7	~78.4	[30]
Material modification	Hot pressing	Nafion-117	Pt	~3.5	~32	[31]
Material modification	Solution casting	Nafion film	Pt	12.3	68.4	[32]
Material modification	Solution casting	Nafion/MWCNTs	Pt	/	35.18	[36]
Material modification	Solution casting	Nafion/FCNT	Au	/	32.3	[52]
Material modification	Solution casting	Nafion/NCNC	Pt	14	29.4	[17]
Material modification	Solution casting	MMT-Nafion/Silicon	Pt	~1.0	88.2	[54]
Material modification	Solution casting	MMT-Nafion/Silicon	Ag	~2.8	49	[54]
Material modification	Solution casting	Nafion/MCNT/TEOS	Pt	10.2	63.7	[55]
Material modification	Solution casting	Nafion film	Pt	13.96	15.68	[55]
Material modification	Solution casting	Nafion/MCNT	Pt	/	35.28	[55]
Material modification	Solution casting	Nafion/TEOS	Pt	9.23	37.83	[55]
Material modification	Solution casting	Nafion/s-MMT /CSA	Pt	17.3	7.15	[56]
Material modification	Solution casting	Nafion/p-Alumina /CSA	Pt	17.4	6.17	[56]
Material modification	Solution casting	Nafion/CSA	Pt	14.2	3.33	[56]
Material modification	Solution casting	Nafion film	Pt	12.2	4.12	[56]
Material modification	Electro-spinning	Nafion/MWNT	Au	16.7	3.33	[57]
Material modification	Hot pressing	PSS-b-PMB	SWCNTs	6.5	/	[59]
Material modification	Solution casting	SSPPC/s membrane	Pt	10.6	2.84	[60]
Material modification	Solution casting	SSPB/S-MMT	Pt	10.2	/	[61]
Material modification	3D printing	Aquivion and Nafion	Pt	/	/	[62]
Material modification	Solution casting	SPEI membrane	Pt	2.75	/	[63]
Surface preparation	Emery paper	GEFC	Pt	6	20	[10]
Surface preparation	Developing process	Nafion-117	Pt/Pd	~7.5	~0.98	[68]
Surface preparation	Plasma treatment	Nafion-1110	Pt	0.068	4.31	[69]
Surface preparation	Hot pressing	Nafion film	Pt	16.1	61	[71]
Surface preparation	Plasma treatment	Nafion-117	Au	~9	~2.75	[72]
Surface preparation	Plasma treatment	Nafion film	Pt	~12.5	~3.2	[73]
Surface preparation	Microneedle roughening	Nafion film	Pd/Au	12.2	/	[74]
Remarkable electrode	Electroforming	Nafion film	Ag/Ni	6.78	2.18	[39]
Remarkable electrode	Electro-spray	Nafion film	MWCNT/graphene	2.33	/	[75]
Remarkable electrode	Hot-pressing	Nafion-117	SWCNT	> 20	/	[76]
Remarkable electrode	Electroless plating	Flemion film	Au	~1.0	/	[77]
Remarkable electrode	Electroless plating	Nafion-117	Pd/Au	~15	/	[78]
Remarkable electrode	Electroless plating	PFSA membranes	Pd/Pt	27	198	[81]
Remarkable electrode	Electroless plating	PSMI-PVDF	Pt	~12	/	[82]
Remarkable electrode	Electroless plating	Nafion-117	Pt	/	/	[83]
Remarkable electrode	Hot-pressing	Chitosan/glycerol	Graphene film	~0.945	/	[84]
Remarkable electrode	Electrophoresis	MWCNT/Nafion	Pd/Pt/MWCNT	6.98	22.28	[14]
Remarkable electrode	CVD	Nafion film	SWCNT	/	/	[93]
Remarkable electrode	Casting	PVDF (HFP)	SWCNTs	5	/	[12]
New EAP actuators	CVD	PVDF (HFP)	Graphene	/	/	[94]
New EAP actuators	Vacuum filtration	PVDF (HFP)	Graphene	/	/	[95]
New EAP actuators	Transfer printing	CEFM	Graphene film	/	/	[96]
New EAP actuators	/	PVDF	RGO/MWCNT	~6.5	/	[99]

Table 1 (continued)

Improving method	Method	Interlayer material	Electrode	Displacement (mm)	Blocking force (mN)	References
New EAP actuators	GO-PSC-IL	Graphene/chitosan	Au	~ 8	/	[100]
New EAP actuators	Hot pressing	CS/chitosan	CS/MWNCT	~ 6	/	[101]
New EAP actuators	/	Chitosan electrolyte	CS/MWCNT	1.0	/	[102]
New EAP actuators	Hot pressing	Chitosan and Genipin	MWCNTs	/	7.3	[103]
New EAP actuators	Hot pressing	Chitosan and glycerol	MWCNTs	/	7.7	[103]
New EAP actuators	Direct casting	Chitosan gel	MWCNT	~ 2	/	[104]

the graphene membrane electrode and producing a higher capacitance (27.6 mF•cm^{-2}) under the input potential [84]. Table 1 summarizes some fabrication methods for improving the blocking force and displacement of IPMC artificial muscles.

3 IPMC Modeling

For IPMC modeling, various approaches have been proposed to model different IPMC properties. The most frequently modeled properties of IPMCs are related to the electromechanical or mechano-electrical transduction of IPMCs, i.e., either the simulation of IPMC tip displacement and/or blocking forces (where the IPMC works as an actuator) or the simulation of an electrical potential on the IPMC electrodes (where the IPMC works as a sensor). Universal models have also been derived to describe both actuation and sensing [105, 106]. Some IPMC models focus on equivalent electrical circuits that can describe the electrical properties of IPMCs or that can be applied to control IPMCs over a broad range of frequencies [107–109], the actuation mechanisms of IPMCs in a sensorless model [110], or solvent loss due to an applied electric potential [111]. There are various challenges in IPMC modeling, such as (i) a strong nonlinearity among key model parameters [110, 112, 113], (ii) a lack of perfectly distinct boundaries between the electrodes and the polymeric membrane, i.e., the density of electrode nanoparticles embedded in the polymeric membrane changes substantially with depth [114], (iii) variations in the electrical resistance of IPMC electrodes during actuation depending on the IPMC curvature and/or distance from the IPMC tip [115, 116], (iv) the effects of hydrolysis and evaporation during the operation of conventional water-based IPMCs [117], (v) the presence of steric effects, i.e., effects that occur when atoms are brought close together, which may arise in the boundary layers between electrodes and the hydrated polymeric matrix when an electrical voltage is applied [118, 119], (vi) shape memory effects in the polymeric membrane of an IPMC [120, 121], and (vii) back-relaxation, structural dynamic effects, and time-varying behaviors [122]. This list of challenges is far from complete.

IPMCs are structurally complex materials that undergo complicated electrochemical processes during actuation or sensing. As no IPMC model can include all physical and chemical effects, some simplifying assumptions must be made. The assumptions applied in various IPMC models are related to the mechanical properties of IPMCs, such as the assumption of a polymeric membrane with isotropic elasticity. Some authors have attempted to improve the simple condition of isotropic elasticity by introducing variability in the elastic constants depending on the level of IPMC hydration or by applying a neo-Hookean model [114, 123, 124] to the polymeric membrane. Deformations may be treated either in a simple manner by using an infinitesimal strain tensor [125] or using the Cauchy–Green strain tensor for large deformations [126]. By assuming a specific shape for the IPMC, some authors have applied beam or shell theory approaches [127, 128]. Linear beam theories, such as the Euler–Bernoulli beam theory, cannot account for nonlinear deformations. Therefore, some authors have adjusted the linear beam theory model to include nonlinearities using the von Kármán approach, which accounts for large deflections [129]. Regarding the conductivity of IPMC electrodes, some models assume a perfect electrical conductivity or electrical resistance depending on the IPMC curvature and/or distance from the IPMC tip. In addition, a constant overall electric permittivity is often assumed for hydrated IPMCs [114, 130, 131]. However, some authors [132] use a variable electric permittivity that depends on polymer depth. The IPMC capacitance is closely related to the IPMC electric permittivity, and thus, the nonlinear character of the IPMC capacitance can be described [113].

IPMC models can be divided into several categories. (i) Black box models [133–138] are based on an empirical relationship between input data and IPMC responses. These models do not explain the governing physical or chemical processes within the IPMC. These models may also use neural network methods. Black box models are generally more suitable for real-time control but are sample dependent. (ii) Gray box models [139–143] are not entirely based on physical principles; instead, they also build upon some empirical patterns. Finally, (iii) white box models [114, 130, 132, 144–153] are completely based on physical principles. Models in this category are also sometimes called physics-based models. Physics-based models generally start with partial differential equations (PDEs) describing the physical and chemical processes occurring in IPMCs. Some authors distinguish another model category: control-oriented, physics-based models [112, 113, 154–156]. Some authors find that white box models are not practical for real-time control purposes (it may be time-consuming to find a numerical solution for nonlinear PDEs), and therefore, they amend a white box model to obtain a model that is suitable for real-time control. These amended models may fall into the category of control-oriented models. The various control types used in these models include proportional integral derivative (PID), proportional integral (PI), and standard H-infinity control ($H\infty$).

In this review, physics-based models are the primary focus. Physics-based models may rely on either numerical [130, 145], analytical [114], or semi-analytical methods [132]. The starting point of these physics-based models is often the Nernst–Planck equation, the Poisson equation of electrostatics (often abbreviated as the PNP equations), and an equation of mechanical equilibrium. However, Del et al. did not consider the Nernst–Planck equation because they assumed that electrostatic forces are primarily responsible for mechanical actuation [151]. Additionally, the authors use the theory of mixtures [157]. Physics-based models may be further divided according to whether they are derived at the microscopic [148, 151, 158] or macroscopic [159] level. As a drawback, microscopic models may present some difficulties for determining model parameters. These physics-based models identify two mechanisms responsible for the bending of IPMCs: electrostatic interactions and ion and water migration. Some models emphasize the former [148], whereas others emphasize the latter mechanism [160]. Porfiri et al. presented a physics-based plate-like model for a thin, flat IPMC derived for actuation and sensing within small deformations [105]. This model was inspired by traditional plate models for moderately thin piezoelectric bimorph plates. In this model, the elastic properties of the electrodes and the polymeric membrane are assumed, and various levels of IPMC hydration are considered. The model combines the Nernst–Planck equation for counterion transport with the Poisson equation and the general principles of multiphase mixtures. The authors showed that the capacitance, which depends on thickness of the boundary layers (the layers between the electrodes and the hydrated polymeric matrix), strongly influences the actuation and sensing performance of an IPMC. Johnson and Amirouche described thermal effects in their model [131]. In addition to the Nernst–Plank and Poisson equations, they also used Fourier's law of heat conduction. The authors both simulated and measured the temperature rise. Zhu et al. presented a physics-based model for an IPMC working as a sensor [124]. The Nernst–Planck equation involved the following transport processes: convection flux under the total pressure gradient, electrical migration governed by the Poisson equation, and the inter-coupling effect between cations and water. A simple step-bending process was simulated for various model parameters, such as the elastic modulus, the hydraulic permeability coefficient, the diffusion coefficients of cations and water, and the drag coefficient of water. Pugal et al. extended upon a physics-based model for an IPMC working as an actuator by coupling the currents in the polymer matrix to the electric current in the electrodes of the IPMC [130]. This coupling was performed on the basis of Ohm's law and the Ramo–Shockley theorem. In their actuation model, the authors used an infinitesimal strain tensor, and the stress tensor was coupled with the charge density in a quadratic manner. Xiao and Bhattacharya [149] introduced a physics-based model starting from the total free energy of a system combined with an IPMC treated as a compressible neo-Hookean material. This model was based on the concept that ion concentration, electric potential, and deformations evolve in such a way as to minimize the total free energy. The Cauchy stress and Maxwell electrostatic stress tensors are expressed, and the obtained equations correspond to the Nernst–Planck diffusion equation, the Poisson equation of electrostatics, and the equation of mechanical equilibrium. IPMC actuators/sensors are commonly plates with a uniform thickness used in bending mode, with rods or tubes being less common. Sharif et al. modeled and fabricated an IPMC torsional sensor by considering ion diffusion, electromigration, and convection [152]. The PNP equations alone are not sufficient to obtain the induced electrical potential. Therefore, the anion concentration is expressed based on the local deformation induced by a mechanical stimulus. An identical relation in a general form has also been used [106, 161]. An IPMC sensor with a tubular shape [150], which can bend in all directions, has also been reported.

4 IPMC Applications

Due to their advantages, IPMC actuators have great potential for applications in bio-inspired robotic muscle actuation, opto-mechatronic systems, and biomedical engineering.

4.1 Robotic Actuators

IPMCs have the characteristics of natural muscle and have been praised as an "artificial muscle." Thus, IPMCs have broad potential as robot muscle actuators [162], including applications in robotic fish with tail fin muscles [163–171],

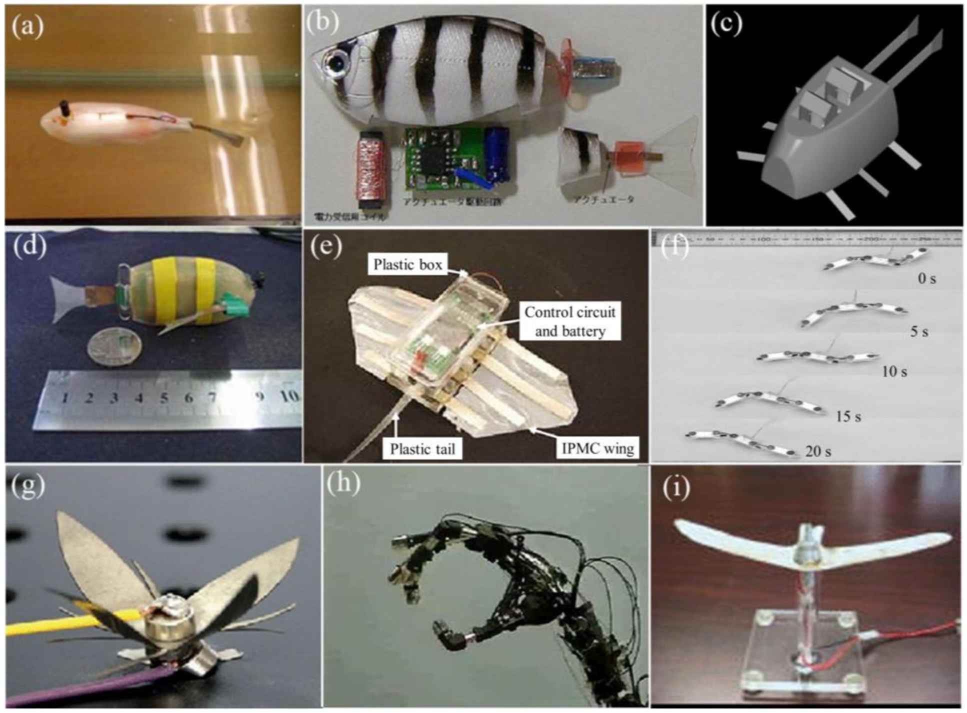

Fig. 7 The application of IPMC in robot actuators. **a**–**d** Robotic fish with the tail fin muscle [163–169]; **e** Manta ray robot derived by IPMC [172]; **f** Snake-like robot using IPMC [174]; **g** The bionic flower made by IPMC, which can effectively simulate the expansion and closing of the flower [175]; **h** The gripper with multiple freedom joints made by IPMC [176]; **i** The bionic flapping wing device made by IPMC can simulate the vibration of the flapping wing [123, 177]

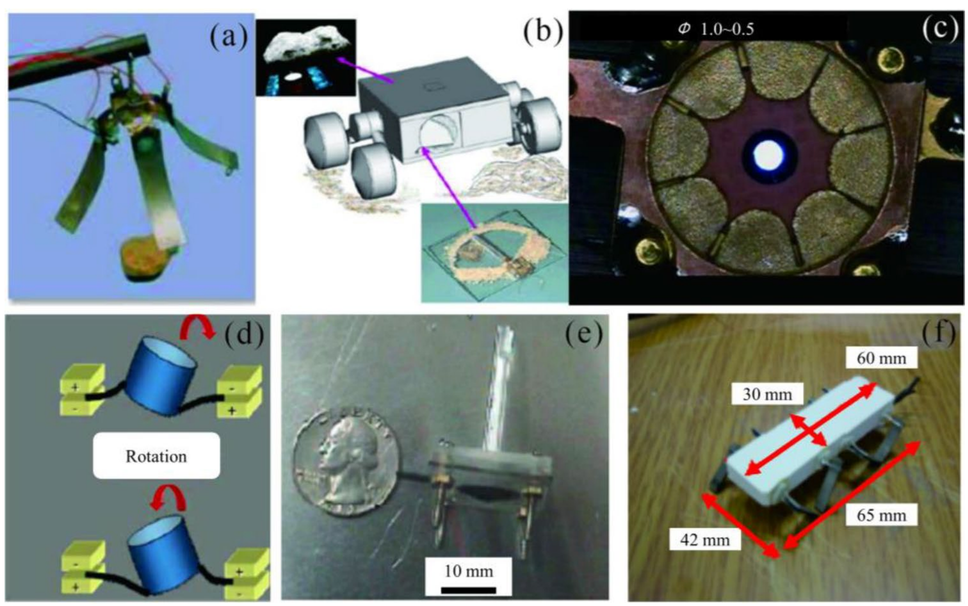

Fig. 8 The application of IPMC in photoelectric system. **a** Four claw catcher [178]; **b** Dust removal device [180]; **c** Lens driving unit [181]; **d** The driving element of removing the position of capsule endoscopy's lens [182]; **e** Micro pump [183]; **f** Eight-legged robot [184]

ray robots with pectoral fin actuators [172, 173], snake-like robots [174], and bionic blooming flowers [175], as shown in Fig. 7a–g The Jet Propulsion Laboratory (JPL) at the National Aeronautics and Space Administration (NASA) has developed IPMCs as artificial joints to realize a robotic arm with multiple degrees of freedom [176], and the Environmental Robots Incorporated Company has developed a flapping wing device [123, 177], as illustrated in Fig. 7h, i.

4.2 Opto-Mechatronics Systems

Figure 8a shows a four-claw gripper produced by the JPL with the use of an IPMC [178]. An IPMC-based softer gripper that can grasp objects with various features was further developed by He et al. [179]. The Institute of Space and Aeronautical Science and NASA have established a Hayabusa asteroid exploration plan called the MUSES-C task, which aims to collect sample material from the surface of an asteroid and return it to the earth. Analyses

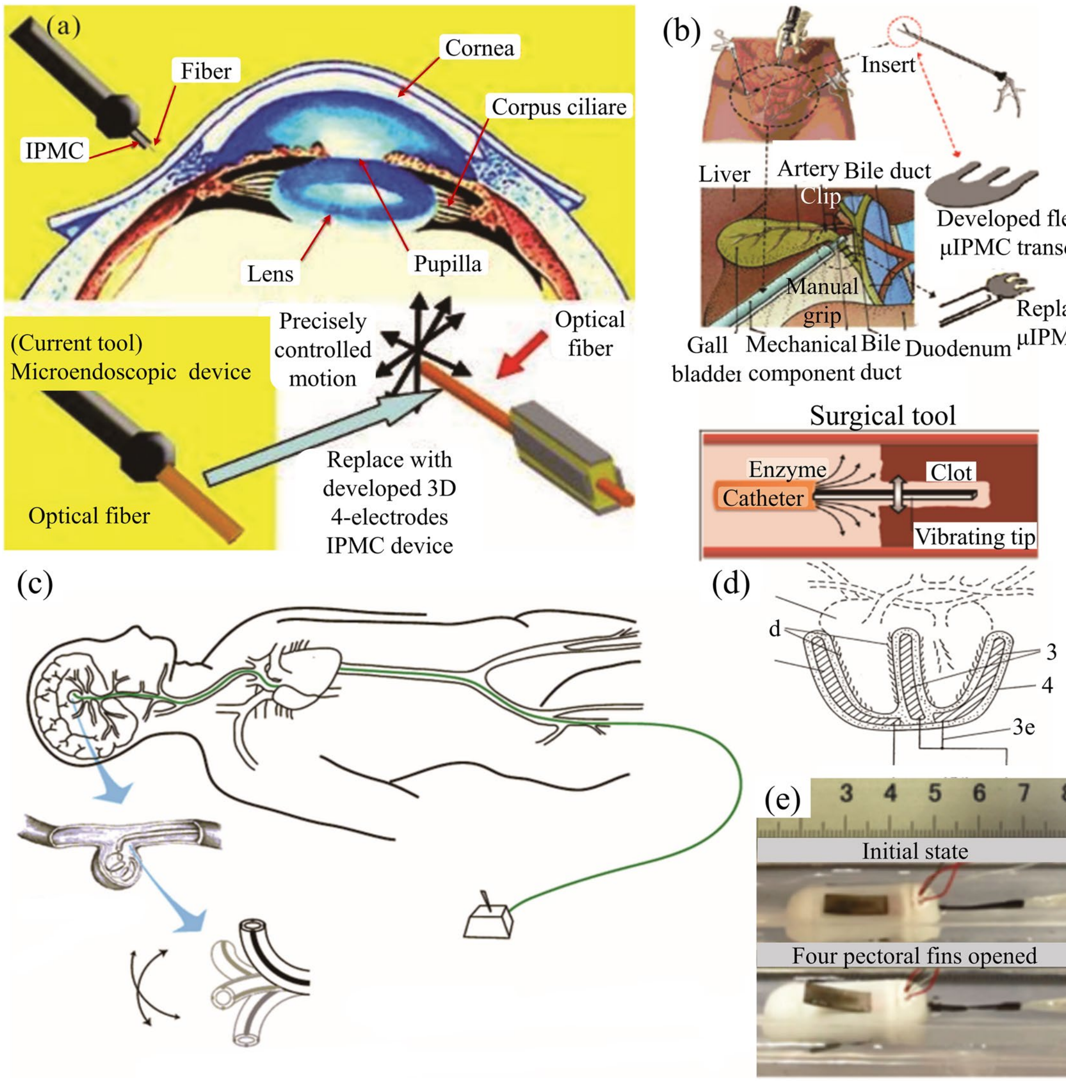

Fig. 9 The application of IPMC in medical engineering. **a** Optical fiber actuator for microscopic eye surgery [186]; **b** Microsurgical forceps [187]; **c** Active catheter [192]; **d** Cardiac auxiliary diastolic device [193]; **e** Capsule endoscopic control [194]

have revealed that the camera lens dust removal device carried by the "rovel'' mobile robot (MUSES-CN) in the spacecraft can be driven by an IPMC, as shown in Fig. 8b [180]. The Eamex Corporation studied lens actuators for mobile phones and cameras, as displayed in Fig. 8c [181]. Kim et al. creatively used an IPMC as the driving element to modulate the position of a capsule endoscopy lens, as shown in Fig. 8d [182]. Figure 8e and f present a micro pump actuated by a round IPMC membrane [183] and an eight-legged robot fabricated by an IPMC actuator, respectively [184].

4.3 Medical Engineering

IPMCs have great application prospects in medical engineering based on their characteristics of flexibility, large deformation, and biocompatibility [185]. Feng and Tsai used a four-electrode cylindrical IPMC to improve the accuracy of an optical fiber actuator for microscopic eye surgery, as shown in Fig. 9a [186]. They also studied microsurgical forceps, as shown in Fig. 9b [187], and artificial cochlea actuated by micro IPMCs [188]. In addition, numerous studies have focused on active catheters [185, 189–191]. The Eamex Corporation used a cylindrical IPMC for brain catheters, the end of which can achieve two-dimensional active bending, as demonstrated in Fig. 9c [192]. Shahinpoor et al. developed an artificial heart with an IPMC as an auxiliary diastolic device (Fig. 9d) [193]. Li et al. [194] utilized an IPMC to modulate the movement of micro-capsule robots in the body (Fig. 9e).

The sensing characteristics of IPMCs have led to broad application prospects in engineering [20, 89, 195]. Recently, the Robert Bosch (South East Asia) Corporation in Singapore belonging to the Bosch group in Germany has focused on the development of electrodynamic polymer actuators and their applications in flexible wearable electronics. The mechano-electric transduction of IPMCs will be presented in detail in the future.

At present, many scholars have explored applications of IPMCs in the fields of bionic drives, flexible sensing, and biomedicine. The application potential of IPMCs has gradually arisen in bionic robotic fish, underwater robots, blooming flowers, wings that can flap in the air, flexible grippers, space wipers, precision micro pumps, endoscopic driving units, surgical micro forceps, heart assist devices, and surgical catheters. Due to the unique material properties and driving performance of IPMCs, it is more advantageous to apply IPMCs to water or humid environments because water can provide the medium required for ion migration. Therefore, from a comprehensive point of view, the current IPMCs have great application prospects in the field of underwater bionic robots to realize underwater environmental investigation, water quality monitoring, or underwater cultural relic salvage. Furthermore, IPMCs can also be used as a guide device [196] for interventional catheters [192] in human vascular interventional operations, where traditional surgical catheters are difficult to operate and cannot be actively guided. IPMCs are beneficial, because they take advantage of liquid environments, such as underwater environments or human blood vessels. Meanwhile, these two applications still face some challenges. For example, the water-tightness of the device in a water environment must be considered, and safety must be evaluated for the human body, representing indispensable steps for future applications.

4.4 Summary and Perspectives

In summary, many scholars have comprehensively analyzed the mechanical and driving performance of IPMC materials. These works have laid a solid foundation for the development and application of IPMCs. Although the performance of IPMCs has greatly improved, because of the actual application environment and the limitations of IPMCs, the following aspects must be studied in the future.

First, it is necessary to prepare non-metallic flexible IPMC electrode materials in an efficient manner to reduce production costs. At present, most IPMC electrode materials are produced from rare precious metals (Pt, Pd, Au, and Ag). This practice increases the cost of IPMC production; moreover, the metal electrode may suffer from cracks and oxidation after many cycles of operation, which greatly reduces the compactness, water retention ability, and conductivity of the electrode, thereby reducing the service life and driving performance of the IPMC. In addition, the preparation of metal IPMC electrodes is cumbersome and requires a long preparation time; thus, the development of a method for efficiently and conveniently preparing low-cost flexible electrodes is important for future research.

Second, the preparation of high-performance air-operating IPMCs is necessary. Traditional water-based IPMCs have a short working time in air (only a few minutes). As the working time in air increases, the driving performance gradually decreases (creep phenomenon), and it is difficult to ensure that the IPMC has a stable driving performance in air. Although ionic liquid-based IPMCs have a much longer working time in air, these IPMCs are far below traditional water-based IPMCs in response speed, output force, and displacement performance. Future IPMC applications will require not only a high response speed and stable driving performance, but also the ability to operate in air for a long time. These characteristics represent a focus for future research, along with some challenges.

Third, we should investigate new applications in which the advantages of IPMC performance can be fully exploited. Because of the unique material properties and driving mechanism of IPMCs, the application of IPMCs in water quality monitoring and underwater reconnaissance and salvage should be considered in the future. IPMCs can also be used as a guide device for human interventional surgical catheters. In addition, multi-degree IPMC can be demanded in space soft robotics after the success development of ionic liquid-based IPMC.

5 Conclusion

This paper has reviewed research progress on improvements, modeling, and applications of IPMC artificial muscle. Improvements include enhancements to the matrix membrane, surface treatment of the matrix membrane, electrode preparation, and new EAP actuators to overcome the shortcomings of traditional IPMC actuators, such as a low power output and short effective air operation time.

The performance of IPMC actuators can be enhanced by adding nanoparticles to the polymer to strengthen the Nafion substrate membrane, which improves the mechanics, thermal stability, conductivity, and versatility of the substrate. The performance of IPMCs can also be improved by treating the surface of Nafion membranes and increasing their surface area. As another method for improvement, one can utilize superior electrode materials with high conductivity, high capacitance, and stability, and one can prevent cracks that occur in metal electrodes and nanoparticle oxidation. A new type of ionic EAP actuator has been prepared using graphene film, chitosan, and ionic liquids. These new IPMC actuators have more advantages than traditional IPMC actuators. In addition to methods for improving the performance of IPMCs, this paper reported on IPMC modeling, including current model challenges and several model categories. Challenges such as nonlinearity, boundaries, varying resistance, solvent evaporation, steric effects, shape memory effects, and back-relaxation exist in IPMC modeling. Black, gray, and white box models as well as control-oriented, physics-based models were also described. This review primarily focused on physics-based models, which rely on numerical, analytical, or semi-analytical methods. At present, IPMCs have been widely used in bionic robots, photoelectric systems, and medical engineering. In the near future, IPMCs will be applied in various aspects of industry.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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