Supporting information

A Molecular-Regulation Strategy towards Low-voltage Driven, Multi Degree of Freedom IPMC Catheters

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Experimental Section

Materials: Hydrogen peroxide (H\textsubscript{2}O\textsubscript{2}), Sodium sulfite (Na\textsubscript{2}SO\textsubscript{3}), sodium carbonate (Na\textsubscript{2}CO\textsubscript{3}) and sulfuric acid (H\textsubscript{2}SO\textsubscript{4}) were purchased from Sinopharm Chemical Reagent Co., Ltd. Chloroauric acid (HAuCl\textsubscript{4}) was purchased from Shanghai Chemical Reagent Co., Ltd. 1,10-Phenanthroline monohydrate (Phen) was purchased from Sigma-Aldrich. Methanol, ethanol, propanol, acetone, ethyl acetate and diethyl ether were purchased from China National Medicines Co., LTD. Nafion tube (diameter of 3 mm) was purchased from the HALMA company. Deionized water was homemade.

Synthesis of [Au(Phen)Cl\textsubscript{2}]Cl solution: Firstly, Na\textsubscript{2}CO\textsubscript{3} powder (10 mg) was mixed with HAuCl\textsubscript{4} solution (8 mL, 10 g L\textsuperscript{-1}). Then, the mixture was heated to 50 \textdegree C under stirring state for 1h to obtain NaAuCl\textsubscript{4} solution. After that, Phen solution (12 mL, 5 g L\textsuperscript{-1}) was added dropwise into the NaAuCl\textsubscript{4} solution at 95 \textdegree C under stirring state for 30 mins. Finally, the [Au(Phen)Cl\textsubscript{2}]Cl solution was obtained after cooling down to room temperature.

Fabrication of IPMC catheter: Au electrode was plated on Nafion tube by an
chemical reduction procedure. Before catheter fabrication, we anneal the Nafion tube at 140 °C for 2h to release internal stress, and the curved tube becomes straight one. Firstly, Nafion tube was etched by plasma equipment to obtain a rough surface. Secondly, hydrogen peroxide (5% mass concentration) and sulfuric acid solution (1 mol L\(^{-1}\)) were used to rinse the Nafion tube for removing residual organic impurities. Then, the cleaned tube was immersed into [Au(Phen)Cl\(_2\)]Cl solution for 18 h for ionic exchange process. After that, faint yellow Nafion tube was put into deionized water heating to 90 °C. Na\(_2\)SO\(_3\) solution (3% mass concentration) was added dropwise to the solution. The reduction continued for 5h until Nafion tube was plated with Au electrode. This reduction procedure was repeated several times to obtain high-quality Au electrode layer. Finally, IPMC catheter with four individual electrodes were obtained through laser cutting technique.

**Regulation process of Nafion materials:** Firstly, Nafion tube was etched by plasma and then rinsed by hydrogen peroxide and sulfuric acid solution. Then, the cleaned Nafion tube was immersed into the alcoholic solution (10 mL, 1.3 mol L\(^{-1}\)) under water bath of 55 °C for 30 mins. After that, the tube was washed by deionized water. Finally, the alkanol molecules regulated Nafion tube was obtained after drying for 24h at room temperature.

**Material Characterizations:** SEM and TEM images were measured by Hitachi S-4800 and FEI Tecnai G2 F20 equipment, respectively. X-ray powder diffraction (XRD) analysis was conducted by Philips X’Pert PRO diffractometer with nickel-filtered Cu K\(_\alpha\) radiation. Stess-strain measurements were conducted by universal tester (Shimadzu, AGS-X 500N) in tension mode. The electrode conductivity was tested by a multifunction digital four-probe tester (ST-2258C). Actuation displacements were recorded by laser locator (Keyence, LK-G80). Testing cell for actuation measurements showed in Figure 2g was customized from Dongguan Ying An Plastic Mould Co., Ltd.

**Electrochemical Measurements:** Electrochemical properties of IPMC catheters were tested by electrochemical work station (Shanghai Chenghua, CHI760D). Specific capacitance (C, F g\(^{-1}\)) of the device was calculated by using the following equation.\(^1\)
\[
C = \frac{S_{\text{Area}}}{2v \cdot \Delta V \cdot m}
\]

Where \( S_{\text{Area}} = \oint I \, dV \) is the loop area of CV curve, \( v \) is the scan rate, \( \Delta V \) is the potential window, \( m \) is the mass of Au electrode.

Ionic conductivity \((\sigma, \text{ S cm}^{-1})\) of the device is calculated from the impedance spectroscopy according to previous works below:\(^2\)

\[
\sigma = \frac{D}{(R_b \cdot S)}
\]

Where \( D \) is the thickness of Nafion tube, \( S \) is the interface contact area, \( R_b \) is the equivalent series resistance.

**Additional Figures and Results**

Fig. S1 Surface and cross-sectional SEM images of the electrode incision caused by laser micro-dissection.

Fig. S2 Actuation displacement as a function of frequency of the catheter.
Fig. S3 a) DSC traces and b) XRD patterns of Nafion tubes regulated by diethyl ether, ethyl acetate and acetone, respectively.

Fig. S4 a) Young’s Modulus of Nafion tubes regulated by methanol, ethanol and propanol, respectively. b) Young’s Modulus of Nafion tubes regulated by diethyl ether, ethyl acetate and acetone, respectively.

Fig. S5 a) CV curves of IPMC catheters regulated by methanol, ethanol and propanol, respectively. b) Capacitance and ionic conductivity of the catheters regulated by diethyl ether, ethyl acetate and acetone, respectively.
Fig. S6 Actuation displacement as a function of frequency of catheters regulated by diethyl ether, ethyl acetate and acetone, respectively.

Fig. S7 Stress-strain curve of Nafion treated with pure water under the same condition.

Table S1 Values of crystallinity for Nafion and modified Nafion. (Calculated through method reported in previous works)³⁻⁵

<table>
<thead>
<tr>
<th>Sample</th>
<th>Nafion</th>
<th>MeOH</th>
<th>EtOH</th>
<th>PrOH</th>
<th>Ethyl acetate</th>
<th>Acetone</th>
<th>Diethyl Ether</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystallinity (%)</td>
<td>25.13</td>
<td>20.84</td>
<td>18.72</td>
<td>23.25</td>
<td>23.67</td>
<td>24.43</td>
<td>24.68</td>
</tr>
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References