CHAPTER 11

An Overview of Preparation, Properties and Applications of Ionic Polymer Composite Actuators

Inamuddin and Ajahar Khan

Department of Applied Chemistry, Aligarh Muslim University, Aligarh, India

*Corresponding author E-mail address: inamuddin@rediffmail.com

Abstract

It deals with the basic information regarding to the materials and their types used for manufacturing IPMC actuators using various fabrication processes under optimum experimental parameters and possible applications. Among the variety of electroactive polymers recently developed IPMCs are good candidates for use in bio-related, robotics and aerospace applications because of their biocompatibility. Several fabrication processes, their performance and mechanical characteristics, a number of recent applications of IPMCs have been reported in this chapter. The control and various factors affecting performance of IPMC have also been reported.

Keywords

Ionic Polymer Metal Composites, Bending Actuator, Electroless Plating, Structure of IPMC

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The development of mankind and human civilization can be correlated to the invention and manufacture of new materials. In present times, new materials are discovered and manufactured at a very high frequency. We could christen this time to be the decade of smart materials. Composite materials that respond to an external stimulus in real or near real time are known to be as “smart” materials. Smart materials are thus defined as materials that exhibit a fast, repeatable, reversible and significant change in at least one of their physical properties in response to an external stimulus[1]. Smart materials are present in all the basic forms (solid, compound, composite and solution). Smart materials on the basis of external stimulus can be classified as ‘property changing’ or ‘energy exchanging’ [2]. ‘Property changing’ materials are those in which the input external energy causes a mechanical, chemical, thermal or electrical change in material properties. Energy exchanging materials involve the changes in their internal energy level while the materials properties remain the same. The response of the materials in either category is reversible and direct. Smart materials in these two categories along with their characteristic properties are listed in **Table 1**. Among the different variety of materials listed in **Table 1**, electroactive polymers (EAPs) have been extensively studied and received significant attention due to their inherent attractive features such as mechanical flexibility, light weight, easy processing, flexibility and fracture tolerance. These polymers exhibit large property changes in response (change shape or size) to electrical stimulation which adds significant benefits to their potential applications. In addition to these properties, these polymers respond to an electric field or mechanical deformation [3–11]. Since the invention in the mid 1970’s, EAP materials have been the subject of extensive studies by several research groups. EAPs are considered to be alternatives to predictable sensor and actuators [12–14] because of their low electric voltage dependence, flexibility, light weight, bio-compatibility, compliant properties, ability to perform in air and aquatic media and easy fabrication processes.

EAP materials respond mechanically to external stimulation with considerable change in their shape and size. Generally, electroactive polymers are classified into two major categories [2,15,16] **Table 2**. The classification is based on their activation mechanism responsible for actuation as electronic EAPs (which are driven by an electric field or coulomb forces) and ionic EAPs (which change shape by mobility or diffusion of ions and their conjugated substances). The electronic EAPs such as electrostrictive, electrostatic, piezoelectric and ferroelectric generally require high activation fields (>150

$\text{V } \mu\text{m}^{-1}$) which are close to the breakdown level of the material. Beside requiring high activation electric potential, electronic EAPs can be controlled in a better manner as they can hold the induced displacement on the application of dc voltage. The property of these materials to hold the induced displacement, when a dc voltage is applied, makes them potential materials in robotic applications and these materials can be operated in air without major constraints. The electronic EAPs also have high energy density as well as a rapid response time in the range of milliseconds. In general, these materials have a glass transition temperature inadequate for low temperature actuation applications [1]. On the other hand, ionic EAPs are materials that involve mobility or diffusion of ions and they consist of two electrodes and an electrolyte. The activation of ionic EAP can be achieved by relatively low voltage and generally induce bending displacement. Ionic EAP materials such as gels, ionic polymer-metal composites, conducting polymers and carbon nanotubes require low driving voltages, nearly equal to 1-6 V. One of the constraints of these materials is that they must be operated in a wet state or in solid electrolytes. Ionic EAPs predominantly produce bending actuation that induces relatively lower actuation forces than electronic EAPs. Often, operation in aqueous systems is plagued by the hydrolysis of water. Moreover, ionic EAPs have slow response characteristics compared to electronic EAPs. The amount of deformation of these materials is usually much more than electronic EAP materials and the deformation mechanism bears more resemblance to a biological muscle deformation. The induced strain of both the electronic and ionic EAPs can be designed geometrically to bend, stretch or contract [16]. But it is difficult to maintain their position constant under dc activation [1]. They can be used as actuators or sensors. As actuators, they are characterized by being able to undergo a large amount of deformation when they are stimulated. Similarly, when they exhibit the inverse effect, they can be used as sensors. **Table 3** shows comparison of electronic and ionic EAPs characteristics [10].

Table 1: Classification of smart material [2].

S.No.	Property changing			Energy changing		
	Material	Stimuli	Change	Material	Stimuli	Effect
1.	Magnetorheological	Magnetic field	Viscosity	Thermoelectric	Electrical current	Temperature differential
2.	Thermochromic	Thermal energy	Spectral reflectivity	Photovoltaic	Radiation energy	Electrical current
3.	Shape memory	Thermal energy	Crystalline phase change	Photoluminescent	Radiation energy in the ultraviolet spectrum	Radiation energy in the visible spectrum
4.	Thermotropic	Thermal energy	Phase change (conductivity, transmissivity, solubility and volumetric expansion)	Electroactive polymer	Electrical Current	Strain

Table 2: List of leading electroactive polymers.

S.No.	Ionic EAP	Electronic EAP
1	Ionic polymer gels (IPG)	Electrostrictive paper
2	Conducting polymers (CP)	Electrostatic
3	Carbon nanotubes (CNT)	Dielectric EAP
4	Ionic polymer metal composite (IPMC)	Piezoelectric
5		Electro-viscoelastic elastomers
6		Ferroelectric polymers
7		Liquid crystal elastomers (LCE)

Table 3: Properties of electronic and ionic EAPs [10].

S.No.	Properties	Ionic EAPs	Electronic EAPs
1	Controllability	Difficult	Easy
2	Activation voltage	Small	Large
3	Energy density	Poor	Good
4	Electrolyte	Present (wet)	Absent (dry)
5	Mechanism	Mobility or diffusion of ions	Coulomb forces
6	Displacement	Bending	Changes shape or dimensions
7	Examples	Ionic polymer gels (IPG), Ionic polymer metal composites (IPMC), Conductive polymers (CP), Carbon nanotubes (CNT)	Electrostrictive, Elastomers, Electrostatic, Piezoelectric, Ferroelectric, Polymers

EAP materials have been termed as “artificial muscles” because of their electromechanical response under electrical stimulation, exhibiting large strain. For this capability, EAPs are particularly attractive in biomimetic, since they can be used to mimic the movement of animals, humans and insects for making biologically inspired mechanisms. These materials also have the potential for application in the field of robotics, where large linear movement is often needed. The general characteristic properties of EAPs and their significance [10] are summarized in **Table 4**.

Electroactive polymers have great potential as polymer-based actuators and sensors. The actuation in electronic EAPs (e-EAPs) is driven by the electrostatic mechanism due to the presence of dipoles. On the other hand, ionic EAPs (i-EAPs) actuate due to the diffusion or transport of ions within the polymer membrane. While e-EAPs can hold induced displacement under dc activation, i-EAPs require an electrolyte or solvent for actuation. However, due to their bi-directional actuation and low voltage requirements, i-EAPs are being considered for biomedical, space exploration, robotics and naval applications. Among the EAPs, dielectric elastomers, conducting polymers, ionic gels, carbon nanotubes based actuators and ionic polymer metal composites (IPMCs) actuators are

considered to be the most promising and attractive materials to researchers because of their light weight, large bending displacement under low applied voltage and small force generation capability with dexterous behaviour [17–26].

Typically, an IPMC consists of a semipermeable polymer membrane sandwiched between Pt, Au or Ag metal electrodes and is neutralized with certain counter ions that balance the electrical charge of the anions covalently fixed to the back-bone membrane or in simple words an IPMC comprises of a neutralized semi permeable ion-exchange polymer membrane coated with metal Pt or Au as electrode at both surfaces of membrane and water as inner medium for the dissociation for metal cations. Ionic polymer metal composites (IPMCs) are capable of electromechanical and mechanoelectrical response to applied voltage and mechanical deformation respectively [27–30]. **Figure 1** depicts the outline of general introduction of IPMC.

Table 4: The general characteristic properties of EAPs and their significance [10].

S.No.	Measurement		Properties	Significance
1	Mechanical		Tensile strength (Pa)	Mechanical strength of the actuator material
			Stiffness (Pa)	Required to calculate blocking stress, mechanical energy density and mechanical loss factor/ band width.
			Thermal expansion coefficient (ppm/C)	Affects the thermal compatibility and residual stress.
2	Electrical		Maximum voltage (V)	Necessary to determine limits of safe operation.
			Impedance spectra (Ohms and phase angle)	Provides both resistance and capacitance data. Used to calculate the electrical energy density; electrical relaxation/dissipation and equivalent circuit.
			Nonlinear current (A)	Used in the calculation of electrical energy density, quantify nonlinear responses/driving limitations.
			Sheet resistance (Ohms per square)	Used for quality assurance.
3	Electro active properties	Strain	Electrically induced strain (%) or displacement (cm)	Used in calculation of blocking stress and mechanical energy density.
		Stress	Electrically induced force (N) or mechanically induced charge (C)	Electrically induced force/torque or stress induced charge.
		Stiffness	Stress/strain curve.	Voltage controlled stiffness.
4	Microstructure		Thickness (electrode & EAP), internal structure, uniformity, anisotropy and hosted defects	These are features that will require establishing standards to assure the quality of the material.
5	Environmental behavior		Operation at various temperatures, humidity and pressure conditions	Determine material limitations at various conditions.

Figure 1: Outline of general introduction of IPMC.

2. History of IPMC

As mentioned earlier, IPMC is a type of i-EAP which gives a mechanical response upon applied voltage for the first time in 1992, was discovered by Shahinpoor [31], Oguru *et al.* [32] and Shadeghipour *et al.* [3]. Shadeghipour *et al.* developed a novel accelerometer cell based on nafion coated with platinum and also calibrated it. Shahinpoor prepared swimming robotic structures using ionic polymer gel muscles. In this conceptual design, all the onboard electronics was to be placed in a head-like structure. A flexible membrane was attached to this head and was filled with an aqueous ionic gel. It was possible to electrically control the expansion and contraction of this polymeric fiber filled gel. Oguru *et al.* showed that upon given voltage, mechanical deformation is produced across the thickness of a nafion based membrane. Scanning electron microscope (SEM) image of a nafion based IPMC is shown in **Figures 2**. **Figure 3** shows the chemical structure of nafion based IPMC [28,33].

3. Working principle of IPMC

The actuation response of IPMCs were discovered in 1992 by Shahinpoor, Sadeghipour *et al.* and Oguro *et al.* [3,31,32]. When an adequate potential ($\sim 1-5$ V) is applied across the thickness of an IPMC actuator, the hydrated counter cations are able to break away from the fixed anionic groups and move toward the negatively charged surface. The extent of bending is related to the magnitude of the applied potential and the direction of the bending motion is reversed by reversing the polarity of this potential. The polymer membrane allows selective ions to pass through the membrane while blocking others ions in the hydrated state. The water present on the surface of IPMC serves as a medium for ion migration, so the quantity of water molecules present on the surface of the IPMC is a very important factor which influences the performance of the IPMC. During hydration, water molecules surround the cations and make the entire hydrated entity mobile. There are two possible physical forces responsible for actuation of IPMC. One is the fluid-induced swelling force [34]. In this case when an electrical potential is applied across the IPMC actuator, cations are redistributed and move towards the cathode side (cation rich region) bringing with their water molecules resulting in a locally imbalanced net charge density which produces internal stresses acting on the backbone of polymer causes differential swelling across the polymer thickness. In the anion-rich region, the polymer chains relax, while in the cation rich region, they further extend, consequently, the IPMC will bend towards the anode side (cation poor region). The force is the electrostatic force that arises due to the imbalanced net charges in the polymer membrane [35]. On the other hand, if a mechanical force is applied to the IPMC and makes it bend, the ion movement

will generate sensing charges on the electrodes. So an IPMC can be used both for sensing and actuation [35,36]. The pressure from the strained polymer matrix causes water to diffuse out of the cation rich areas, resulting in a slow relaxation toward the cathode. Under dry conditions, the cross-linked cations are not free to move. However, in wet condition (on hydration) cations are surrounded by water molecules to make the whole film mobile. Thus, transduction in the IPMCs film is because of the movement of cations with the water molecules [37]. They operate best in a humid environment and can be made as self-contained encapsulated actuators to operate in dry environments as well. They have been modelled as both capacitive and resistive element actuators that behave like biological muscle and provide an attractive means of actuation as artificial muscles for biomechanics and biomimetics applications [38]. Transport of hydrated cations within an IPMC beam under the applied voltage and associated electrostatic interactions lead to the bending of an IPMC beam. The responsible factors are:

- a) Swelling and contraction caused by the water content.
- b) Electrostatic force generated by deviation of fixed charges of sulphonic acid groups.
- c) Momentum conservation effect concerning the ion migration and water travel.
- d) Conformation change of the polymer structure according to the ionic migration.

Figure 2: SEM image showing surface morphology of a nafion based IPMC membrane (a) and actual view of fabricated nafion based IPMC membrane actuator (b).

Figure 3: Chemical structure of nafion (perfluorinated sulfonic acid ionic polymer).

This conformation change actually represents bending of the polymer. The transduction principles of an IPMC are illustrated in **Figure 4**.

A bending response under the influence of a low voltage of about 1-5 V of an IPMC was first reported by Oguro *et al.*[32] and Shahinpoor *et al.* [31] also reported similar findings at the same time. Use of mechanoelectric transduction for the development of a pressure sensor has been reported by Sadeghipour *et al.*[3]. Relatively high bandwidth (several hundred Hertz), flexibility, operational feasibility in aqueous environment and chemical stability are some of the advantages inherited by IPMCs. A lifetime of up to 250000 cycles has been recorded for an ionic liquid coated IPMC operating in air [39]. Hunter *et al.* [40] did an extended comparison of the performances of artificial actuators. Comparison of some of the smart materials with strength [10] is shown in **Table 5**. Thus, from the **Table 5**, it can be concluded that IPMC has potential as an actuator in low-mass, large bending displacement, sensors and other applications. From static test results such as those blocking force tests and displacement, an IPMC actuator can be corresponding to a composite beam [41]. Coulomb forces and diffusion are associated with reverse effects and considered as bending [35].

Figure 4: Graphical representation of bending mechanism of IPMC membrane actuator.

Table 5: Comparison of an IPMC with other smart materials [10].

S.No.	Smart Materials	Strain (%)	Stress (MPa)	Efficiency (%)
1	Magnetostrictive	0.2	-	< 50
2	Piezoelectric	0.1	35	> 75
3	SMA	> 4	> 300	> 3.8
4	IPMC	> 40	0.3	> 30

Generally, IPMC is capable of actuating and sensing in harsh conditions. It can responds at low temperature as -100°C where an increase in voltage allows compensation for the loss in efficiency. Furthermore, IPMC based artificial muscle becomes less conductive because decrease in temperature increases electrical resistance. The results come out to resist the generally accepted fact that resistance of metallic conductors with

increasing/decreasing temperature increases/decreases respectively. It has been reported that the IPMC displays less ionic current activities and becomes less active at lower temperature [28]. Upon varying the applied frequency signal the point where large deformations are observed at a critical frequency called resonant frequency, beyond which the actuator response is diminished. Mechanical hysteresis and time varying performance is the main drawbacks of IPMC materials. With the research over the past decade, the materials have become more stable but these problems are not completely resolved. In the recent years, research pertaining to IPMC materials has prolonged more in terms of better adaptability to the environment applicability and getting higher efficiency [42,43].

4. Structure of IPMC

As previously mentioned, an IPMC actuator imparting a layered structure consists of a polymer or composite material membrane coated with novel metal electrodes. Scanning electron microscopes of an IPMC membrane actuator, shows a maximum of three layered (**Figure 5**). The intermediate polymer layer is followed by outer most layers of metal electrodes on both the surfaces. The intermediate polymer layer comprises of Pt, Au or Ag metal particles dispersed or deeply embedded inside the polymer matrix. **Figure 5** represents the cross sectional view of three layers in an electroless plated IPMC membrane. In a sputter-coated sample the number of layers is limited to two: the polymer layer and the electrode. Each layer contributes to the overall electromechanical and mechanoelectric effects in the IPMC. Research has been conducted to understand the electrode layer comprising of porous electrodes [44–48], the intermediate layer [49–51] and the polymer layer [52–56]. The role of these different layers in the transduction of IPMC actuator can be better explained in the following section.

Figure 5: Scanning electron micrograph shows the three layers in an electrolessplated nafion based IPMC membrane actuator [34].

4.1 Polymer layer

Typically, an ion-exchange composite and polymer membrane with selective ion diffusivity and ion conductivity are used for fabricating an IPMC actuator. The ion-exchange membrane acts as a pathway for selective ions to move. The transduction in the IPMC actuator depends on the counter ions, ionomer and the state of hydration of the polymer membrane. Two ionomers with the name of nafion (perfluorosulfonate) by DuPont or flemion (perfluoro carboxylate) by Asahi Glass are commonly available. The major differences between these two ionomers are in the ion-exchange capacities and functional groups (nafion having sulfonate and the flemion having carboxylate groups). The carboxylate group has better mechanical strength and higher ion-exchange capacity corresponding to the sulfonate group. Larger ion-exchange sites cause a higher interfacial area between the metal electrode and the polymer in flemion based samples [57]. A flemion based IPMC has higher water uptake, larger bending, higher stiffness, no back relaxation, higher dielectric strength, better mechanical strength and higher capacitance than nafion based one [58,59]. However, due to commercial availability, nafion is used as

a base polymer for fabricating IPMC because of several advantages such as higher working temperature (up to 120°C), chemical resistance and high ion conductivity of around 0.1 S cm^{-1} in 1 M H_2SO_4 at 20°C measured using a dc current pulse as compared to other polymers [60,61]. Nafion is a perfluorosulfonate ionomer that contains a poly(tetrafluoroethylene) backbone and perfluoroether side chains that terminate in a sulfonate group. The anions are attached to the polymer chain while the cations are free to move upon hydration [62]. Its derivatives can be synthesized normally by the copolymerization of a derivative of a perfluoro (methyl vinyl ether) with sulfonyl acid fluoride and tetrafluoroethylene. Beside the various advantages, perfluorinated polymers have some limitations including low blocking force, back relaxation, high cost and environmental unfriendliness. Therefore, to replace the perfluorinated polymers and improve the performance of IPMCs actuators, non-perfluorinated EAPs are considered to be important alternatives [63]. It has also been shown that the IPMC deflection and force are positively affected by an increase in ionic conductivity. Since the key role of the ionomers in the transduction of IPMC is charge transport, hence an attempt to improve the ionic conductivity has been made. Other properties that affect the transduction capabilities are the ion-exchange capacity, water uptake and elastic modulus.

4.2 Layer of electrode

The electrode layer plays an important role in IPMC transduction. Different electrode properties like electrode thickness, morphology and the types of metal used influence the performance of IPMC actuators. It is also much easier to modify the electrode layer to change the IPMC properties than the ionomer or intermediate layers.

4.3 Morphology of electrode

The morphology of electrode affects the transduction property of an IPMC actuator [64]. The overall capacitance of the IPMC increases as the surface area of the electrode increased. Porosity and thickness of the novel metal electrode play a very important role in controlling the resistance of the electrode [45,65]. On the other hand, the particle gap, thickness and the distance between the surfaces of particles in the intermediate layer directly affect the capacitance of the electrode. In addition, the transduction of IPMC actuator is affected by the surface area of the electrode, which in turn is controlled by the morphology. IPMC transduction can also be improved by increasing the surface area which improves the capacitance of an IPMC. The effect of surface roughening on IPMC properties was studied by Jin *et al.* [66] and Tiwari [67]. Both studies confirmed that transduction in IPMC is improved by roughening. An improvement in tensile modulus and capacitance due to better electrode adhesion and electrode penetration was observed. The electrode morphology is also affected by the type of metal used for electroding. Gold

and palladium electrodes form smoother electrode surfaces thus show better transduction [68,69].

4.4 Thickness of electrode

Increase or decrease of the electrode thickness and conductivity positively affects the transduction in an IPMC. This is because the resistivity, ρ , of the electrode is inversely proportional to the charge transfer:

$$\frac{dQ}{dT} = \frac{VL_tW}{2h\rho} \dots\dots\dots (1)$$

Where Q is the charge, V is the voltage across the thickness of the electrode ($2h$), length (L_t) and width (W) of the IPMC.

Table 6. Effect of plating cycle and reduction temperature on the particle size and capacitance of an actuator [2,49].

S.No.	Studies	No. of plating cycles			Reduction temperature (°C)		
		1	2	3	30	40	50
1	Capacitance @10 Hz (mF cm ⁻²)	1	1.9	2.48	0.2	0.5	1
2	Surface electrode thickness (μm)	1.5	2.5	3	0.9	1.1	1.5
3	Particle size (nm)	60 (±10)	85 (±15)	110 (±20)	40 (±20)	75 (±15)	90 (±10)
4	Sheet resistance (Ω/sq)	9.8	4.95	4.3	13	11	6
5	Intermediate layer thickness (nm)	300	200	<200	1500	400	200
6	Particle gap in intermediate layer (nm)	100	50	<50	<200	<100	<50

Lee and Yoo [70] studied the effect of alcohols (methanol, n-propanol and ethanol) on the thickness of the electrode layer in the first deposition step. It was found that ethanol

produced more uniform and thicker layer whereas in cases of propanol and methanol a thinner electrode were produced compared to water based samples. An alcohol based samples was found to have higher surface resistance than water based sample (methanol: $9 \text{ k}\Omega \text{ cm}^{-1}$; ethanol: $3.8 \text{ k}\Omega \text{ cm}^{-1}$; 1-propanol: $7 \text{ k}\Omega \text{ cm}^{-1}$). It was observed that the electrode thickness increases with the number of plating cycles used for electroding and the reduction in temperature employed for the fabrication of IPMC actuator. The first reduction temperature mainly affects the electrode thickness while the second reduction temperature has more effect on the electrode density [71]. The results of Kim et al. [45] study summarized in **Table 6** shows the effect of temperature reduction and the plating cycle on electrode properties. An increase in capacitance and particle size was observed with the reduction temperature and plating cycle.

4.5 Electroding metal

The hydration is required for IPMCs operation hence care must be taken with the type of metal used for electroding. Platinum [72,73] and gold remain stable under hydration, resist corrosion and have high conductivity, hence are commonly used for electroding. Platinum has good chemical stability and availability in different cationic forms. The properties of platinum electrodes could be modified by using palladium [68] or silver nano powders [48]. Gold electroding on IPMCs actuator also offer several advantages such as high electric conductivity, softness, non-toxicity and high conductivity ($454\text{-}500 \text{ S cm}^{-1}$) but a low surface area of $0.4\text{-}0.75 \text{ m}^2 \text{ g}^{-1}$. Three different techniques have been used for electroless plating of Au electrode: galvanic displacement, autocatalytic and substrate-catalyzed. The impregnation reduction (IR) technique for gold electroding was patented by Fujiwara *et al.* [74,75]. Gold flakes have also been used for electroding [76,77]. Despite their favorable properties both gold and platinum are expensive.

Due to high availability (in the form of a powder, woven cloths, belts, fibers [78] or tapes), low price, specific capacitance ($10 \mu\text{F cm}^{-2}$) with a density of around 0.6 g cm^{-3} and large surface area of $1900 \text{ m}^2 \text{ g}^{-1}$, carbon black has been widely used for electroding [76,79]. However, electroding using hot pressing causes the carbon black electrode to delaminate. Carbon nanotube (CNT) has demonstrated large force generation and small strain of around 0.1-1 % [80]. These advancements have led to the manufacture of IPMCs with CNTs as electrodes [71,80–85]. Single wall nanotubes (SWNTs) have a high surface area of $1000 \text{ m}^2 \text{ g}^{-1}$. However, the poor dispersion inside the membrane and high cost limit their application [86]. Iridium [87] and ruthenium [88] due to their specific capacitances and large surface areas of $80\text{-}1000 \mu\text{F cm}^{-2}$ and $50\text{-}70 \text{ m}^2 \text{ g}^{-1}$ respectively, are preferred over Au and Pt. Other metals such as silver, lead [89], copper [90,91], palladium [72] and nickel [69,89,92] have also been used for electroding because

of their similar properties to Pt and Au electroded IPMCs actuators. However, nickel, silver and copper are corrosive and have limited applications in aqueous media. Organic conductors such as polyaniline (PANI), polypyrrole and Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) have also been used for the development of all organic conductors with a typical conductivity of 100 S cm^{-1} [93].

4.6 Intermediate layer

The research on IPMCs actuators has been focused mostly on the polymer layer and the electrode layer. The intermediate layer plays an important role in the transduction of IPMC by affecting the interfacial area between the polymer electrode and the IPMC capacitance. The role of the intermediate layer in IPMC transduction has been overlooked [67,94]. It has been observed that with an increase in the interfacial area, bending responses as well as the charge output of IPMC have improved. Sunghee *et al.*[95] demonstrated the importance of optimizing the interfacial area. They observed that penetration of platinum particles inside the polymer membrane increases after soaking and reaches to saturation after 3 h of immersion. They also concluded that after fourth plating cycle when saturation in the interfacial area was occurred further plating leads to an increase in the electrode stiffness, hence reduce the displacement of the IPMC membrane tip under applied electric potential. Mechanical deformation in the IPMC membrane occurs due to charge accumulation at the interface between the polymer and the electrode because of a stronger correlation between capacitance and strain. The existence of this relation between the actuation of an IPMC and its capacitance was experimentally demonstrated by Akle *et al.*[96].

5. Manufacturing techniques

Multiple studies and various efforts have been performed to manufacture IPMC actuators, but the variability from sample to sample created difficulty to draw definite conclusions and repeatable results that may be readily reproduced [48,97–100]. Fabrication of IPMC starts by selecting the base ion-exchange polymeric materials, typically manufactured from organic or organic-inorganic composite polymers with covalently bonded fixed ionic groups [46,101,102].

The most commonly used ion-exchange materials for the fabrication of IPMCs are perfluorinated hydrocarbon: with short sidechains terminated by ionic groups (*i.e.* nafion from DuPont^{TM}) [34], typically sulfonate or carboxylate ($-\text{SO}_3^-$ or $-\text{COO}^-$) for cation exchange or ammonium cations for anion exchange [103–108]. Perfluorinated ion exchange materials are organofluorine compounds with all hydrogen replaced by fluorine

on a carbon chain having at least one functional group or different atoms. The chemical formula of a typical perfluorinated compound of nafion is shown in **Figure 3**.

The mechanical strength of IPMC materials is determined by the large polymer backbones, while the short side-chains provide ionic groups that interact with the inner solvent and the passage of selective ions. These features are different in other ionic polymers that are primarily limited by cross linking such as styrene/divinylbenzene families, the ability of the ionic polymers to expand (due to their hydrophilic nature). Fabrication of IPMC actuator is a lab-intensive work hence safety measures should be taken in consideration during chemical processing. The initial compositing process and surface electroding process are the two distinct IPMC fabrication processes. Based on the preparation processes, the morphologies of precipitated metals significantly differ.

The purpose of the compositing process is to metalize the polymer membrane by immersing polymer membrane in a salt solution of platinum-containing cations via the ion-exchange process. A proper reducing agent (LiBH_4 or NaBH_4) is used to metalize the polymer. However, the distribution of metallic particles across the polymer membrane is not homogeneous but concentrations of metal particles are nearer to the interface boundaries. Kim and Shahinpoor [109] have presented a detailed description of various techniques and procedures used in manufacturing of IPMCs including their performance and comparison.

The surface electroding process is carried out by using proper reducing agents to carry out reduction, predominantly on the initial platinum electrode layer on the membrane surface formed by the initial compositing process leading to a rather smooth metallic surface. The mechanical and electrical behavior of IPMC is also influenced by the variation of the deposition of electrode material. The process of deposition of metal electrodes on the membrane surface is known as electroding. Irrespective of the method used, electroding is aimed at:

- a) Deposition of metal particles on both the surface of the membrane.
- b) Formation of a uniform metal layer that is chemically and mechanically stable.
- c) Maximization of interfacial area between the electrode and membrane.
- d) Minimization of electrical resistance of the electrode.

The first step is to roughen the material surface where it will serve as an effective electrode. This involves sand blasting or sand papering the surface of the polymer in order to increase the surface area density where platinum salt penetration and reduction occur, as well as ultrasonic cleaning and chemical cleaning by acid boiling (HCl or HNO_3 , low concentrates). The second step is to incorporate the ion exchanging process

using a metalcomplex solution such as tetraamine platinum chloride hydrate as an aqueous platinum complex ($[\text{Pt}(\text{NH}_3)_4]\text{Cl}_2$ or $[\text{Pt}(\text{NH}_3)_6]\text{Cl}_4$) solution. To carry out the ion-exchange process, immerse the membrane in the Pt salt solution, adding ammonium hydroxide solution (5%) to neutralize the solution and leave the membrane at room temperature for a few hours (preferably overnight) [10,63]. Although, the equilibrium condition depends on the type of charge of the metal complex, such complexes were found to provide good electrodes. The third step (initial platinum compositing process) is to reduce the platinum complex cations to the metallic state in the form of nanoparticles by using effective reducing agents such as an aqueous solution of sodium or lithium borohydride (5%) at favourable temperature (*i.e.* 60°C).

Before the reduction is carried out, rinse the polymer membrane with DMW with constant stirring at 40 °C, add 2 mL of the aqueous sodium borohydride solution (5 wt% NaBH_4 aqueous) every 30 minutes for several times while gradually raising the temperature up to 60 °C. Add 20 mL of prepared aqueous solution of NaBH_4 and stirred for 1.5 h at 60 °C. This gives a black layer of Pt particles deposited on the surface of the polymer membrane. To terminate the reduction process, rinse the membrane with DMW followed by immersing in 0.1 N HCl solution for 1 h.

A small amount of sample is warmed with NaBH_4 to check the end point. Precautions should be taken because adding reducing agent (NaBH_4 powder) in a hot solution can cause a gas explosion. If there is any trace of Pt ion remains in the plating solution, the colour of the solution turns to black. In such case, continue deposition of Pt particles with addition of $\text{NH}_2\text{OH}\cdot\text{HCl}$ and NH_2NH_2 solution, until no Pt ion remains.

Finally after washing, treat the membrane with dilute 0.1 N HCl. H^+ in the composite membrane can be simply exchanged for any cation by immersing it in a solution of the chloride salt of the cation, e.g., LiCl in order to exchange it into the lithium cation form. Palladium, silver, gold, and copper have also been successfully used as electrodes. The primary reaction for platinum composites is:

Other IPMC fabrication processes are described below.

5.1 Physical metal loading

Fabrication of IPMCs by physically metal loading method is a novel technique which is faster than conventional methods [47]. The principle of this technique involves two simple steps. First of all, a conductive primary powder is physically loaded into the ionic polymer, forming a dispersed particulate layer. In the second step, the chemical plating

method is used to secure smaller secondary particles using reducing agents, similar to the conventional methods. Furthermore, an electroplating process can be applied to integrate the entire conductive phase intact, which serves as an effective electrode.

5.2 Casting method

The performance of the IPMC membrane varies with its thickness. Hence to achieve a desired thickness of IPMC film other than commercialized thickness, the casting method is widely used. However, the casting method has a reproducibility problem because it requires great attention to process variables such as temperature and concentration of specific solvent [110]. According to the technical information concerning nafion [34], the volume of nafion dispersion is approximately ten times the volume of cast nafion. The overall casting process consists of three steps (a) proper mixing, (b) constant stirring and sonicating and (c) thermal treatment. The mechanical stiffness of the cast polymer film is enhanced by thermal treatment by increasing strength of the molecule's bonding structure. Finally, the polymer film has to be treated with the H_2O_2 solution at temperature ranging between 75-100°C for 1 h followed by boiling in demineralised water for 1 h. This method has been used to achieve IPMC actuators of desired thickness [111].

5.3 Hot-pressing method

The hot-pressing method commonly uses a hot pressing machine to make several thin polymer films to adhere together, which enhances the force performance, reproducibility and bending stiffness (**Figure 6**). Easy to control the thickness, repeatability and simplicity are the main advantages of this method [110]. Preparation of IPMC actuators by this method is described below.

Figure 6: Fabrication of IPMC by stacking and hot press techniques.

Take the polymer films or membranes with proper dimensions, rinse with acetone followed by stacking in a mold with polyimide film. The mold is then pressed at 180°C without pressure for 20 min. in preheated presses, followed by pressing for 10 min. at 50MPa. Cooling the polymer films to ambient temperature in the air then treated with 3wt% sulfuric acid at 70°C for 1 h. Finally treat the film with 10wt% H₂O₂ at 70°C for 1 h followed by rinsing with DMW for 0.5 h. Ion-exchange process is carried out by immersing the stack membrane in an aqueous solution of platinum ammine complex ([Pt(NH₃)₄Cl₂]) for 1-2 days. After rinsing the film with DMW, place it in 500 mL of stirring water at 40°C and add 5 mL of 5wt% aqueous solution of NaBH₄ after every 30 min. at 60°C.

5.4 Electroless plating method

There are several fabrication methods including sputtering, taping with conductive film, or chemical reduction among others. However, electroless plating is the best process to fabricate IPMCs, using conductive metal ions (platinum or gold). Generally, IPMC is fabricated by the chemical reduction method due to the low cost and uniform surface coating with metal by using this method. The adhesion of metal electrode on the surface of the polymer membrane is one of the main problems during the IPMC fabrication. The electroless plating technique can also be used with other methods like electroplating, polymer coating and hot-embossing to modify the adhesion between electrode and polymer, decreasing fabrication time and lowering the cost [21]. Chung *et al.* [112] prepare the EAPs membrane by using a solution casting method and electroless plating for platinum electroding for the study of electrodes behaviours, membrane properties and processing conditions. Their study clearly shows that the smoothness of the surface increase by decreasing the surface roughness to 3 nm from the typical 52 nm but the surface resistance was too high to activate the bending motion. They coated the electrodes again in order to reduce the surface resistance, the fabricated IPMC showed longer bending life time than typical IPMC. Zhang et al. [113] fabricated the IPMC actuator based on Ag-nafion using penetration reduction and chemical plating methods for electroding palladium, platinum and gold due to their high conductivity and corrosion resistance. Metals such as copper have also been used as electrodes [114]. Electrode deposition using sputter coating and electrode deposition on dry and solvated membranes, using a direct assembly process, have also been considered [96,115]. However, the most commonly used manufacturing technique is electrode deposition because by using this technique the mechanical and chemical stabilities of electrodes are enhanced [72]. Kim *et al.* [111] have reported that the performance of IPMCs is directly affected by the concentration and size distribution of platinum particles on the membrane surface. The performance of IPMC can increase by using some of these techniques.

- a) The multiple platinum reduction process has been effective to produce IPMCs exhibiting larger force characteristics.
- b) During the platinum platinization process, use of dispersing agents gives significantly improved force characteristics showing a quick response upon applied electric potential with increased force generation.

The base material stretching process results in the higher permeability compared to the unstretched base material platinum composition process. It was clear from these techniques that the particle penetration within the material is much more effective, and forms a much denser platinum particle phase and distribution.

5.5 Micro fabrication method

Chung *et al.* [48] proposed micro-fabrication technologies which integrates the physical and chemical methods for the fabrication of IPMC actuators free from surface roughening pretreatment for good adhesion reducing both cost and fabrication time. The basic principle of this method is the casting of silver nano-powders with polymer composite followed by plating silver metal electrode by electroless plating method.

6. IPMC actuator: characteristics, performance and measurements

6.1 Generative force

A larger actuation displacement and force with reasonable actuation frequency is necessary for the successful application of IPMCs membrane actuators. The blocking force is mainly dependent on the length of IPMC, thickness and width. According to Kim *et al.* [41], the blocking force is proportional to the square of the thickness of IPMC and linearly proportional to the width and inversely proportional to the length of the IPMC.

With thick IPMC membrane, a high tip force can be generated for working relatively longer duration while its tip displacement and response speed are decreased. A long IPMC membrane cannot produce a high tip force but can create a large displacement. Tip displacement along with the tip force increases with increase in the applied voltage. In this case, electrolysis is accelerated which increase the evaporation of solvent by the heat generated from the IPMC actuator resulting in poor repeatability.

Therefore, a trade off in the actuation displacement, response speed, actuation force and endurance must be considered during the development of IPMCs actuator. Reduced surface resistance of metal electrodes in an IPMC also contributes to improvement of actuation performance and repeatability at the same time.

6.2 Frequency response test

While the deformation and blocking force characteristics of IPMC might be changed due to the micro cracks of the surface electrode that can increase the surface resistance, the natural frequencies are little affected by the surface electrode condition[41]. Therefore, the results of a frequency response test are used to establish the equivalent Young's modulus of IPMC. Frequency response of IPMC contains not only the structural characteristics of IPMC but also electrical characteristics. It was seen that the electrical characteristics change the IPMC system, while the 1st and 2nd natural frequencies are quite similar confirming that the electrical characteristics do not have an effect on the 1st and 2nd natural frequencies. Mechanical characteristics of IPMC also depend on the frequency, generally as the frequency increases there is corresponding decrease of displacement. At low frequencies (0.1~1.0Hz) and an imposed voltage, the effective elastic modulus of the IPMC cantilever strip is small, while at higher frequency (5~20Hz), the elastic modulus is high and the displacement is smaller. It happens due to the simple fact that during low frequency the hydrated ions have time to gush out of the surface electrode while in the high frequency they are rather contained inside the polymer.

6.3 Characteristics of IPMC under the variation of thickness

Nakamura *et al.*[116]established the relationship between the structure of IPMC and force generation as well as process of fabrication by investigating electromechanical and electrochemical characteristics of IPMC using impedance analysis. This study reveals that the generated force is dependent on the repeated plating, preheating and thickness of IPMC. The generated force almost linearly increases for the current range of 0-300 mA. It is assumed that the movement of counter ions followed by trapped water molecules being driven by the applied voltage on both sides of IPMC actuator. It was also seen that the generated force and capacitance of IPMC increased with the increase of the thickness but show non linear behaviour. A similar behaviour was also observed with resistance, an increase in resistance resulting from increasing thickness but the trend of resistance was almost linear to the thickness [117].

6.4 Characteristics of IPMC under the variation of input voltage

As the applying voltage increases, both bending displacement and the generative tip force increase. The linearity of the bending displacement and generative tip force is improved, as the thickness gets thinner. This is because as the thickness of the actuator gets thin, the uniformity of the electrodes is improved [10].

6.5 Effect of different cations

Electromechanical and chemical responses of IPMCs actuator depend on the nature of ion-exchange polymer, the conductivity and morphology of the metal electrodes, cations, and solvent uptake [58,118,119]. The performance and bending displacement towards the positive electrode depend on the type of solvent used. Different alkali metal cations acquire different speeds and total tip displacements for the same IPMC membrane.

After performing the experiments, Kim and Shahinpoor [109] realised that the properties of IPMC differ depending upon different cations. For this nine samples were used and each of which was soaking in an appropriate salt solution (1.5N of NaCl, LiCl, KCl, HCl, CaCl₂, MgCl₂, BaCl₂, CH₃[CH₂]₃NBr, [CH₃]₄NBr), respectively at modest temperature (30°C), for 3 days. **Figure 7** shows the blocking force of different IPMC containing various cations relative to the Na⁺ ions containing IPMC at zero displacement[109]. From this figure it is clear that Li⁺ containing IPMC is superior. It clearly shows that hydration processes with respect to mobile cations play an important role in the actuation behaviour.

Figure 7: Effects of various cations on the actuation of the IPMC muscle [119]

7. Application

IPMCs have many advantages such as (1) produce high displacement, (2) require low drive voltage, (3) can operate very well in wet environment and (4) can be cut into small strips or any desired form. Therefore, IPMC have significant potential in low mass, high displacement actuation and other applications. **Figure 8** demonstrates the applications of IPMC in different fields, a few are discussed below.

Figure 8: Applications of IPMCs in different areas.

7.1 Robotics application

For years efforts have been made in the direction of creating humanoids or robots, which resemble man. With the development of IPMC the goal of creating robots which can walk, talk and more importantly, show emotions like man appears to be closer. Hence, IPMC shows a potential to act like human muscles, thus we might be able to build robots which are closer to a human. Fabrication of biomimetic robots requires (a) development of specific circuitry, (b) power and regulators integrated into the mobility elements to allow a limited degree of self-powered maneuvering [1]. A conceptual design of a swimming robot structure using IPMC was presented by Shahinpoor [31]. Such a swimming robotic structure in the form of a submarine structure partially encapsulated in a flexible membrane filled with an electrolyte like water. Incidentally, the first commercial application of IPMC was a swimming fish robot developed by Eamex, Japan which was exhibited recently. Bar-Cohen *et al.* designed and fabricated a four finger based gripper with hooks miniature robotic arm at Jet Propulsion Laboratory (JPL) [120].

7.2 Space application

To meet the most challenging and technologically demanding space applications new technologies and materials find immediate application in space need to be developed. Bar-Cohen *et al.* developed a planetary dust wiper based on IPMC to remove planetary dust particles from the surface of a Nanorover [30]. It was a joint effort of National Aeronautics and Space Administration (NASA) and National Space Development Agency of Japan (NASDA). NASA found that operations on Mars involve an environment that causes accumulation of dust on hardware surfaces. Hence, they proposed a planetary dust wiper. This dust may cause serious damage to delicate and precious instruments such as a high-resolution camera, etc.

7.3 Human-machine interface

There is demand to develop better human-machine interfaces to help our sensory system efficiently. For example, a highly developed interface is needed for doctors to avoid realizing the difference due to his physical absence in telesurgery. Haptic devices and tactile sensors are some of the important human-machine interfaces [1]. To provide a human operator, artificial tactile feel display with the required stimuli was produced using IPMC actuators [121]. The idea of using EAPs in active tactile display device to present textual and graphical information to a blind person was presented by Bar-Cohen. The display medium can be constructed as a planar array of small cones called ‘reading pins’ [1].

7.4 Medical applications

Ease of operation, flexibility, softness and large displacement similar to biological muscles make the IPMC good candidates to operate as substitutes for human muscles [1]. Shahinpoor [122] used IPMC as artificial muscles. A human body contains large percentage of water and IPMC operates very well in presence of a solvent medium like water which makes it a very good material for making artificial muscles. They can be inserted into the body to perform surgeries, a closed loop real-time control system can control it and a haptic interface like joystick can be used in this closed loop. IPMC can also be used in developing micro-robots, which can guide catheters in the blood stream [1].

7.5 Micro-manipulation applications

Since the last decade there has been tremendous escalation in the field of micro-electro mechanical systems (MEMS). Presently MEMS devices such as micro-pumps fail to achieve very high displacements or forces because of many limitations in terms of applied voltage or energy consumption. EAPs in general and IPMCs in particular because of their large displacements and low power consumption, can be a suitable material in MEMS [1]. IPMC actuators are well suited in micromanipulation devices due to easily micromanipulation in micro-assembly of small parts, in micro-fabrication, etc. Lumia and Shahinpoor gave a design of a micro-gripper, which used both the actuation and sensing capabilities of IPMC [123]. Kim and Bhat initiated an idea of using IPMC strips as fingers for a micro-gripper system [124].

8. Research dimensions

Typically, perfluorinated polymer-based ionomers membranes with the trade name of nafion are being used as actuators and dynamic sensors are commercially available for IPMC actuator applications because of their high proton exchange capacity, thermal, mechanical and chemical stabilities [8,96,104,105,125–129]. However, these polymers have certain drawbacks such as loss of proton conductivity at temperatures higher than 80 °C, high cost of fabrication, hazardous nature, small actuation bandwidth, low blocking force, little humidity due to drops in proton conductivity, as well as membrane dehydration. Therefore, the main objective of this work was to find suitable non-perfluorinated ionic polymers/composite materials as alternative to the perfluorinated ionic polymers for IPMC actuators. Non-perfluorinated polymers have various advantages such as good film forming capability, short processing time, low cost of fabrication, good flexibility, high thermo-mechanical stabilities and easy modification of chemical properties such as ion-exchange and water holding capacities and proton conductivity [8,33]. Therefore, non-perfluorinated polymers based IPMC actuators can provide an easy and reliable solution for the realization of novel actuators as well as the possibility of industrial applications. It is stimulating to build smart material actuators, which can accomplish bio/micro manipulation, bio-sensing, and micro robotic tasks.

The goals of this work are as follows:

Design an IPMC actuator with a low cost and high reliability manufacturing process modified from other previous protocol.

- a) To verify the actuator by a series of dynamic testing in an aqueous environment.
- b) To control the bending motion of the actuator by implementing a PID controller.

9. Literature review

A vast variety of bending actuators based on different types of EAPs have been developed during last decade. It is very difficult to compile all the work carried out in this field. Thus, an attempt has been made to arrange the work carried out during last 15 years. The work published on perfluorinated and non-perfluorinated polymer/composite based different types of actuators during the last fifteen years has been presented briefly in **Table 7**.

Table 7: Perfluorinated and non-perfluorinated polymer or composite based IPMC actuators.

S.No.	Title	Remarks	Ref.
Perfluorinated polymer or composite based actuators			
1	Bending of polyelectrolyte membrane platinum composites by electric stimuli Part II. Response kinetics	A solid composite polymer electrolyte membrane-platinum (SPM-Pt) showing bending response with electric stimuli. A model was developed in which mechanical deformation was applied to the kinetics of the bending response. In the solid polymer electrolyte membrane (SPM) under electric fields via electro-kinetically induced pressure gradients.	[8]
2	Electric field sensitive neutral polymer gels	In this paper, the effects on the motility and shape of polymer gels induced by electric field and new driving mechanism to induce bending deformation of a neutral polymer gels was described.	[130]
3	Morphology of electrodes and bending response of the polymer electrolyte actuator	In this study, the bending behavior of a perfluorocarboxylic acid polymer film plated with Au metal electrode was measured by applied potential in water or saline solution.	[64]
4	Bending deformation of neutral polymer gels induced by electric fields	This study showed when finely distributed colloidal particles were incorporated into a swollen neutral polymer gel bending response under electric field occurs.	[131]
5	State of water and ionic conductivity of solid polymer electrolyte membranes in relation to polymer actuators	Examination of state of the water and the ionic conductivity of various ionic forms of solid polymer electrolyte membranes upon polymer actuators was discussed.	[132]
6	The effects of counter ions on characterization and performance of a	For perfluorinated polymer membrane when exchanged with alkali metal ions as counter ions, the displacement was rapid <i>i.e.</i> fast response under electric	[133]

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	solid polymer electrolyte actuator	stimuli.	
7	Novel ionic polymer metal composites equipped with physically loaded particulate electrodes as biomimetic sensors, actuators and artificial muscles	The novel fabrication method of manufacturing IPMCs actuator based on perfluorinated polymer equipped with physically loaded electrodes as artificial muscles and biomimetic actuators, sensors was proposed.	[47]
8	A novel method of manufacturing three dimensional ionic polymer metal composites (IPMCs) biomimetic sensors, actuators and artificial muscles	Commercially available perfluorinated ion exchange polymer films of proper thickness (100-300 μm) permits fast mass transfer in various chemical processes. Hence can be used as artificial muscles.	[97]
9	Design of high authority flexural actuator using an electro-strictive polymer	In this study, a poly(vinylidene fluoride-trifluoroethylene) polymer based actuator faces laminated with internal electrode has been developed that achieves maximum bending response subjected to an electric field.	[134]
10	Ionic polymer metal composites: II. Manufacturing techniques	Presents a detailed description of various techniques and experimental procedures in manufacturing IPMCs.	[109]
11	Electrochemical characterization of polymer actuator with large interfacial area	Electrochemical and actuating behaviors were strongly dependent on the interfacial area between the polymer electrolyte and electrode. A large surface area composite actuator was fabricated using replication method.	[94]
12	Investigations on actuation characteristics of IPMC artificial muscle actuator	Electromechanical response characteristics of IPMC, having potential for artificial muscle actuators, were investigated using driving methods.	[135]
13	Novel electroactive, silicate nanocomposites prepared to be used as actuators and artificial muscles	A newly developed nanocompositing technique was proposed to optimize the desirable properties of the base ion exchange polymeric materials used for fabrication of EAP actuators and artificial muscles.	[136]

14	Low temperature characteristics of ionic polymer-metal composite actuators	This study describes IPMCs actuators as an attractive solution for cold operation, because of their actuation capability with relatively low electric fields.	[137]
15	A novel fabrication of ionic polymer-metal composites (IPMC) actuator with silver nano-powders	Silver nano-powders applied for the fabrication of IPMC actuator shows good adhesion between polymer membrane and the metal electrodes without surface roughening, high repeatability, low cost and short processing time.	[48]
16	A multi-wall carbon nanotube tower electrochemical actuator	A patterned multiwalled carbon nanotube arrays were synthesized successfully and electrochemical actuation was demonstrated.	[138]
17	A self-sensing ion conducting polymer metal composite (IPMC) actuator	Development of self sensing IPMC actuator as a position sensor.	[139]
18	Characteristics and performance of ionic polymer metal composite actuators based on nafion/layered silicate and nafion/silica nanocomposites	This study shows that the electrical and mechanical properties of the nafion polymer may greatly alter by incorporation of a small amount (3-10 wt.%) of nanoparticulates such as layered silicate (montmorillonite) or fumed silica. These nanocomposites were used for fabrication of IPMCs as polyelectrolytes.	[140]
19	Effects of humidity on the performance of ionic polymer metal composite actuators: experimental study of the back-relaxation of actuators	Study on the effect of water uptake on the displacement, mechanical force, velocity and charging profiles of perfluorinated ionomer-platinum/Li ⁺ based actuators.	[141]
20	An electrode model for ionic polymer metal composites	An IPMC was fabricated by electroless plating of platinum and parametric studies were carried out to estimate the electrical properties (resistance and capacitance) of the electrode.	[45]
21	The cause of nanohole and nano particle	Nanoparticles and nanoholes on the surfaces of gold electrode after actuation	[142]

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	formation on Au electrode after actuation of electro active paper actuator	strongly associated with the degradation of the actuator performance.	
22	Time resolved mapping of water diffusion coefficients in a working soft actuator device	In this study, diffusion weighted imaging was employed to map the distribution of the diffusion coefficient of water in bare, water-soaked, Li ⁺ exchanged, cast nafion and in an nafion based IPMC actuator.	[65]
23	Influence of ordered morphology on the anisotropic actuation in uniaxially oriented electroactive polymer systems	A new anisotropic actuation response was yield by perfluorinated polymer nanostructure when altered by uniaxial orientation, mimicks the structure of biological muscle tissue. IPMCs cut from films oriented perpendicular to the draw direction yield tip-displacement response that is higher than those of unoriented IPMCs actuator.	[143]
24	The role of water in transport of ionic liquids in polymeric artificial muscle actuators	Here in the present study quantifying the delicate interplay between ionic liquid and water absorbed in an ion-exchange polymer membrane as used in ionomeric polymer actuators.	[144]
25	Influence of the conductor network composites on the electromechanical performance of ionic polymer conductor network composite actuators	Investigations regarding the electromechanical performance of the ionic polymer conductor network composite actuatorsinfluence by the conductor network composites (CNCs),manufactured by the direct assembly method with ionic liquids as the solvent was presented.	[145]
26	A highly porous nafion membrane templated from polyoxometalates based supramolecule composite for ion exchange polymer-metal composite actuator	Developments of a new technique for manufacturing a highly porous hybrid SiO ₂ /nafion ionomeric polymer membrane based IPMC actuator, attainingsignificantly high electromechanical performance.	[146]

27	Electrically stimulated gradients in water and counterion concentrations within electroactive polymer actuators	Applications of applied potential atomic force microscopy, neutron imaging, and current sensing atomic force microscopy techniques for investigating actuation mechanism of electroactive polymer actuator.	[147]
28	Enhanced electromechanical performance of graphite oxide-nafion nanocomposite actuator	Graphene oxide/naion based actuators were tested in terms of conductivity, tip displacement, bulk and surface morphology and blocking force.	[148]
29	Ionic polymer metal composite material as a diaphragm for micropump devices	Two different IPMC diaphragms was prepared and experimentally tested. The stroke volume of each IPMCs diaphragm was examined with different values of applied electrical potential.	[149]
30	Electro-chemical operation of ionic polymer metal composites	The potential origin of the relaxation phenomena of IPMC actuator related to electro-chemically induced surface reactions with electrodes were discussed.	[107]
31	Preparation and performance of IPMC actuators with electrospun Nafion®-MWNT composite electrodes	A new IPMC actuator was prepared with nafion-117 ionomer membrane and electrodes made of an electrospun nafion/multiwalled carbon nanotube web were tested. To reduce the surface resistance the composite electrodes were coated with gold layers of 2-3 μm thickness.	[150]
32	Synthesis and performance evaluation of thin film PPy-PVDF multilayer electroactive polymer actuators	A spin coating technique was used to fabricate a porous polyvinylidene fluoride thin membrane with a thickness of 32 μm with electrochemically deposited polypyrrole layers on both sides to make ultra thin membrane polymer actuators.	[151]
33	Equivalent circuit modeling of ionomer and ionic polymer conductive network composite actuators containing ionic	This study described the electrical equivalent circuits that model the complex frequency dependent impedance of 1-ethyl-3-methylimidazolium trifluoromethanesulfonate containing ionic polymer conductor network	[152]

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	liquids	composite devices and electroactive polymer membranes.	
34	High actuation response of PVDF/ PVP/ PSSA based ionic polymer metal composites actuator	The tip displacements of polyvinylidene fluoride (PVDF)/polyvinyl pyrrolidone (PVP)/polystyrene sulfonic acid (PSSA) based IPMCs actuators were measured at low AC and DC voltages. The results were compared with commercially obtain nafion polymer based IPMC actuator.	[105]
35	Enhanced electromechanical response of ionic polymer actuators by improving mechanical coupling between ions and polymer matrix	Poly[(vinylidene difluoride)-co-(chlorotrifluoroethylene)] (P(VDF-CTFE)) and P(VDF-CTFE)/poly(methyl methacrylate) (PMMA) cross-linked blends were used as i-EAP actuators with electrolyte ionic liquid 1-ethyl-3-methylimidazolium trifluoromethane sulfonate [C ₂ mim][TfO] for the first time. The results were compared with perfluorinated nafion or aquivion ionomers.	[153]
36	Enhanced and fast actuation of fullerenol/PVDF/PVP/PSSA based ionic polymer metal composite actuators	This study reports the enhanced and fast actuation of the fullerenol (Ful)/polyvinylidene fluoride (PVDF)/polyvinyl pyrrolidone (PVP)/polystyrene sulfonic acid (PSSA) composite polymer membrane based IPMC actuator, at low AC and DC voltages as compare to the PVDF/PVP/PSSA membrane based IPMC actuator.	[154]
37	Effect of nucleation time on bending response of ionic polymer metal composite actuators	An autocatalytic electroless plating of nickel metal as electrode is attempted to substitute electroless impregnation reduction technique in IPMC actuators to reduce processing time and cost.	[155]
38	Electrochemical and electromechanical properties of high-performance polymer actuators containing vapor grown carbon	The effects of ionic liquid and metal oxide on the electromechanical and electrochemical properties of poly(vinylidene fluoride-co-hexafluoropropylene) polymer based actuators were investigated using a metal	[156]

	nanofiber and metal oxide	oxide containing a vapour grown carbon nanofiber (VGCF)-IL gel electrode.	
39	Ionic polymer metal composite actuators based on triple layered polyelectrolytes composed of individually functionalized layers	In this paper an IPMC actuators were fabricated based on two types of triple-layered nafion polymer membranes via consecutive solution recasting and electroless plating methods.	[157]
40	Repetitive control of a trilayer conjugated polymer actuator	This study demonstrate a polypyrrole based conducting electroactive polymer actuator desired to track repeating commands for position control applications.	[158]
41	Durable and water floatable ionic polymer actuator with hydrophobic and asymmetrically laser scribed reduced graphene oxide paper electrodes	An asymmetrically and hydrophobic laser-scribed reduced graphene oxide paper electrodes were employed to developed a highly durable and water floatable ionic polymer artificial muscle.	[159]
42	Effect of dehydration on the mechanical and physicochemical properties of old-and palladium -ionomeric polymer metal composite (IPMC) actuators.	This paper describe the dehydration effect on physicochemical and mechanical properties of Au (gold) and Pd (palladium) metal electrode based IPMC using water as solvent.	[160]
43	Single active finger IPMC microgripper	A new designcomprising of one stationary and one actuating finger for a single active finger IPMC microgripper was proposed.	[61]
Non-perfluorinated polymer or composite based IPMC actuators			
44	A tunable dendritic molecular actuator	In this study an electroresponsive molecular actuator was developed based upon a diblock copolymer of a negatively charged linear chain and a positively charged dendrimer.	[161]
45	Actuator based on MWNT/PVA	This study reveals the development of first actuator based on multiwalled	[162]

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	hydrogels	carbon nanotube (MWNT) hydrogels with poly(vinyl alcohol) through simple hydrogelation.	
46	Electric field frequency and strength effects on Au electrode damage for an electroactive paper actuator coated with polypyrrole	The causes of electrode damage to an electroactive paper actuator coated with Au and polypyrrole were investigated at various electrical potential frequencies and strengths.	[163]
47	Polypyrrole actuators with inverse opal structures	Paper polypyrrole (PPy) films synthesized by electrochemical polymerization exhibited higher actuation rates than those of the actuators made from flat PPy films.	[164]
48	A mechanical actuator driven electrochemically by artificial molecular muscles	A microcantilever, coated with redox-controllable, bistable rotaxane molecules shows reversible deflections by alternating oxidizing and reducing electrochemical potentials. One surface of micro-cantilever devices were prepared by precoating with a gold film.	[165]
49	Polymer actuator based on PVA/PAMPS ionic membrane: Optimization of ionic transport properties	In this paper, water soluble poly (vinyl alcohol) and highly ionic conductive poly 2-acrylamido-2-methyl-1-propanesulfonic acid (PAMPS) composite was blend to synthesized and develop electroactive polymer as artificial muscle.	[166]
50	Polypyrrole actuator with a bioadhesive surface for accumulating bacteria from physiological media	A gold/polypyrrole based bilayer actuator was prepared by electrochemical deposition, with surface modification with a bioadhesive polymer, polydopamine. The actuator exhibited high actuation performances in physiological media.	[167]
51	Poly(ethylene oxide)-poly(butadiene) interpenetrated networks as electroactive	Molecular dynamics (MD) techniques have been used to study coordination stability and ionic transport in an interpenetrating polymer (IPN) network used	[168]

	polymers for actuators: A molecular dynamics study	as electrolyte for actuator devices.	
52	Electroactive artificial muscle based on crosslinked PVA/SPTES	Based on a crosslinked ionic networking membrane of sulfonated poly(arylenethioethersulfone) copolymer (SPTES) and poly(vinyl alcohol) (PVA), a novel biomimetic artificial muscle with electrically driven bending deformation was developed.	[169]
53	Electric-stimuli-responsive bending actuator based on sulfonated polyetherimide	In this study, biocompatible sulfonated polyetherimide (SPEI) membrane based bending actuator was fabricated by a solution casting method. It shows good electro-chemical properties, IEC, water uptake and proton conductivity required for a fast and high bending response of ionic polymer actuator.	[170]
54	Electroactive polymer actuators with carbon aerogel electrodes	Carbon aerogel was used for fabrication of nanoporous electrodes for EAP actuators. Their electromechanical and electrochemical characteristics were compared with actuators based on carbide-derived carbon and activated carbon electrodes.	[171]
55	High-performance, low-voltage and easy-operable bending actuator based on aligned carbon nanotube/polymer composites	The coefficient of thermal expansion of the polymer matrix can remarkably reduce by incorporating super-aligned carbon nanotube sheets into a polymer matrix (polydimethylsiloxane). The fabricated actuator was easily operable and capable to generate large bending actuation with controllable actuation.	[172]
56	Sulfonated polystyrene-based ionic polymer metal composite (IPMC) actuator	A comparative study was carried out between sulfonated polystyrene (sPS) and perfluorinated nafion based IPMC. The tip displacement, the current density, the response rate and the blocking force were significantly higher for the sPS-IPMC than that of the nafion IPMC actuator.	[173]

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57	Nanoporous carbide-derived carbon based actuators modified with gold foil: Prospect for fast response and low voltage applications	Carbide-derived nanoporous carbon (CDC) based ionomeric actuators were developed in combination with gold foil. The gold-foil modified actuators required remarkably low operating voltage and exhibited high frequency response.	[174]
58	Novel sulfonated styrenic pentablock copolymer/silicate nanocomposite membranes with controlled ion channels and their IPMC transducers	A novelelectrically driven IPMC actuator has been design and fabricated by ionic thermoplastic elastomer, poly((t-butyl-styrene)-b-(ethylene-r propylene)-b-(styrene-rstyrene sulfonate)-b-(ethylene-r-propylene)-b-(t-butyl-styrene)) (tBS-EP-SS-EP-tBS; SSPB) pentablock copolymer and its nanocomposites with sulfonated montmorillonite (s-MMT).	[175]
59	Electric-field-actuation of in situ composites that contain silver-coated carbon fibers in sodium sulfonate ionomers	Anelectric-field-stimulated actuation system was developed by impregnating sodium-sulfonated ionomers, poly(styrene-co-butyl acrylate-co sodium allyl sulfonate, PSBS) with <i>in-situ</i> deposited silver-modified carbon fibers (SCCF).	[176]
60	Imidazolium sulfonate containing pentablock copolymer ionic liquid membranes for electroactive actuators	Thermally stable electroactive actuators have been fabricated by imidazolium sulfonate containing pentablock copolymer ionic liquid composite. The fabricated actuator exhibit effective actuation response under a low potential	[177]
61	Comparative displacement study of bilayer actuators comprising of conducting polymers, fabricated from polypyrrole, poly(3,4 ethylene dioxythiophene) or poly(3,4-propylene dioxythiophene)	A report on comparative study of the displacement of bilayer actuators manufactured using different conducting polymers was presented.	[178]

62	Enhancement of actuation ability of ionic-type conducting polymer actuators using metal ion implantation	The effect of gold-ion implantation on the actuation performance of ionic type conducting polymer actuators has been examined.	[179]
63	Fabrication of ionic polymer actuator with graphene nanocomposite electrodes and its characterization	The polystyrene based ionic polymer actuator was fabricated by conventional solvent mixing using graphene nanocomposite (GN) as its electrodes.	[180]
64	Novel sulfonated polysulfone ion exchange membranes for ionic polymer metal composite actuators	A series of sulfonated polysulfone (SPSU) membranes with different degrees of sulfonation were prepared for fabricating cost effective and high performance IPMC actuators.	[181]
65	Kraton based ionic polymer metal composite (IPMC) actuator	The tip displacement, proton conductivity, current density, water uptake and ion-exchange capacity of kraton non-perfluorinated IPMCs were examined and compared with nafion based IPMCs membranes.	[63]
66	Ionic polymer actuator based on anion conducting methylated ether linked polybenzimidazole	A new type of ionic polymer actuator based on anion conducting methylated poly[(1-(4,4-diphenylether)-5-oxybenzimidazole)-benzimidazole] (PBI-OO) membranes and a carbon composite electrode material was successfully developed.	[108]
67	Low-voltage and high-performance electrothermal actuator based on multi walled carbon nanotube/ polymer composites	Environment friendly electrothermal bimetallic actuators was fabricated by multi-walled carbon nanotubes with the matrix of water borne polyurethane or silicone rubber.	[182]
68	Development of sulfonated poly(vinyl alcohol)/polypyrrole based ionic polymer	A novel sulfonated poly(vinyl alcohol)/polypyrrole polymer membrane sandwiched between platinum (SPVA-Py-Pt) was fabricated for a bending	[33]

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	metal composite (IPMC) actuator and its characterization	actuator which can be used in microrobotic applications.	
69	Study and preparation of highly water stable polyacrylonitrile-kraton-graphene composite membrane for bending actuator toward robotic application	Polyacrylonitrile-kraton-graphene (PAN-KR-GR) ionomeric polymer membrane sandwiched between Pt electrode based IPMC actuator is developed. The study opened a route to design and prepare multifunctional IPMC membrane for robotic application.	[42]
70	Fabrication of silver nano powder embedded kraton polymer actuator and its characterization	A novel silver nano powder (Ag Pw) embedded kraton (KR) ionic polymer actuator was fabricated. The KR-AgPw ionic polymer metal composite (IPMC) membrane was prepared by a solution casting method.	[29]

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