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Actuation and blocking force of stacked nanocarbon polymer actuators

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ABSTRACT

We have developed stacked nanocarbon polymer actuators that are composed of several nanocarbon polymer actuator films using nonwoven fabric as insulation layers. The nonwoven fabric prepared through electrospinning methods has extremely-low-density structures, which do not significantly prevent the motions of each nanocarbon actuator layer. Therefore, stacking several thin nanocarbon polymer actuators using nonwoven fabric as insulation layers is expected to increase generated force without decreasing the displacement of a one-layer actuator. We have prepared stacked actuators with one, two, three, four, and seven layers using this method. The displacement and blocking force of these actuators are measured and compared with those of one-layer actuators of different thicknesses. Displacement is weakly dependent on the thickness of the actuator films of the stacked actuators. On the contrary, it decreases considerably as the thickness of the actuator film of the one-layer actuator increases. In both cases, blocking force is proportional to the thickness of actuator films. We have developed a stacked actuator model based on a trilayer actuator model and confirmed the experimental results using the model.

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1. Introduction

In recent years, interest in the research field of electroactive polymer (EAP) actuators has increased significantly because they provide several advantages compared to conventional actuators, such as large actuation strains, large compliance, low mass density, and easy processing [1,2]. They are expected to be applied to multiple human-friendly applications such as bionic implants, limb prosthetics, tactile display, and biomedical applications. EAP actuators can be divided into two main types [2], i.e. ionic, which are activated by the electrical transport of ions and/or solvents, and electronic, which are activated by an electric field. The major advantage of ionic EAP actuators is their low driving voltage (lower than 3 V) and large displacement as compared to that of electronic EAP actuators (typically kV) or even other conventional actuators.

In previous studies, we developed nanocarbon polymer (NCP) actuators based on carbon-nanotube (CNT) electrodes, which are a type of ionic EAP actuators [3,4]. (Figure 1(a)) Our

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actuator adopts a bimorph configuration with a polymer-supported internal ionic liquid (IL) electrolyte layer sandwiched between NCP electrode layers composed of CNTs, IL, and base polymers; this configuration allows for rapid and long-term operation in air at low applied voltage. ILs are nonvolatile and characterized by high ionic conductivities and wide potential windows that are advantageous for rapid responses in actuation and high electrochemical stabilities of components, respectively.

A small-sized NCP actuator (e.g. 50-μm thick, 10-mm long, and 2-mm wide) exhibits large displacement (a bending angle of more than 90°) and relatively high response (a maximum of 100 Hz) to low voltage (less than 2 V) [5]. It shows the displacements of 0.86 mm, 0.19 mm, 0.51 mm and 2.2 mm (resonance) at applied square wave voltage of 4 Vp-p with frequencies of 0.1 Hz, 1 Hz, 10 Hz and 81 Hz, respectively. (The displacement was measured at a distance 5 mm from the fixed point) [6]. Moreover, the generated stress and strain of nanocarbon electrodes are estimated to be several MPa and 2%, respectively, based on the displacement of actuators and the elastic modulus of electrode layers [7,8].

Our NCP actuators are operable in air at low applied voltage, and they achieve stability for more than 100,000 cycles. However, the scale effect is an issue while applying them to power applications such as soft actuators for robotics.

When low voltage is applied, internal ions transfer to electrode layers, and these ion transports most likely result in swelling of the cathode layer and shrinkage of the anode layer, as cations are larger than anions. This results in the bending motion of an actuator to the anode side [9,10]. In a previous paper, the bending motion of an NCP actuator was successfully analyzed based on the trilayer polymer actuator model [11]. In our NCP actuator, the thickness of the NCP electrodes is large as compared to that of electrolyte

Figure 1. Nanocarbon polymer actuators. (a) One-layer actuator. (b) Stacked actuator.
layer. Therefore, we can neglect the thickness of the electrolyte layer in the model. In that case, according to this model, bending displacement is inversely proportional to the thickness of an actuator film and bending force is proportional to the square of the thickness. Furthermore, bending speed is inversely proportional to the square of the thickness. Therefore, there are trade-off relations between the thickness of the actuator film (electrode and electrolyte) and bending displacement, speed, and force.

In several studies, stacking several thin actuators has been used as a method of solving this issue of trade-off relations [12–17]. (Figure 1(b)) In this case, the most important issue is that the insulation layer between actuator electrodes prevents the contraction/elongation of each electrode layer, which decreases the motion of the stacked actuator. Therefore, we should use an insulation-layer material that exhibits extremely low friction. In this paper, we propose a new method of stacking thin NCP actuator films using nonwoven fabric as insulation layers between the actuator layers. The fabric is created using the electrospinning method, and it is expected that the fabric will not prevent the bending motion of each actuator layer significantly.

2. Experimental method

2.1 Materials

Single-walled CNTs were synthesized through water-assisted CVD, which is referred to as the super-growth (SG) method [18]. SG-CNTs were supplied by CNT-Application Research Centre, AIST. Other reagents were received from NEC Co. Ltd., (carbon nanohorn (CNH)), IoLiTec Co. Ltd. (1-ethyl-3-methylimidazolium tetrafluoroborate (EMIBF₄)), Arkema Chemicals Inc. (poly(vinylidene fluoride-co-hexafluoropropylene); Kynar Flex 2801® (2801) and 2851®(2851)), Kishida Chemicals Co. (N-methyl-2-Pyrrolidone (NMP)), Sigma Aldrich (propylene carbonate (PC)), and Wako Pure Chemicals (N,N-dimethylformamide (DMF) and Acetone (Ac)).

The base polymer 2851 is harder than 2801. Therefore, 2851 was used for the base polymer for the hard nanocarbon electrode layer and 2801 was used for the soft electrolyte layer and polymer nonwoven fabrics as the insulations layers since the electrode layers generate the electromechanical stress, while the electrolyte layers and insulation layers are passive layers for bending motion.

2.2 Preparation of nanocarbon electrode films

A suspension of the SG-CNTs (0.5 wt%) was dispersed uniformly in NMP (99.5 wt%) using a Nanovater® (Yoshida Kikai Co. Ltd) for fabricating the SG-CNT dispersion. A mixture of CNH (2 wt%), 2851 (2 wt%), and EMIBF₄ (12 wt%) was dispersed uniformly in NMP (84 wt%) for 15 min at 2,000 rpm using a Planetary Centrifugal Mixer (THINKY model ARE-110 (ARE-110)) for fabricating the CNH/2851/EMIBF₄ dispersion.

Electrode solutions were obtained by mixing the SG-CNT solution (20 g) with the CNH/2851/EMIBF₄ solution (5 g) for 15 min at 2,000 rpm using ARE-110. A portion of the electrode solution was coated on a flat glass plate using a doctor blade heated to 80°C. The solvent was evaporated for one day, and a nanocarbon electrode film was obtained. (Figure 2)
Nanocarbon electrodes of four different thicknesses, i.e. E81 (0.081 mm), E99 (0.099 mm), E137 (0.137 mm), and E188 (0.188 mm), were fabricated by varying the gap size between the glass plate and doctor blade.

2.3 Preparation of the electrolyte film

A mixture of 2801 (10 wt%) and EMIBF₄ (10 wt%) in a mixed solvent composed of PC (15 wt%) and DMF (65 wt%) was stirred for 2 h at room temperature (RT). Then, a portion of the resulting gelatinous mixture was coated on the flat glass plate at a gap size of 0.2 mm using a doctor blade heated to 65°C. The solvent was evaporated for 2 h, and a polymer gel electrolyte film with a thickness of 15 μm was obtained. (Figure 2)

2.4 Preparation of the polymer nonwoven fabrics

2801 (20 wt%) was stirred in a mixed solvent composed of Ac (40 wt%) and DMF (40 wt%) for 2 h at RT. Then, a portion of the resulting gelatinous mixture was injected to a collector covered with aluminum foil using an injection syringe. We obtained the polymer nonwoven fabric on the collector by applying a voltage of 20 kV between the injection syringe and collector using an electrospinning system (MECC model NANON). The distance between injection nozzle and collector was 15 cm, the injection rate was 0.2 ml/hour, and the nozzle diameter was 0.21 mmφ. The thickness of the fabric was 10 μm. Figure 3 shows the electrospinning system and the SEM image of the fabric. As
shown in the figure, the fabric is composed of a number of 2801 fibers with diameters of hundreds of nanometers. The density of the non-woven fabrics estimated from its weight and volume was 0.658 g cm\(^{-3}\), the value of which is very low as compared to that of 2801 (1.77 – 1.80 g cm\(^{-3}\)) from catalogue [19] and represents the porous structure of non-woven fabrics. The strain-stress test of the prepared non-woven fabrics was carried out with SEIKO TMASS 6100. The obtained strain-stress curve was shown in Figure S1. From Figure S1, we estimated the value of Young’s modulus of non-woven fabrics, 21.5 MPa.
2.5 Preparation of the stacked actuator strips

Figure 4 shows the preparation of the stacked actuator strips. The electrolyte film was sandwiched between two NCP electrode films and subsequently placed between the hot-press plates of a small a crew-type hot-press apparatus. Then, the three layers were pressed (2 Nmm\(^{-2}\)) at 70°C for 5 min to adhere them with each other, and the NCP actuator was obtained.

For the preparation of the stacked actuators, the nonwoven fabric was sandwiched between the NCP actuators and then pressed (0.2 Nmm\(^{-2}\)) at RT for 5 min to adhere the layers. Finally, area A (A, A’, and A”) and area B (B, B’, and B”) were pressed (5 Nmm\(^{-2}\)) at 70 °C for 5 min, as shown in Figure 4, to adhere the NCP electrodes to connect electrical pass, and the stacked NCP actuators were obtained.

Table 1 summarizes the types of NCP electrodes, number of stacked layers (stacking number), and thickness of the NCP actuators.

2.6 Characterization

Actuation testing was performed using a displacement measurement system composed of a laser displacement meter (KEYENCE model LK-GD500), a potentiogalvanostat (HOKUTO DENKO model HAL 3001), a waveform generator (Tektronix model AFG 3022), and an oscilloscope (YOKOGAWA ELECTRIC model DL 850 E).
The blocking force testing of the NCP actuator was performed using an ultra-low-capacity load cell (KYOWA ELECTRONIC INSTRUMENTS model LTS-50GA) and an instrumentation amplifier (KYOWA ELECTRONIC INSTRUMENTS model WGA-670B).

One end of an NCP actuator strip was clipped by two gold disk electrodes. Displacement and blocking force were measured at a point located 4 mm away from a fixed point under applied square-wave voltages of ± 2 V and ± 2.5 V, respectively, at various frequencies. (Figure 5)

3. Results and discussion

The purpose of this study is to solve the issue of the trade-off relations between the thickness of the NCP actuator and displacement, displacement speed, and generated force by stacking several thin actuator films. The most important technical issue in

| NCP electrode film | Stacking number | Thickness of actuators (mm) |
|--------------------|----------------|----------------------------|
| Actuator 81–1 E81  | 1              | 0.141                      |
| Actuator 81–2 E81  | 2              | 0.303                      |
| Actuator 81–3 E81  | 3              | 0.463                      |
| Actuator 81–4 E81  | 4              | 0.634                      |
| Actuator 81–7 E81  | 7              | 1.225                      |
| Actuator 99–1 E99  | 1              | 0.195                      |
| Actuator 137–1 E137| 1              | 0.266                      |
| Actuator 188–1 E188| 1              | 0.337                      |

Figure 5. Schematic view of displacement and generated force measurements for nanocarbon polymer actuators.
achieving this is that the insulation layer between each actuator electrode prevents the contraction/elongation of each electrode layer, which decreases the motion of the stacked actuator. Therefore, we should use an insulation-layer material that exhibits extremely low friction. We propose a new method of stacking thin nanocarbon actuator films using nonwoven fabric as insulation layers between each actuator layer. The fabric is created using the electrospinning method, and it is expected that the fabric will not prevent the bending motion of each actuator significantly.

By using the method as shown in Figure 6(a), the SEM image of the cross section of the stacked NCP actuator during bending motion was obtained. When the voltage was applied to the stacked NCP actuator, it bends. Then, the middle of the actuator was cut during the bending by opening the connecting between two NCP electrodes. The cross section of the actuator film was observed by the SEM, which was shown in Figure 6(b). As seen in Figure 6(b), the nonwoven fabric adheres extremely well to both electrode layers of each actuator layer. However, its density appears to be considerably low. Therefore, it is considered that the nonwoven fabric with the low-density structure does not significantly prevent the elongation and contraction of each nanocarbon electrode layer driven by electric voltage. Based on this result, the stacking of several thin nanocarbon actuators using nonwoven fabric as insulation layers is expected to increase generated force without decreasing the displacement of the one-layer actuator.

Figure 7(a) shows the frequency dependence of the displacement of one-layer actuator strips of different thicknesses driven by a square-wave voltage of ± 2 V. It is clearly seen from figure that displacement decreases considerably as the thickness of the actuator film increases. Figure 7(b) shows the frequency dependence of the blocking force of the actuator films of various thicknesses driven by a square-wave voltage of ± 2.5 V. Blocking force
increases slightly with the thickness of the actuator film. In all data on frequency dependence of the displacement and the blocking force, the displacement and the blocking force decreases with increasing the frequency. The frequency dependence of displacement for the NCP actuator was reported previously [20,21]. It was reported that their frequency dependence can be analyzed by taking into account the electrochemical equivalent circuit for the NCP actuator. In this paper, we will not further discuss this point.

Figure 8(a) shows the frequency dependence of the displacement of stacked actuator strips with various stacking numbers driven by a square-wave voltage of ± 2 V. In contrast to the case of the change in the thickness of the actuator film, displacement is only weakly dependent on the stacking number of the actuator films. It decreases slightly as stacking number increases. Figure 8(b) shows the frequency dependence of the blocking force of the stacked actuator films. As compared to the case of the change in thickness, blocking force increases considerably with stacking number.
Figure 9 shows the plots of the displacement and blocking force at the lowest frequency against the thickness of the actuator film of the one-layer and stacked actuators. As shown in Figure 9(a), displacement is inversely proportional to the thickness of the actuator film of the one-layer actuator. On the contrary, it is weakly dependent on the thickness of the actuator films of the stacked actuator. In both cases, blocking force is proportional to the thickness of the actuator films, as shown in Figure 9(b).

As previously mentioned, several authors reported that multilayer of thin actuators was used for developing actuator showing both large displacement and force [12–17]. They also reported the multilayer actuator model. However, the multilayer actuator models previously reported in other references are under different conditions and they are difficult to apply to the experimental results in this paper. In order to analyze the experimental results, therefore, we develop a stacking actuator model based on a tri-layer actuator model [11,22].

As mentioned in the introduction, when voltage is applied between two nanocarbon electrodes, cations transfer to the cathode and anions transfer to the anode; this results in the elongation and contraction of each layer. The strain generated in each electrode layer when voltage $V$ is applied between two electrode layers is assumed to be independent of the duration of the application of voltage $V$ and the position in the electrode layer. In this case, the strains generated in the elongated cathode layer and contracted anode layer are
assumed to be $\alpha$ and $-\alpha$, respectively. In the case of the bending ionic EAP actuator, the trilayer model is based on the following assumptions: (Figure 10(a))

1. The electrode and electrolyte layers are elastic and isotropic. The Young moduli and Poisson’s ratio of both layers remain unchanged throughout the range of bending considered.
2. The change in the thickness of the electrode layer under applied voltage is negligible as compared to the thickness of the electrode layer.
3. The rate of ions entering and leaving the electrode layers during charging/discharging processes is constant along the actuator length.
4. The strain at any cross section of the actuator is symmetric about the neutral axis, and the stresses generated in the actuator are caused solely by the elongation or contraction of the two nanocarbon electrode layers.

Under these assumptions, the total bending moment, which is composed of the induced internal bending moment and the moment created by external force $F$, is always zero at the cross section along the actuator. This gives the following equation:

$$\frac{1}{R}EI + aE_1bh_1(h_1 + h_2) - FL = 0,$$

where $EI = E_1I_1 + E_2I_2$ is the flexural rigidity of the entire actuator, $E_1$ and $E_2$ are the elastic moduli of the electrode and electrolyte layer, respectively, and $I_1$ and $I_2$ are the area moments of inertia of each layer, which are given by the following equations:
\[ I_1 = \frac{2b((h_2/2) + h_1)^3}{3} - \frac{2b(h_2/2)^3}{3} \]  

(2)

\[ I_2 = \frac{2b(h_2/2)^3}{3} \]  

(3)

where \( h_1 \) and \( h_2 \) represent the thicknesses of the electrode and electrolyte layer of the actuator, respectively, and \( b \) and \( L \) are the width and length of the actuator film, respectively.

\( 1/R \) is the curvature of the actuator (\( R \): curvature radius), which is given by the following equation, provided that external force is 0 in Equation (1):

\[ \frac{1}{R} = -\frac{E_1 bh_1(h_1 + h_2)}{EI} \]  

(4)

Displacement \( \delta \) is easily related to curvature by the following equation:
Similarly, blocking force $F$ is given by the following equation:

$$F = -\frac{??E_1 b h_1 (h_1 + h_2)}{L}$$  \hspace{1cm} (6)$$

The generated strain, $\alpha$, can be considered to be proportional to the induced charge in the electrode layers. Therefore, it is given by the following equation:

$$\alpha = kCV$$  \hspace{1cm} (7)$$

The response speed of generated strain $\alpha$ is apparently proportional to that of induced charge. Electrochemical response time is represented by the equivalent circuit of the nanocarbon actuator film, which is simply represented by a serial equivalent circuit of the electric resistance of an ionic gel layer ($R$) and the double layer capacitance of electrode layers ($C$).

According to Equation (4), bending displacement (curvature) is inversely proportional to the thickness of the electrode layer, which is in agreement with the experimental results, as shown in Figure 9(a). According to Equation (6), blocking force is proportional to the square of the thickness of the actuator film. The data in Figure 9(b) is not sufficient for comparison with Equation (6).

Here, we propose a new bending actuator model of the stacking actuator based on the trilayer actuator model and apply it to the experimental results shown in Figure 7–9. Figure 10(b) shows a schematic view of the stacked-actuator model. Subscripts 1, 2, and 3 denote the electrode layers, electrolyte layer, and insulation layers, respectively. Under the same assumptions of the trilayer model described previously, the total bending moment for one focused layer is always zero.

Then, we obtain the following equation:

$$n \left[ \frac{1}{R} (E_1 l_1 + E_2 l_2 + E_3 l_3) + a E_1 b h_1 (h_1 + h_2) \right] - FL = 0,$$  \hspace{1cm} (8)$$

where $n$ is the number of the stacking layers, $E_1, E_2, l_1, l_2, a, b, h_1, h_2, F$, and $L$ are given by the previous equations, $E_3$ the elastic modulus of the insulation layer, and $l_3$ is given by the following equation:

$$l_3 = \frac{2b \left( h_3^2 + h_2^2 + h_1 \right)^3}{3} - \frac{2b \left( h_2^2 + h_1 \right)^3}{3}$$  \hspace{1cm} (9)$$

where $h_3$ the thickness of the insulation layer.

Curvature, which is directly related to bending displacement according to Equation (5), is given by

$$\frac{1}{R} = \frac{2\delta}{L^2 + \delta^2};$$  \hspace{1cm} (5)$$

Blocking force $F$ is given by the following equation obtained by substituting $1/R = 0$ in Equation (8):
Bending displacement depends on the rigidity of the insulation layer, \( E_3 I_3 \). If \( E_3 I_3 \ll E_1 I_1 + E_2 I_2 \), it is independent of the thickness of the insulation layer. As shown in Figure 9(a), bending displacement is almost independent of the thickness of the stacked actuator, and hence, it is independent of stacking number. The slight decrease in the displacement above a thickness of 0.6 mm is owing to the problem of the fabrication of multiple stacked layers. In the case of more than 4 stacked layers, it is difficult to obtain a good electrical connection and good actuation response. Blocking force is proportional to stacking number. Therefore, it is proportional to the thickness of the stacked actuator film, which is in agreement with the experimental results shown in Figure 9.

4. Conclusions

We developed stacked NCP actuators composed of several NCP actuator films using non-woven fabric as insulation layers. The nonwoven fabric prepared using the electrospinning method has extremely-low-density structures, and it does not significantly prevent the motions of each nanocarbon actuator layer. Therefore, stacking several thin NCP actuators using nonwoven fabric as insulation layers is expected to increase generated force without decreasing the displacement of the one-layer actuator. We prepared stacked actuators with one, two, three, four, and seven-layers using this method. Their displacement and blocking force were measured and compared with those of one-layer actuators of different thicknesses. Displacement was weakly dependent on the thickness of the actuator films of the stacked actuators, whereas it decreased significantly as the thickness of the actuator film of the one-layer actuator increased. In both cases, blocking force was proportional to the thickness of the actuator films.

We developed a stacked actuator model based on a trilayer actuator model. Based on the model, in the case of the one-layer actuator, bending displacement was inversely proportional to the thickness of the electrode layer and blocking force was proportional to the square of the thickness of the actuator film. In the case of the stacked actuator, bending displacement was independent of stacking number and blocking force was proportional to stacking number. The experimental results shown in Figure 9 were in good agreement with modeling results.

Disclosure statement

No potential conflict of interest was reported by the authors.

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