Application of the multi-walled carbon nanotube and sodium alginate based sol-gel coatings to enhance the electrically actuated performance of muscle-like biomass hydrogel paper actuators

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Research Article

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Abstract

To comply with green chemistry and achieve sustainable development, an electrically actuated membrane and two non-metallic electrode membranes are assembled into a muscle-like Biomass Hydrogel Paper Actuator (BHPA) similar to ‘sandwich’ structure, which has a great potential in engineering application due to the excellent properties such as light weight, low driving voltage and good flexibility. And the contact resistance between every two membranes of the BHPA is the key factor to improve its output force characteristics and tremor behavior. Thus, in this paper, based on the orthogonal experiment and control variates, under the excitation of electric field, the effect and mechanism of Multi-walled Carbon Nanotube (MWCNT) - Sodium Alginate (SA) based sol-gel coating and its thickness on electrically actuated performances of the BHPA were studied in depth and the results were compared with that of the BHPA samples assembled by the traditional hot laminating technology. Furthermore, with equivalent circuit model, the preliminary quantitative relationship between peak current and contact resistance of the BHPA was derived by a mathematical expression. The results demonstrated that the optimum dimension and testing voltage of the BHPA were 35 mm × 8 mm × 1 layer (0.352 mm) and 4 V, where its output force density and service life both reached the maximum values of 13.34 mN/g and 330 s with the lightest tremor behavior. Moreover, the internal resistance and elastic modulus of the BHPA achieved the minimum values of 2.13 Ω and 3.1 MPa respectively, and its specific capacitance acquired the maximum value of 81.3 mF/g under the sol-gel coating of 1 coating thickness. Generally, it is of great value to modify interface properties of the BHPA for actuation enhancement in demanding working conditions.

Keywords

Biomass hydrogel paper actuator; Electrically actuated performance; Orthogonal experiment; Multi-walled carbon nanotube; Sodium alginate; Sol-gel coating thickness.

Declarations

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Conflicts of interest/Competing interests

The authors have no conflicts of interest to declare that are relevant to the content of this article.

Availability of data and material

Not applicable.

Code availability
Not applicable.

Authors’ contributions
All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Junjie Yang, Tao Yu, Jintong Yao, Siyong Wang and Kang Wei. The first draft of the manuscript was written by Junjie Yang and Tao Yu, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.
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ABSTRACT

To comply with green chemistry and achieve sustainable development, an electrically actuated membrane and two non-metallic electrode membranes are assembled into a muscle-like Biomass Hydrogel Paper Actuator (BHPA) similar to ‘sandwich’ structure, which has a great potential in engineering application due to the excellent properties such as light weight, low driving voltage and good flexibility. And the contact resistance between every two membranes of the BHPA is the key factor to improve its output force characteristics and tremor behavior. Thus, in this paper, based on the orthogonal experiment and control variates, under the excitation of electric field, the effect and mechanism of Multi-walled Carbon Nanotube (MWCNT) - Sodium Alginate (SA) based sol-gel coating and its thickness on electrically actuated performances of the BHPA were studied in depth and the results were compared with that of the BHPA samples assembled by the traditional hot laminating technology. Furthermore, with equivalent circuit model, the preliminary quantitative relationship between peak current and contact resistance of the BHPA was derived by a mathematical expression. The results demonstrated that the optimum dimension and testing voltage of the BHPA were 35 mm × 8 mm × 1 layer (0.352 mm) and 4 V, where its output force density and service life both reached the maximum values of 13.34 mN/g and 330 s with the lightest tremor behavior. Moreover, the internal resistance and elastic modulus of the BHPA achieved the minimum values of 2.13 Ω and 3.1 MPa respectively, and its specific capacitance acquired the maximum value of 81.3 mF/g under the sol-gel coating of 1 coating thickness. Generally, it is of great value to modify interface properties of the BHPA for actuation enhancement in demanding working conditions.

Keywords: Biomass hydrogel paper actuator; Electrically actuated performance; Orthogonal experiment; Multi-walled carbon nanotube; Sodium alginate; Sol-gel coating thickness.

(Some figures may appear in colour only in the online journal)

1. Introduction

Since the 21st century, with the deepening of robot development and the birth of new smart materials, a muscle-like Biomass Hydrogel Paper Actuator (BHPA), as the key driver of intelligent mechatronics system, is now setting off a global research upsurge and has become a focus in the fields of soft robot (Yufeng et al. 2020; Park et al. 2020), nanocomposite hydrogel (Cunfeng et al. 2021; Qingli et al. 2020; Haraguchi et al. 2002), piezoelectric actuator (Liang et al. 2022; Bar-Cohen et al. 2006) and biomedical engineering etc (Gu et al. 2020; Foroughi et al. 2011). Due to the limitation of materials, Electro Active Polymer (EAP) paper actuators have been in a very slow stage of development until the emergence of new material such as the biomass hydrogel (Junjie et al. 2020, 2022; Zhe et al. 2021), which has the characteristics of high strain, good flexibility, light weight and no noise. Thus the BHPA recognized as the most suitable paper actuator, has many similar characteristics with muscle, and even surpasses muscle in some actuation aspects.

As an EAP paper actuator, the BHPA can produce bending deflection and output force under the excitation of electric field, so that the electric energy is capable of transforming into the mechanical energy directly and isothermally. However, the BHPA always has the problem of weak and unstable output response, which seriously
hinders its practical application and development. Considering this issue, researchers at home and abroad have carried out a large number of relevant studies (Sun et al. 2018, 2020; Junjie et al. 2021, 2022; Zhu et al. 2021; Chendong et al. 2022). For instance, Cho and Ha et al. (Cho et al. 2022; Ha et al. 2020) have provided an engineered fabrication platform of multi-responsive actuator by combining hydrogel lithography and electrospinning methods, which gives an insight into the design of soft actuator with stimulus specific, multi scale and functional properties. Shin et al. (Shin et al. 2021) has designed electrically responsive poly(sodium 4-vinylbenzenesulfonate / 2-hydroxyethyl methacrylate / acrylic acid) and poly(sodium 4-vinylbenzenesulfonate / 2-hydroxyethylmethacrylate / acrylamide) hydrogels, and their electrically responsive bending actuation can be adjusted by ion strength, cross-linking density, applied voltage, geometrical parameters and monomer composition. Kim et al. (Kim et al. 2020; Duan et al. 2020) has fabricated cellulose nanocrystal-reinforced polyvinyl alcohol-cellulose physical hydrogels utilizing a simple blending technology for actuator application, and its immense output displacement was observed at low electrolyte concentration. Yang et al. (Yang et al. 2021) has exhibited the studies and syntheses of mechanical properties for series novel inorganic-organic hydrogels via using vinyl functionalized silicon nanoparticles as the cross-linker, where the quick response in swelling and deswelling behaviors are owing to the improved rigidity of hydrogel network and the modified surface water diffusion on nanoparticles, respectively. Collectively, these studies demonstrated that among various paper actuators, hydrogel actuators and soft actuators had drawn much attention because of their smart low electric field control, simple structure and light specific gravity, whereas the covered performances were not enough due to their limited electrical actuation mechanism of conventional electrochemical reaction.

Therefore, in this work, for the remaining challenge of assembling the electrically actuated membrane and two non-metallic electrode membranes into the muscle-like BHPA owing to its non-adhesive and slippery nature, and from the perspective of the BHPA under different assembly process factors, a reliable and facile approach of the Multi-walled Carbon Nanotube (MWCNT) - Sodium Alginate (SA) based sol-gel coating was proposed in achieving a good membrane bonding of the BHPA with improved interface contact resistance and enhanced electrically actuated performance. Furthermore, under the excitation of electric field and based on the orthogonal experiment and control variates, effect and mechanism of the MWCNT-SA based sol-gel coating and its thickness on electrically actuated performances of the BHPA were researched through the testing platform and compared with that of the BHPA samples assembled by the traditional hot laminating technology. And then an equivalent circuit model of the qualitative relationship between contact resistance and peak current of the BHPA was derived by a mathematical formula, which was characterized by Electrochemical Impedance Spectroscopy (EIS) and Cyclic Voltammetry (CV), tensile property, diffraction of x-rays (XRD) pattern and infrared spectroscopy (IR) methods; additionally, the surface and cross-sectional microstructure of the BHPA prepared by the natural polymer SA were captured and analyzed through the electron spectroscopy and cold field-emission scanning electron microscope (SEM) images.

2. Experiments and tests
2.1. Preparation technique

The essential materials were the SA (analytical reagent), the MWCNT aqueous dispersion of 10.0 wt%, glycerol (chemically pure) and distilled water. And the experimental instruments included an electronic analytical balance (brand of LICHEN, FA324C, Zhejiang LICHEN Instrument Technology Co., Ltd), magnetic stirrers (brand of LICHEN, SN-MS-H280D, Zhejiang LICHEN Instrument Technology Co., Ltd) and a vacuum drying box (brand of LICHEN, DZF-6050A, Zhejiang LICHEN Instrument Technology Co., Ltd). The preparation process of the BHPA samples was shown in Fig. 1. Concretely, similar to a ‘sandwich’ structure, the BHPA consisted of two non-metallic electrode membranes and an electrically actuated membrane caught in the middle. To prepare the electrode membrane, the MWCNT aqueous dispersion of 10 mL was injected into 40 mL SA solution in a beaker to obtain the electrode solution, and then it was heated with 60 °C water bath and was stirred at the speed of 800 r/min with 1 mL glycerol dripping for 25 min. Subsequently, the electrode solution was put into a vacuum drying box to obtain the electrode membrane by vacuum drying with the vacuum degree of -0.080 MPa and the drying temperature of 60 °C for 36 h. Next, to prepare the electrically actuated membrane, the SA of 1.5 g was dissolved in 50 mL distilled water in a water-bath heating to 50 °C and then was mixed with 3 mL glycerol stirring at 600 r/min for 45 min. Afterwards, the electrically actuated solution was evenly coated on the electrode membrane inside the vacuum drying box. After completely drying, another part of the electrode solution was coated on the electrically actuated membrane in the same way. By repeating the above steps, the BHPA samples as the experimental group were acquired with the MWCNT-SA based sol-gel coating of 1, 2 and 3 coating thicknesses respectively, and some other samples had also been obtained by the traditional hot laminating technology as the control group.

![Fig. 1. Preparation process of the BHPA samples.](image)

2.2. Test methods

The testing platform of the BHPA was presented in Fig. 2, where the electronic analytical balance with high precision of mN level was used to measure its output force characteristics. The BHPA sample was clamped on the lead screw elevator with a clamp wrapped in copper sheets. Its vertical displacement was controlled by adjusting the knob, and the operation was stopped when the sample was just in contact with the scale pan of the electronic analytical balance that was connected with a computer through serial port line 232 and data acquisition software. Under the excitation of electric field provided by a DC regulated power supply (brand of RIGOL, DP813A, RIGOL Technologies, Co. Ltd), the BHPA sample would produce output force with a testing period unified as 2400 s and was recorded every second, and the peak current of the sample was measured in real time by a digital multimeter (brand of DELIXI ELECTRIC, DE22, DELIXI Electric, Co. Ltd), which was fitted
and analyzed by a software of OriginPro 9.1.

3. Results and discussion

3.1. Orthogonal experimental design and analysis

Orthogonal experiment was suitable for the multi-factor and multi-level experimental testing, and had characteristics of high efficiency, fast and economy. Then based on the principle of orthogonality, the most representative combination samples with the advantage of “uniform disperstiveness and regular comparability”, were selected to achieve the research results equivalent to that of comprehensive tests, and the conclusions obtained were clear and reliable. As shown in Fig. 3, four influencing factors of the BHPA were length (A), width (B), thickness (C) and testing voltage (D), respectively, where each factor had three testing levels. With constant thickness of the electrode membrane, thickness of the BHPA changed with its electrically actuated membrane, and was concisely expressed by its number of layers.

Accordingly, the time when violent output-force tremor occurred for the first time was defined as service life of the BHPA, which was adopted as evaluation index of the orthogonal experiment. And its influence degree of each factor and level on the experiment results was obtained by calculating and analyzing the sum of service life $k_i (s)$, the average service life $\bar{k}_i (s)$ and its Range ($R$) of various BHPA samples under the same influencing factors.
factor, which were calculated by following formulas and their results were listed in Table 1, respectively.

\[ K_i = \sum_{1}^{n} T_i, \quad (1) \]

where \( K_i \) was the sum of the BHPA service life of level \( i \) under various process influencing factors; \( T_i \) was its service life; \( n \) and \( i \) were separately the number and serial number of level \( i \) under each influencing factor.

\[ k_i = K_i / n, \quad (2) \]

where \( k_i \) was the average BHPA service life of level \( i \) under various process influencing factors.

\[ R = \max [k_i] - \min [k_i], \quad (3) \]

where \( R \) was the range of service life of the BHPA under various process influencing factors.

Table 1 Results and analysis of the BHPA samples based on orthogonal experiment.

<table>
<thead>
<tr>
<th>Experiment serial number</th>
<th>Influencing factor</th>
<th>Service life (( T ), unit: s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
</tr>
<tr>
<td>01</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>02</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>03</td>
<td>1</td>
<td>3</td>
</tr>
<tr>
<td>04</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>05</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>06</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>07</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>08</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>09</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>( K_1 )</td>
<td>600</td>
<td>575</td>
</tr>
<tr>
<td>( K_2 )</td>
<td>635</td>
<td>585</td>
</tr>
<tr>
<td>( K_3 )</td>
<td>575</td>
<td>620</td>
</tr>
<tr>
<td>( k_1 )</td>
<td>200</td>
<td>191.67</td>
</tr>
<tr>
<td>( k_2 )</td>
<td>211.67</td>
<td>195</td>
</tr>
<tr>
<td>( k_3 )</td>
<td>191.67</td>
<td>206.67</td>
</tr>
<tr>
<td>Range (( R ))</td>
<td>20</td>
<td>15</td>
</tr>
</tbody>
</table>

\[ \Sigma T_i = 1795 \]

It could be seen that the longest service life of the BHPA sample (No. 06) was 305 s during 9 groups of orthogonal experiments, and among the average service life \( k_i \) of various BHPA samples under the same influencing factor A, the level 2 reached the maximum value of 211.67 s, which meant that the best level was A2.

Similarly, under different influencing factors of B, C and D, the average service life \( k_i \) of various BHPA samples reached the maximum values of 206.67 s, 231.67 s and 288.33 s, respectively, thus the relevant best level was B3, C1 and D2, and then the optimal combination of each factor and level on the BHPA was A2B3C1D2. Besides, under the four influencing factors, Range (\( R \)) of average service life \( k_i \) of the BHPA with different levels was 20, 15, 63.34 and 195 respectively, thus the priority of influencing factors was D > C > A > B. To sum up, the
optimum dimension and testing voltage of the BHPA were 35 mm × 8 mm × 1 layer (0.352 mm) and 4 V. It was capable of being verified by output force characteristics and tremor behavior of the BHPA, as shown in Fig. 4, where its sample (No. 06) was the optimum with a relatively stable output force, the slowest frequency and the smallest amplitude of output-force tremor behavior, and the consequently longest service life.

Fig. 4. Based on orthogonal experiments, output force characteristics and tremor behavior of the BHPA with 9 groups of samples: (a) No. 01, (b) No. 02, (c) No. 03, (d) No. 04, (e) No. 05, (f) No. 06, (g) No. 07, (h) No. 08 and (i) No. 09.

Moreover, Fig. 5 presented the output force characteristics and service life of BHPA samples under different influencing factors of testing voltage, length, width and thickness. Concretely, under the testing voltage of 4 V, stable output force of the BHPA was the largest with the longest service life, and it firstly increased and then decreased with the rise of length, which was because the short BHPA sample was not conducive to be clamped and its stiffness was strong with poor output force characteristics and a large tremor behavior; conversely, the long BHPA sample, as a polymer material, had viscoelasticity and a relatively small stiffness, thus suitable length of the BHPA was 35 mm. Besides, its service life continued to ascend gradually with the width rising, because it could appropriately adjust stiffness of the BHPA with a constant length. When aspect ratio of the BHPA was about 4.3, its stable output force was the largest and was not easy to produce the output-force tremor behavior, thus the optimum width of the BHPA was 8 mm. Since its much thickness was capable to raise stiffness and interior stress of the BHPA, it would cause the violent tremor behavior and the short service life; on the contrary, too thin of the BHPA was difficult to provide sufficient output force actually required, where its proper thickness was 1 layer (0.352 mm).
Fig. 5. Output force characteristics and service life of the BHPA samples based on different influencing factors: (a) testing voltage, (b) length and width, and (c) thickness and testing voltage.

3.2. Output force characteristics and tremor behavior of the BHPA

According to control variates, under the excitation of electric field, the effect of MWCNT-SA based sol-gel coating and its thickness on output force characteristics and tremor behavior of the BHPA was studied in depth and the results were compared with that of the BHPA samples assembled by the traditional hot laminating technology. The variation curves of output force characteristics and tremor behavior of the BHPA samples with time were shown in Fig. 6. It could be seen that the electrically actuated performance of every BHPA sample was various under different assembly processes and the sol-gel coating thicknesses. But from the whole test time range, the complete working process of each sample was roughly the same, which could be divided into three stages; rapid rise, stable work and fatigue decline of the output force. Among them, for the sample assembled by the traditional hot laminating technology, it did not appear the fatigue decline stage of output force but with a poor tremor behavior; in comparison, ignoring the sol-gel coating thickness, all the BHPA samples with MWCNT-SA based sol-gel coatings presented excellent output force characteristics and the improved tremor behavior.
Fig. 6. Output force characteristics and tremor behavior of the BHPA samples based on various assembly process factors: (a) the traditional hot laminating technology, (b) the MWCNT-SA based sol-gel coating of 1 coating thickness, (c) that of 2 coating thicknesses and (d) that of 3 coating thicknesses.

Specifically, as shown in Fig. 6(a), assembled by the traditional hot laminating technology, the stable value of output force of the BHPA was the minimum (1.347 mN) of all samples, and it was always accompanied by the output-force tremor throughout the test range, which was defined as the permanent tremor behavior without effective means of regulation and inhibition. In contrast, as shown in Fig. 6(b), adopting the MWCNT-SA based sol-gel coating of 1 coating thickness, total thickness of the BHPA sample was about 0.538 mm, and its stable output force (3.501 mN) and service life had been immensely enhanced, which were all around triple that before it adopted; meanwhile, the output-force tremor had also been greatly improved, which became the persistent tremor behavior with a duration of ≥ 600s. Furthermore, Fig. 6(c) and Fig. 6(d) presented the significantly promoted stable values of output force (3.549 mN and 6.551 mN) of the BHPA samples assembled with the sol-gel coating of 2 coating thicknesses about 0.773 mm and that of 3 coating thicknesses around 1.08 mm respectively; however, the output-force tremor and service life had been fairly damaged instead of being continuously modified, and there were extremely irregular output-force tremor behavior of paroxysmal tremor (≤ 60 s) and persistent tremor (Junjie et al. 2021). In addition, under different assembly processes and the MWCNT-SA based sol-gel coating thicknesses, the evaluation parameters of output force characteristics and tremor behavior of the BHPA samples were listed in Table 2.

<table>
<thead>
<tr>
<th>Assembly process factors</th>
<th>Traditional hot laminating technology</th>
<th>MWCNT-SA based sol-gel coatings</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mass of sample (g)</td>
<td>0.2131</td>
</tr>
<tr>
<td></td>
<td>Stable value of output force (mN)</td>
<td>1.347</td>
</tr>
<tr>
<td></td>
<td>Output force density (mN/g)</td>
<td>6.32</td>
</tr>
<tr>
<td></td>
<td>Service life (s)</td>
<td>84</td>
</tr>
<tr>
<td></td>
<td>Duration of tremor behavior (s)</td>
<td>2316</td>
</tr>
<tr>
<td>Duty ratio of tremor behavior</td>
<td>96.5%</td>
<td>86.25%</td>
</tr>
<tr>
<td>-------------------------------</td>
<td>-------</td>
<td>--------</td>
</tr>
<tr>
<td>Number of tremors</td>
<td>181</td>
<td>193</td>
</tr>
<tr>
<td>Frequency of tremor (per minute)</td>
<td>4.69</td>
<td>5.59</td>
</tr>
<tr>
<td>Amplitude of tremor behavior</td>
<td>15.06%</td>
<td>17.91%</td>
</tr>
</tbody>
</table>

Correspondingly, to intuitively analyze the electrically actuated performance of BHPA samples based on different assembly process factors, the trend curves were drawn by selecting the representative parameters listed, as shown in Fig. 7. Thereinto, Fig. 7(a) presented that the greatly enhanced service life of the BHPA sample decreased rapidly with the increase of the MWCNT-SA based sol-gel coating thickness, and the service life reached the largest value of 330s under the sol-gel coating of 1 coating thickness, which was about four times of the shortest value 84s of the sample assembled by the traditional hot laminating technology. Conversely, the largely reduced duty ratio of tremor behavior increased sharply with the sol-gel coating thickness rising, but the duty ratio achieved the minimum value of 86.25% under the sol-gel coating of 1 coating thickness, which was decreased by 10.62% of the maximum value 96.5% of that sample adopting the traditional technology. Although the service life and duty ratio had completely opposite trends, their main turning points were the same. Fig. 7(b) showed that under the traditional technology, the frequency of output-force tremor of the BHPA sample was the lowest value of 4.69 times per minute, and it increased promptly to the highest value 6.45 times per minute of the sample under the sol-gel coating of 3 coating thicknesses. On the other hand, the amplitude of tremor behavior first increased and then decreased; subsequently, it obtained the maximum value of 17.91% under the sol-gel coating of 1 coating thickness, and the amplitude of tremor behavior gained the minimum value 15.06% of the sample assembled by the traditional technology. In summary, these evaluation parameters listed were capable of well reflecting electrically actuated performance of the BHPA from different angles. Under the MWCNT-SA based sol-gel coating, the contact resistance between membranes of the electrode membrane and electrically actuated membrane was effectively reduced inside the BHPA, so its stable output force and service life grew markedly with the dwindling duty ratio. Whereas, with the increase of the sol-gel coating thickness, elastic modulus of the BHPA sample amplified gradually, which resulted in the boosting frequency and abating amplitude of the output-force tremor behavior. Thus, under the sol-gel coating of 1 coating thickness, it could significantly improve output-force tremor behavior of the BHPA, to ensure working stability of the BHPA and make it have practical application value.

![Fig. 7. Variation law of representative performance evaluation parameters of the BHPA samples on the output process factors.](image)
force characteristics and the tremor behavior: (a) service life and duty ratio, and (b) frequency and amplitude.

3.3. Electrochemical properties of the BHPA

For acquiring the effect of different assembly process factors on electrochemical properties of the BHPA, its internal resistance and specific capacitance were compared by the EIS and CV methods respectively, as shown in Fig. 8, thus the specific capacitance \( C_s \) (mF/g) and internal resistance \( R_\Omega \) (\( \Omega \)) were obtained by the formula (4) and formula (5). Specifically, Fig. 8(a) presented that Nyquist diagram of the BHPA under the EIS signal was a good linear relationship between the real part resistance and the imaginary part capacitive reactance of the cross-linking impedance signal, where the slope of the graphed line was internal resistance of the BHPA sample. Fig. 8(b) showed that the samples had typical electrical double-layer capacitance of electrochemical interface, which possessed the charge-discharge characteristics at a constant rate, and the similar electrochemical behaviors with excellent capacitance properties.

\[
C_s = \frac{C}{m} = \int_{U_1}^{U_2} i \cdot dU \left( m \cdot u \cdot \Delta U \right), \quad (4)
\]

where \( C \) (mF), \( m \) (g), \( i \) (A) and \( U \) (V) separately represented capacitance, mass, electron current of the BHPA and the excitation intensity of electric field; \( u \) (V/s), \( \Delta U \) (V), \( U_1 \) (V) and \( U_2 \) (V) were the scanning speed, potential window, starting and terminating voltages of the CV experiment, respectively.

Meantime, in the RC equivalent circuit of the BHPA electrochemical system, its total impedance was equal to the sum of impedance of each series element. Since there was no inductive element such as coil, the inductive reactance \( R_L \) (\( \Omega \)) was 0. Besides, under a high frequency of sinusoidal alternating current, the capacitive reactance \( R_C \) (\( \Omega \)) tended to 0, and then the total impedance was only electrical impedance \( R_\Omega \) (\( \Omega \)), i.e. internal resistance of the BHPA, which was tested by the following formula.

\[
Z = Z' + jZ'' = R_\Omega + j(R_L - R_C) = R_\Omega - j(1/2\pi \cdot f \cdot C), \quad (5)
\]

where \( Z \) (\( \Omega \)), \( Z' \) (\( \Omega \)) and \( Z'' \) (\( \Omega \)) represented the total impedance, real impedance and imaginary impedance of a series circuit respectively; similarly, \( R_\Omega \) (\( \Omega \)), \( R_L \) (\( \Omega \)) and \( R_C \) (\( \Omega \)) was its real electrical impedance (i.e. internal resistance), imaginary inductive reactance and imaginary capacitive reactance; \( C \) (F) was the capacitance and \( f \) (Hz) was the frequency of sinusoidal alternating current.

As shown in Fig. 8(c), specific capacitance of the BHPA decreased violently with the increase of the MWCNT-SA based sol-gel coating thickness, but changed gently under the traditional hot laminating technology. Among them, specific capacitance of the BHPA reached the maximum value of 81.3 mF/g and its internal resistance achieved the minimum value of 2.13 \( \Omega \) under the sol-gel coating of 1 coating thickness, which was respectively around five times of 17 mF/g and was decreased by 12.7% of 2.44 \( \Omega \) of the sample assembled by the traditional hot laminating technology. In comparison, under the MWCNT-SA based sol-gel coating, the contact resistance between membranes of the electrode membrane and electrically actuated membrane was greatly abated in the BHPA, which improved its internal resistance and specific capacitance with the growing accumulation and migration rate of charged ions, and played a vital role in enhancing its electrically actuated performance.
Fig. 8. Electrochemical properties of the BHPA samples based on various assembly process factors: (a) Nyquist diagram of the EIS signal; (b) scanning potential and current, and (c) specific capacitance and internal resistance.

3.4. Tensile property of the BHPA

Under the excitation of electric field, the bending deflection angle and output force characteristic of the BHPA basically occurred in its elastic deformation stage. And its tensile property was capable of being expressed by the elastic modulus ($E$, unit: MPa) based on an electronic universal material testing machine (CMT5504, Shenzhen Sansi Test Instrument Co., Ltd) and a software of OriginPro 9.1, which was calculated as follows:

$$E = \frac{\sigma}{\delta}, \quad (6)$$

where $\sigma$ (MPa) represented tensile stress of the BHPA and $\delta$ (%) was its tensile strain.

As shown in Fig. 9(a), even under different assembly process factors, the BHPA samples did only have elastic deformation stage, but no obvious yield and plastic hardening stages. Through size measurement, it was found that the size of the samples after fracture were consistent with their overall size before the tensile test, which indicated that the BHPA was conformed to the typical mechanical properties of hydrogel materials. And they possessed soft and weak textures commonly with low tensile strength and small tensile strain. Fig. 9(b) presented that with the rise of the MWCNT-SA based sol-gel coating thickness, elastic modulus of the BHPA was increasing, while its output force density was decreasing. And elastic modulus of the BHPA reached the lowest value of 3.1 MPa and its output force density achieved the largest value of 13.34 mN/g under the sol-gel coating of 1 coating thickness, which was separately reduced by 32.61% of 4.6 MPa and was enhanced about 15.10% of 11.59 mN/g of that sample adopting the traditional hot laminating of 1 layer. To summarize, the larger the sol-gel coating thickness of the BHPA, the greater its elastic modulus and the more the bending interior stress needed to be overcome with the better output force characteristics.

Fig. 9. Tensile properties of the BHPA samples based on various assembly process factors: (a) tensile stress and tensile strain, and (b) elastic modulus and output force density.

4. Enhancement mechanism analysis
### 4.1. Microscopic structure of the BHPA

SEM images of the BHPA samples were used for their analysis and capture of the surface and cross-sectional microstructure by different assembly process factors. As shown in Fig. 10(a) ~ Fig. 10(d), with the increase of the MWCNT-SA based sol-gel coating thickness, surface microstructure of the BHPA was more and more uneven and unpolished than that of the sample by the traditional hot laminating technology, which reduced its tightness between membranes of the electrode membrane and electrically actuated membrane. Therein, by the sol-gel coating of 1 coating thickness, surface microstructure of the BHPA was very uniform and smooth with few gaps, so that the connection between the membranes was reliable and the contact resistance was reduced, which enhanced electrically actuated performance of the BHPA effectively. Fig. (e) ~ Fig. (h) presented that the electrode membrane and electrically actuated membrane of the BHPA could be clearly distinguished, and their contact surfaces penetrated and fitted each other with few gaps and spacing in the interface. With the increase of the MWCNT-SA based sol-gel coating thickness, the folds in each membrane raised and its internal cracking phenomenon gradually appeared, but it was still better than the cross-sectional microstructure of the BHPA sample by the traditional hot laminating technology. Since fluidity of the sol-gel coating filled the gaps and spacing between the membranes, their chimerism and connectivity were the best by the sol-gel coating of 1 coating thickness, which significantly reduced contact resistance of the BHPA with little possibility of cracking during the work, and improved its specific capacitance with good output force characteristics.

![Fig. 10. Surface and cross-sectional microstructure of the BHPA samples based on various assembly process factors (5,000 times): (a) ~ (d) the surface microstructure by the sol-gel coating of 1, 2 and 3 coating thicknesses, and by the traditional hot laminating technology respectively, (e) ~ (h) that of the cross-sectional microstructure.](image)

### 4.2. Electron spectroscopy of the BHPA

Energy spectrum of the BHPA before and after its output-force tremor test was utilized to quantitatively explore the distribution change of constituent elements, as shown in Fig. 11. It was illustrated that in the failure sample, the contents of element carbon (C), element oxygen (O), element sodium (Na) and element hydrogenium (H) increased slightly, was basically unchanged, reduced and lessen dramatically respectively, because the cations migrated directionally and accumulated a large amount of Na⁺ in the interface in contacting with the electrode membrane, resulting in a serious decrease in the amount of element Na inside the electrically actuated membrane under the excitation of electric field. Besides, it was penetrated with a small amount of the element C passed through the interface, but the element O was fixed in the polymer structure, and a considerable amount of the element H moved with hydrated electric ions from the surface to the other side of the BHPA due to its interior electrostatic force, i.e. Coulomb force.
Meanwhile, Fig. 12(a) presented that the XRD peak value of SA was the lowest owing to its weakest crystallinity, and the XRD peak angles of the MWCNT and MWCNT-SA were the same. In high molecular polymers, the crystallinity basically represented the conductivity of electric ions, and the small crystallinity made molecules closely stacked, thus the electric ion mobility increased and the conductivity promoted. It was proved that as a non-metallic material, the XRD peak value of the MWCNT was directly proportional to its internal crystallinity, whose drying state formed the MWCNT-SA and its XRD peak value was very high indicating excellent conductivity. Additionally, as shown in Fig. 12(b), the characteristic peak of SA was strong and had a large moving displacement towards low wave number, which were more remarkable than that of the MWCNT-SA by chemical reaction, along with the weak hydrogen bond between molecular structures.

Fig. 12. Material properties of the BHPA based on different characterization methods: (a) diffraction of x-rays (XRD) pattern, and (b) Infrared Spectroscopy (IR).

4.3. Enhancement mechanism

In order to further study the interaction between electrically actuated performance of the BHPA and its own electrochemical properties, an equivalent circuit model for the BHPA was presented in Fig. 13, where its middle layer was the electrically actuated membrane regarded as an ionic conductive material, and it could be modeled in parallel with capacitor \( C_1 \) and resistor \( R_1 \). Both of two contact interfaces of the BHPA had contact resistance between the electrically actuated membrane and the electrode membrane bonded on both sides, which could be modeled by two resistors \( R_{C1} \) and \( R_{C2} \) respectively. Although surface resistance of the electrode membrane made of the MWCNT-SA was far less than \( R_1 \) and \( R_C \), it could not be ignored about 9.44 \( \Omega/cm \), which was represented by two resistors \( R_{S1} \) and \( R_{S2} \) respectively. Further analysis showed that the excitation intensity of electric field needed to be moderately increased by reducing \( R_C \), so as to enhance the electrically actuated performance of the BHPA with a reliable working efficiency. This was because the excessive excitation intensity would make the
electrostatic force (Coulomb force) inside the BHPA relatively weak with an accelerating migration speed of electric ions, and the ion aggregation at the contact interface continued to increase with a rising block level of the ion channels, which was easy to break down the electrically actuated membrane, resulting in its failure and shortening its service life. By contrary, the too low excitation intensity of electric field could lead to the prominent electrostatic repulsion between electric ions in the BHPA, and it was macroscopically manifested in the small output force and its poor tremor behavior.

Fig. 13. Equivalent circuit model for electrically actuated performance of the BHPA.

Further, based on the equivalent circuit model for electrically actuated performance of the BHPA, its peak current $I$ (A) could be deduced comprehensively, which was obtained from the formula:

$$I = \frac{U}{R_{R1} + R_{R2} + R_1} + \frac{\varepsilon_r \varepsilon_0 S}{4\pi kd} \frac{dU}{dt},$$  \hspace{1cm} (7)$$

where the first term represented the steady-state current from the resistor $R$ and the second term represented the transient current from the capacitor $C$; $U$ (V) was the excitation intensity of electric field; $S$ ($m^2$), $d$ (m) and $\varepsilon_r$ was the cross-sectional area, thickness and dielectric constant of the electrically actuated membrane respectively; and $k$ was considered as constant and the dielectric constant $\varepsilon_0$ of the vacuum medium was about $8.85 \times 10^{-12}$ F/m.

In the actual working process, the load on the BHPA did not exceed its elastic limit. And then under the excitation of electric field, the qualitative formula was derived in describing the bending deformation, peak current and bending stiffness of the BHPA based on its experimental testing data and basic physical parameters, which was expressed as follows:

$$D = \frac{I}{K} = \frac{12h \delta}{kh^2 \sigma},$$  \hspace{1cm} (8)$$

where $D$ (mm) was the bending deformation; $I$ (A) was the peak current; $K$ (N/m) was the bending stiffness; $b$ (mm) was the aggregate thickness; $h$ (mm) was the free end length; $\sigma$ (MPa) was the tensile stress and $\delta$ (%) was the tensile strain.

5. Conclusions

Through the orthogonal experiment and control variates, it was concluded that the best size and testing voltage of the BHPA were 35 mm $\times$ 8 mm $\times$ 1 layer (0.352 mm) and 4 V respectively, and under the MWCNT-SA based sol-gel coating of 1 coating thickness, both of its output force density and service life achieved the maximum values of 13.34 mN/g and 330 s with the lightest tremor behavior, which was increased by 15.10% of 11.59 mN/g and was about four times of 84 s of that sample adopting the traditional hot laminating of 1 layer. Moreover, by the test and analysis of tensile property and electrochemical properties, elastic modulus
and internal resistance of the BHPA reached the minimum values of 3.1 MPa and 2.13 Ω, and its specific capacitance obtained the largest value of 81.3 mF/g under the sol-gel coating of 1 coating thickness, which was separately decreased by 32.61% of 4.6 MPa and 12.7% of 2.44 Ω, and was around five times of 17 mF/g of the sample assembled by the traditional hot laminating technology. Based on the microscopic structure, electron spectroscopy and equivalent circuit model of the BHPA, it was verified that under the sol-gel coating of 1 coating thickness, the elastic modulus and contact resistance ($R_C$) between membranes of the electrode membrane and electrically actuated membrane were greatly reduced in the BHPA, which improved its bending interior stress, internal resistance and specific capacitance with growing accumulation and migration rate of charged ions, and played a significant role in enhancing its electrically actuated performance with good output force characteristics. In conclusion, this work provided an effective approach and theoretical basis for the working stability and the development of new applications of the BHPA in the future.

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