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Research paper

Investigating the effect of different anisotropic surface roughening methods on ionic polymer metal composites behavior

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Abstract

As the smart materials, ionic polymer-metal composites (IPMCs), have a layered structure consisting of a polymeric membrane based on perfluorinated alkene, which is sandwiched between two noble metal-based electrodes, such as gold and platinum, and they can be bent significantly under applying a low-range of voltage. IPMCs are used in many applications, such as robotics, biomechanics, and medical purposes. In order to improve the performance of IPMC, in this article, three different anisotropic surface roughening methods with new and optimized fabrication instructions are used, and samples are compared. The experiments are applied to measure three main factors of IPMCs: displacement, blocking force, and lifetime. The results obtained from plasma samples show that the maximum displacement is 36.23 mm, the blocking force is 4.08 etching, 18 percent higher lifetime than micro sandblasting, and sandpaper under applying a voltage range between 1-7 V; as a result, the plasma etched IPMC sample has the most efficiency.

1. Introduction

In the last few decades, scientists and researchers always have been seeking to develop and improve composite functions due to the significance of composite materials in various industrial and research subject areas [1, 2]. Indeed, investigating composites' behavior, capabilities, and structures has enabled scientists to fabricate new composites with various materials, shapes, sizes, and applications [3, 4]. One of the most notable new-built composites is polymeric, which is massively used primarily in smart materials due to its properties, including lightweightness, the capability to be produced efficiently in industrial quantities, and remarkable elastic flexibility [5, 6]. Since the '70s, generally, materials with the ability to convert mechanical force to electrical energy and vice versa have been significant; considering unique chemical and physical attributes, Electroactive Polymers (EAPs) are the types of materials that are capable of converting electric force to mechanical work directly and demonstrate a significant change in size and shape under applying voltage [7-9]. According to the operation mechanisms, EAPs can be sorted into two main classifications: 1-ionic and 2-electronic [10].

Electronic or field-activated EAPs (eEAPs) are activated by a Coulomb force that creates an electric field between two electrodes and can have displacement under applying a DC voltage. In addition, they can be operated in the air without any constraints [11]. These types of EAPs, such as piezoelectric, electrostatic, and ferroelectric, require high voltage levels (>10 V). Hence, they can be used in robotic applications with high efficiency [12]. Electronic EAPs have some advantages, for example, high mechanical energy density, large actuation forces, and rapid response. However, there are some disadvantages [13]. Although it may seem that eEAPs are suitable for use in artificial organs such as muscles because of their robustness, the need for high voltage to activate them hazardous for the human body. Indeed, the high voltages can cause blood clots or injury due to potential voltage breakdown and shorting to the body [14, 15].

Ionic metal-polymer composite (IPMC) is the most notable ionic EAP responding to applying a low voltage by showing a large displacement. IPMC consists of an electrolyte polymer membrane based on perfluorinated alkene containing short side-chains terminated with an ionic group such as SO_3^- such as Flemion®, Nafion®, Selemion®, and Aciplex® plated between two noble metals electrodes including platinum, gold, and palladium [16-18]. A common ionic polymer is Nafion® with a chemical formula of C7HF13O5S.C3F7, a perfluorosulfonated proton conductor (H^+) compounding perfluorovinyl attached to $SO_3^-H^+$ groups over a Teflon backbone. In addition, its pores provide the transport of cations but block the transport of anions. Flemion® is another choice for the fabrication of IPMCs and has a similar chemical structure to Nafion® [19, 20]. However, it is not common to use Flemion in IPMC fabrication since it cannot work in the air and is dynamically slower than Nafion®. There are four main methods to fabricate Nafion®-based IPMC: 1. Physical metal loading, 2. Hot pressing method, 3. Metallization by soaking method, and 4. Electrode coating method [21].

The first step of IPMC fabrication is the preparation of membrane surface, well known as surface treatment; in fact, the Nafion® surface should achieve suitable roughness to coat noble metal particles. Generally, there are two methods to apply roughness to the membrane: physical and chemical [22]. Chemical roughening methods such as O_2 plasma etching, well known as chemical etching, change the chemical structure of the membrane and remove oxygen atoms leading to converting SO^{-3} to SO^{-2}. Consequently, the rate of reactivity increases, and it may react with impurities in the surroundings [23]. On the other hand, physical roughening methods such as microneedles, sandpapers, Ar plasma etching (well known as physical etching), and sandblast do not change the chemical structure of the membrane and just remove some particles from the surface [24]. Applied roughness by both methods leads to creating some pattern on the surface, called surface texture, that can be isotropic and anisotropic. Isotropic surfaces have the same material properties, specifically roughness, in any direction and points of surfaces; on the contrary, in anisotropic surfaces, roughness and properties differ in various points and directions [25].

The purpose of surface roughening is to increase the surface area of the electrode. The importance of roughening process on IPMC specifications is undeniable because it improves the IPMC ion exchange rate and enhances capacity and tensile modules. In addition, it can help to increase the thickness of electrodes [26]. In this article, three IPMC samples are fabricated by three different anisotropic surface roughening methods and are compared their factors consisting of displacement, maximum deflection, and lifetime under applying a range of voltage with the experimental method.

2. Structure and function

IPMCs have a layered structure consisting of a membrane sandwiched between two electrodes. Ion transduction occurs in the membrane layer with specific properties such as ionomer nature and hydration; in fact, the membrane layer is an ion exchanger. As explained, Nafion and

Felemion are common candidates owing to the application. However, Nafion is the first choice because it has better mechanical strength and higher ion exchange capacity [27]. Nafion has poly and Perfluoroether side chains, and the chemical structure is shown in Fig. 1.

The hydrated cations move freely and migrate through the polymer network, while the anions are fixed at the polymeric chain. Electrode layers play a significant role in ion exchange (transduction), and the thickness and metal types of electrodes can affect IPMC behavior. The types of metal used in electrode layers must be unchanged in physical and chemical properties under hydration, have excellent corrosion resistance and have high conductivity, as shown in Fig. 2. Considering the properties, Platinum and Gold are mainly used for electrode layers because their suitable properties and accessibility. However, Gold is a better choice for electrodes due to its higher conductivity and flexibility and the fact that it is not hazardous to the environment [28, 29].

As an actuator, IPMCs can be bent significantly on the application of an electrical field, as shown in Fig. 3(a). The bending mechanism is caused by the transfer of ions and water molecules through the ionic channels from the anode to the cathode. When an electrical potential emerges due to the electrical field between two electrode layers in the membrane, cations are enclosed by water molecules, known as hydrated cations, and move to the cathode; therefore, the electromechanical transactions happen as shown in Fig. 3(b) [30].

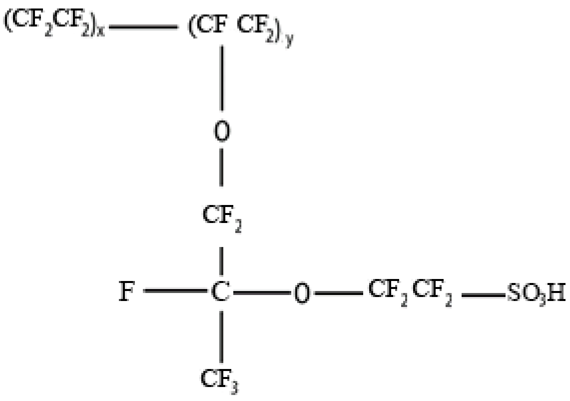

Fig. 1. Chemical structure of Nafion.

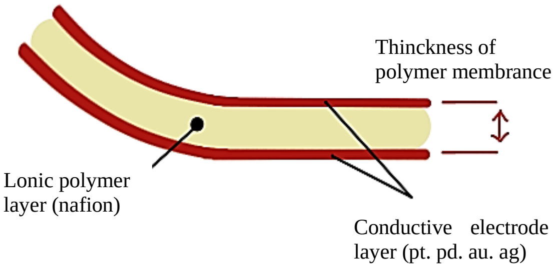

Fig. 2. The cross-section of structure of Nafion.

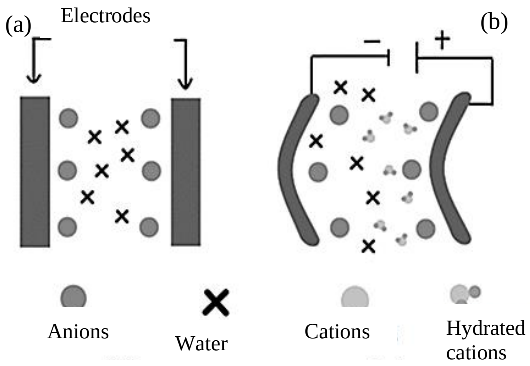

Fig. 3. (a) Schematic diagram of an IPMC actuator and (b) actuator under applied voltage.

3. Fabrication methods

In this research, Nafion 117, shown in Fig. 4, with 118 μm thickness and platinum are used to fabricate IPMC because of their accessibility and low price. As an innovation in this study, a Teflon (Polytetrafluoroethylene) beaker is used to enhance the rate of Platinum adsorption on the membrane surface and improve the solution's circulation rate. The electrode coating method is employed, which is described below:

Three different anisotropic methods are used to prepare the surface; indeed, the purpose of this step is to remove 10μm of both sides of the membrane and to rough the surface. The first method is sandblasting by micro-sand blast machine. The process speed is approximately 1 to 1.5 s/cm^2, and the bead material is glass (GP 105A, Toshiba Co. Ltd.). As the second method, Ar plasma etching is applied to treat the surface by plasma etcher (manufactured by SRBIAU), shown in Fig. 5, with RF power: 100 W and gas flow: 50 SCCM (standard cubic centimetres per minute) for 6 min etching. In the third method, sandpaper 3000 grit is utilized. After the plasma treatment process, all membranes must be put in an ultrasonic bath cleaner for at least 15 minutes at room temperature and 75% efficiency since the contaminations need to be demolished. The mechanism of the plasma can be seen in Fig. 6. The mechanism has four-level: 1. producing active gas, free radicals, and ions, 2. the transition of active gas from plasma to the surface, 3. ion bombarding the surface which creates etching, and 4. driving out the contamination from the gas chamber.

Fig. 4. Nafion 117.

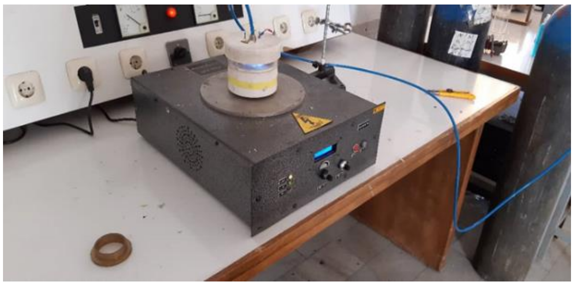

Fig. 5. Plasma etcher.

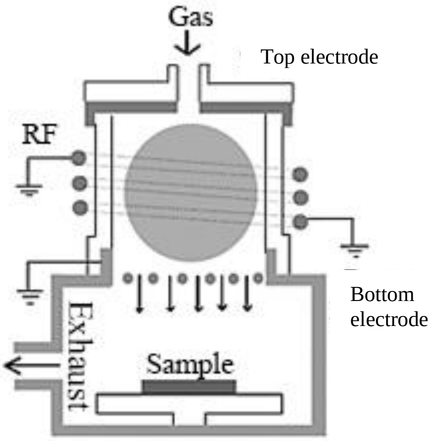

Fig. 6. Schematic representation and mechanism of plasma etching.

Adsorption: The membrane should be immersed in platinum (Pt $(NH_3)_6$)Cl_4 solution of 2 Pt/ml for at least 3 hrs until all Pt is adsorbed and ammonium hydroxide solution is added. It should be mentioned that the required amount of Pt is more than 45 ml per 30 cm^2 of Nafion surface area, as shown in Fig. 7.

Reduction: First, the Nafion membrane is immersed in string water at 40°C, and then 2 ml of sodium borohydride ($NaBH_4$) is added to the primary solution every 15 min for 14 times. Afterwards, the temperature is raised to 60°C slowly. Then, 20 ml of Pt solution is added to 100 ml stirring water for 1.5 hrs at 60°C. At the end, the membrane must be put into HCl solution for 1 hr. A black layer appears on the surface of the membrane at the final stage of the process, as shown in Fig. 8.

Developing: In the reduction step, the amount of deposited Pt on the membrane surface could be more considerable, depending on the Pt quality and thickness. In this step, it is expected that the amount of Pt will be maximum on the surface, and the membrane will have a transparent and silver cover.

Fig. 7. The sample after the adsorption process.

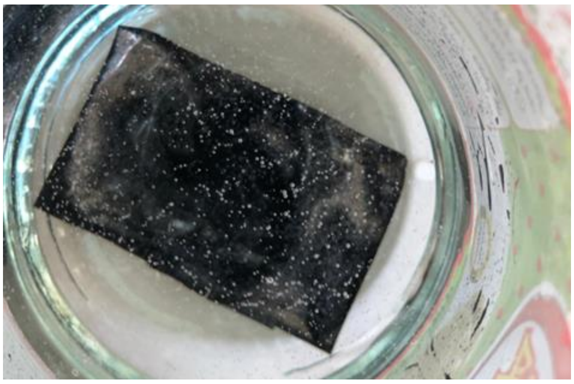

Fig. 8. The sample at the reduction step.

The 120 mg of the Pt solution is added to 120 ml distilled water, then 5 ml ammonium hydroxide is added. Temperature is raised to 40°C, and 3 ml hydroxylamine hydrochloric solution (NH_2OH-HCl) and 1.5 ml hydrazine hydrate ($NH_2NH_2.H_2O$) are added each 15 min for precisely 4 hrs. During the addition, the temperature is slowly raised to 60°C. In the next step, the solution is let to cool to room temperature. Then 5 g sodium borohydride is added to the solution helping the remaining Pt to adsorb. To remove ions, the membrane is immersed in a very dilute HCl solution stirring for 1 hr. In the final step, the IPMC must be removed from the acid solution and rinsed with distilled water. To preserve IPMC from the dry condition, which decreases flexibility, IPMC is kept in distilled water. The fabricated IPMC should have some properties, such as a shiny and homogenous surface, as shown in Fig. 9.

The IPMC needs to be tested by applying a DC voltage. It is expected that IPMC is bent significantly during applying a low voltage (1-3 V). In the present research, if the IPMC cannot deform significantly, it is commonly a problem in depositing Pt in the developing step. It is better to repeat this step until the membrane surface has a homogenous coated Pt. Finally, the IPMCs are cut to three samples described in Table 1.

4. Experimental setups

In this research, the maximum deflection of an IPMC fixed beams is analyzed by measuring the maximum rate of displacement using 1-7 V applied by a DC supply, and the length of the beams is 40 mm. The experimental setup devices for the test include a displacement laser, a DC supply, a monitor, and a controller, which are schematically shown in Fig. 10.

Fig. 9. Final fabricated IPMC.

Table 1. Specification of samples.

Sample	Code	Dimension (mm)
Plasma etching	P	40 × 10
Sandblasting	Sb	40 × 10
Sandpaper	Sp	40 × 10

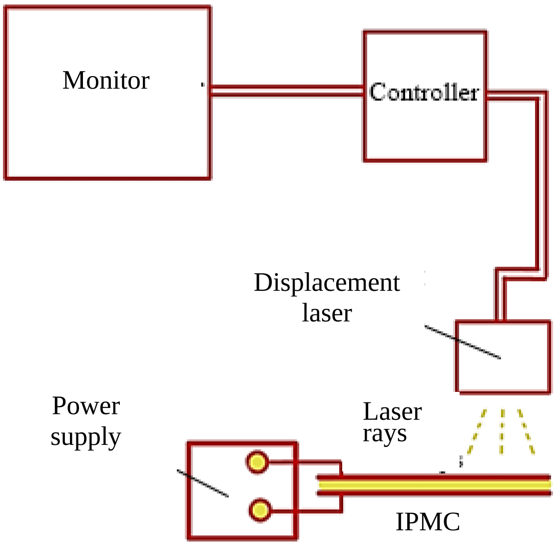

Fig. 10. Schematic diagram of a displacement sensor device.

5. Results and discussion

5.1. Displacement

Two factors impact IPMCs displacement: Young's modulus and surface resistance. More significant displacements need lower Young's modules and surface resistance. Fig. 11 compares the maximum displacements of the IPMC beams under applying a voltage range of 1-7 V.

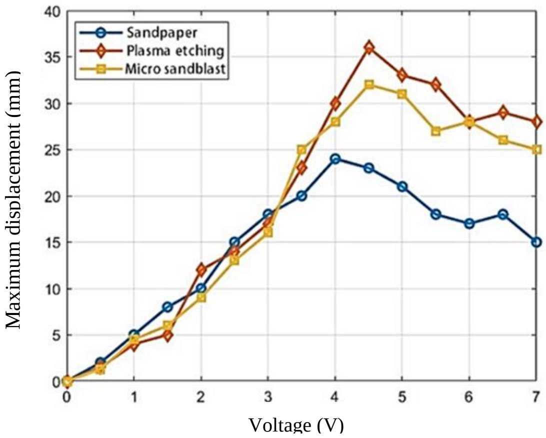

Fig. 11. Displacement chart.

According to the data, it is evident that the displacements is negligible since the resistance of the Nafion prevents the increase in displacement. By increasing the voltage rate, the displacements reache a higher value until the voltage of 4 V, and the maximum displacement of samples occurs in the range of 4-5 V.

5.2. Lifetime

For comparison, the actuator's lifetime is defined as the actuating time until the displacement of each sample reaches 15 -20% of the maximum displacements of each sample. The lifetime results are achieved in the same method as the displacement measurement, shown in Fig. 12. It is observed that the slope of the lifetime plot is related to the electrode density and thickness in the early stage.

5.3. Young modulus

The density and thickness of the electrode affect the resistance of the electrode. The applied force and the deflection are measured by a load cell and a laser displacement sensor. The cross-section of the beam is rectangular, and Young's modulus of the IPMC fixed beam is calculated from the simplified expression based on the Bernoulli-Euler beam theory as follows (Eq. 1):

$$E = \frac{4PL^3}{\delta b h^3} \tag{1}$$

where E is the Young's modulus, P is the applied force, L is the length of the beam, δ is the measured tip displacement of the actuator, b is the width, and h is the thickness. Young's modulus of the IPMC is a value in the average because the IPMC is composed of layers of polymer and metal electrodes, and the results are shown in Fig. 13.

5.4. Blocking force

The force or blocking force of the IPMC is generated by internal stress in Nafion. Blocking force is caused by the movement of water molecules in the area between two electrodes. Therefore, the force is almost inversely proportional to the resistance, as shown in Fig. 14. The data is collected and given in Table 2.

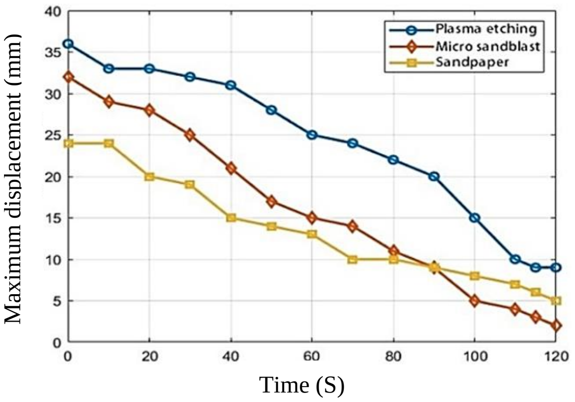

Fig. 12. Lifetime chart.

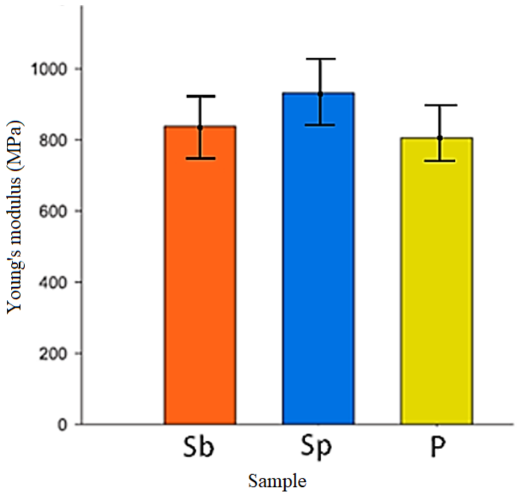

Fig. 13. Young's modulus of samples.

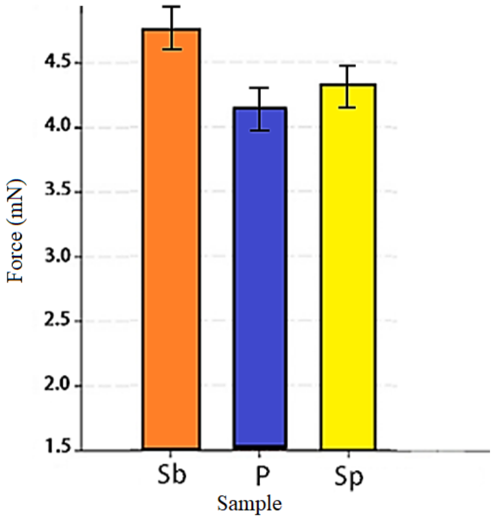

Fig. 14. Blocking forces.

Table 2. The polynomial coefficient of samples.

Sample	Code	Blocking force (mN)	Maximum displacement (mm)
Plasma	P	4.21	36.23
Sandblast	Sb	4.39	32.39
Sandpape	Sp	4.7	24.71

5.5. Maximum deflection equation

Due to IPMC fundamentals, the deflection of an IPMC beam is related to the stiffness and applying voltage. According to the membrane's internal resistant, the deflection rate is not high when applying voltage is poor. Increasing the voltage enhances the ion transfer; hence, the bending rate will be high.

The IPMC beam reaches maximum bending rate when the electric field in the membrane is balanced by applying voltage and ion distribution. In this research, it is now possible to extract an equation describing the maximum deflection of a linear equation of an ionic polymer metal composite-based fixed beam calculated by Eq. (2). In addition, the result of each sample is shown in Table 3.

$$f(x) = p_1.x^4 + p_2.x^3 + p_3.x^2 + p_4.x + p_5 \quad (2)$$

6. Conclusions

The surface area of the membrane is one of the essential elements in depositing electrodes, and by increasing the surface area with surface roughening methods, the electrode is deposited deeply and ununiformly, leading to improve IPMC performance. The results show that plasma etching is the best anisotropic process for surface roughening compared to the other methods, because the experiments demonstrate that the highest maximum displacement value is 48.21% higher than sandpaper method and 13.41% higher than microsandblasting method in the same voltage range. In addition, the slope of the displacement chart has the lowest fluctuation; and it has the longest lifetime when 4.5 V is applied.

Table 3. The polynomial coefficient of samples.

Sample/Quantity	P	SB	SP
p_1	0.08685	0.065	0.06782
p_2	-1.325	-1.281	-1.292
p_3	5.437	6.982	6.863
p_4	-0.7973	-4.805	-4.77
p_5	0.6279	1.27	1.306

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