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Yutaka Chida^a, Tsuyoshi Morita^a, Ryuichi Machida^a, Daiki Hoshino^a & Yasushiro Nishioka^a

^a Department of Precision Machinery Engineering, College of Science and Technology, Nihon University, Funabashi-shi, Chiba, Japan

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Fabrication of Bi-Directional Bending Organic Actuators Consisting of Stacked Polypyrrole Films With Different Expansion and Contraction Ratios

YUTAKA CHIDA, TSUYOSHI MORITA,
RYUICHI MACHIDA, DAIKI HOSHINO, AND
YASUSHIRO NISHIOKA

Department of Precision Machinery Engineering, College of Science and Technology, Nihon University, Funabashi-shi, Chiba, Japan

An organic conductor, polypyrrole has been extensively studied as one of the prospective soft actuator materials. In this paper, simple bimorph actuators consisting of an electropolymerized polypyrrole (PPy) film and an adhesive tape were fabricated. It turned out that those actuators nonuniformly bent depending on the distance between the actuator and the counter electrode. Then, a structure consisting of polypyrrole/both-side adhesive tape/polypyrrole, whose two polypyrrole films have different extension/contraction ratios, has been fabricated, and it was found that the actuator exhibited more uniform bending deformation which was nearly independent of the distance between the actuator and the counter electrode.

Keywords Actuator; conducting polymer; polypyrrole; PPy

1. Introduction

Organic soft actuators attract strong attentions because they have many advantages compared to conventional mechanical actuators. Organic soft actuators are generally light and flexibly deformed. In addition, they operate under low voltage ranges as low as 1 volt or so, and generate no sound during deformation. Amongst those soft actuators, bending soft actuators are of special interest because small volume change in organic materials can cause a large bending displacement. For example, soft actuators consisting of Ionic Conducting Polymer Films (ICPF) have been widely used as bending actuators [1]. However, the fabrication processes for ICPF actuators seem complicated. In contrast, polypyrrole films synthesized using electropolymerization as a new material for organic soft actuators have been

Address correspondence to Yasushiro Nishioka, Department of Precision Machinery Engineering, College of Science and Technology, Nihon University, 7-24-1 Narashinodai, Funabashi-shi, Chiba 274-8501, Japan. Tel.: +81-47-469-6482; Fax: +81-47-469-6482; E-mail: nishioka@eme.cst.nihon-u.ac.jp

extensively studied [2–6]. In addition, the amount of the volume change can be modified by altering the electropolymerization conditions. Hara *et al.* recently reported that the expansion and contraction ratio of their polypyrrole actuators exceeded 40%, which is very encouraging [3]. Bending actuators can be easily fabricated by forming a bimorph structure consisting of a polypyrrole film and other film material.

In this work, simple bimorph actuators using an electropolymerized polypyrrole (PPy) film and a both-side adhesive tape were fabricated. It turned out that those actuators nonuniformly bent depending on the distance between the actuator and the counter electrode. We fabricated a structure consisting of polypyrrole/both-side adhesive tape/polypyrrole whose two polypyrrole films have different extension/contraction ratios, and found that the actuator exhibited more uniform bending deformation which was nearly independent of the distance between the electrodes.

2. Principle of PPy Actuator

Figure 1 describes the principle of the PPy actuator functions. A PPy actuator and a counter electrode are placed in an electrolysis solution. When a negative voltage is applied to the actuator, negative ions in the solution will be driven out from the actuator, the actuator shrinks. This is called dedoping process. When positive voltage is applied to the actuator, the negative ions are absorbed into the PPy actuator (doping process), and the actuator expands. As shown in the lower figure, the conductive PPy polymer networks are loosely dangled, and the spacing between the polymer chains can expand and shrink during the doping and dedoping processes.

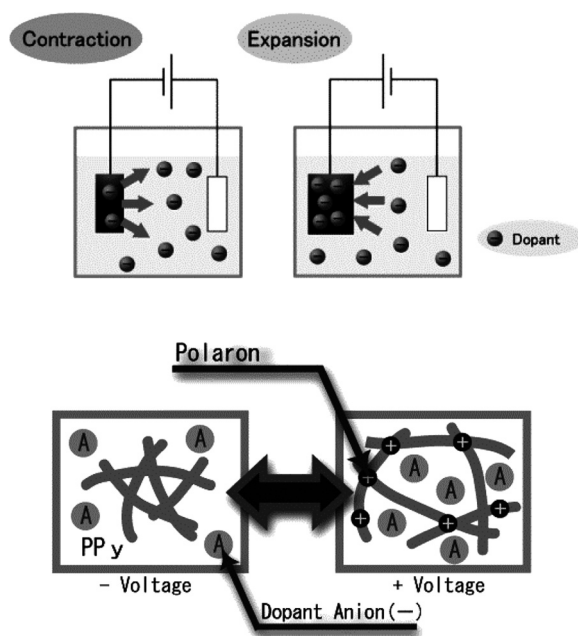


Figure 1. Principle of PPy actuator function.

3. Experimentals

3.1. Galvanostatic Electropolymerization of Pyrrole

Figure 2 describes the experimental setup for the Galvanostatic electropolymerization to form PPy film. A counter electrode (Ti), a reference electrode (Ag/AgCl), and a working electrode (Ti) were immersed into a solvent containing pyrrole monomers and an electrolyte, and the bias voltage was controlled to keep constant current between the counter electrode and the working electrode during the PPy polymerization. The PPy film deposited on the working electrode was peeled off, and was used as an actuator material. The characteristics of PPy polymers had very different characteristics influenced by different synthetic conditions of the Galvanostatic electropolymerization. For example, if different electrolytes were used, the expansion contraction ratios were very different [7]. The polymerization was done using a computer controlled potentiostat (HZ-5000, Hokuto Denko Corp.).

Here, two PPy films with different expansion/contraction behaviors were prepared under the conditions listed below.

- (a) PPy1; Electrolyte: N,N-Diethyl-N-methyl-N-(2-methoxyethyl) ammoniumbis (trifluoro methane sulfonyl) imide, Solvent: Methyl Benzoate, Room temperature, Current: 0.55 mA/cm^2 , Deposition time: 4 hrs)
- (b) PPy2; Electrolyte: tetra-*n*-butylammonium trifluoromethanesulfonate (TBACF₃SO₃), Solvent: Methyl Benzoate, Room temperature, Current: 0.50 mA/cm^2 , Deposition time: 2 hrs)

Here, two films of PPy1 with the thickness of $68.8 \mu\text{m}$ and $138.8 \mu\text{m}$, and a film of PPy2 with the thickness of $28.6 \mu\text{m}$ were prepared. Three samples of PPy1 ($68.8 \mu\text{m}$)/adhesive tape, PPy1 ($138.8 \mu\text{m}$)/adhesive tape, and PPy1 ($138.8 \mu\text{m}$)/adhesive tape/PPy2 ($28.6 \mu\text{m}$) were fabricated. The PPy film and the adhesive tape film were stacked together. The stacked structure was cut to form actuators with the appropriate size. The area of the samples investigated here was $5 \times 20 \text{ mm}^2$.

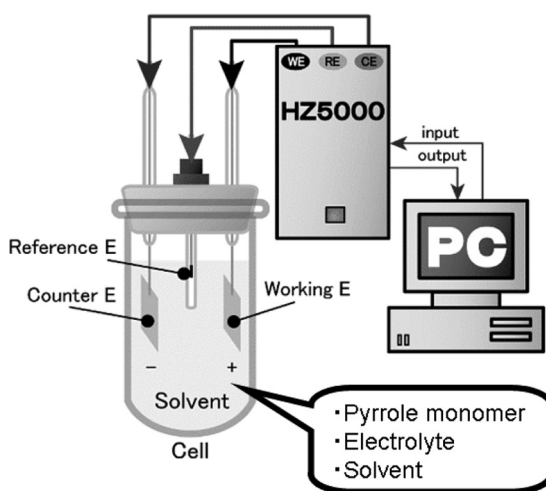


Figure 2. Setup for Galvanostatic electropolymeriation.

The adhesive tape used here is a both-side adhesive tape (NW-10S) produced by Nichiban Inc.

3.2. Actuator Characterizations

The bending experiments for the fabricated devices were performed using the same experimental setup for the PPy polymerization as shown in Figure 2. Here, the potentiostat HZ-5000, Hokuto Denko Corp. was also used to perform the actuation experiments. The top part of the PPy actuator was connected to the working electrode using a metal clip to make the electrical contact. Then, the counter electrode, the reference electrode, and the the actuator were immersed in a water solution of an electrolyte, lithium bis-trifluoromethane sulphonyl imide (LiTFSI). A bias voltage was given at the working electrode during the bending experiments. The bias ranges was $-1.0 \sim +1.0$ V, and the bias sweep rate was 10 mV/s. Please not that only the lower part of the actuator was immersed in the LiTFSI solution, and that the conducting PPy actuator acts as the working electrode.

4. Results and Discussions

Figures 3a and 3b show the photographs during deformation of the PP1 ($68.8 \mu\text{m}$)/adhesion tape actuator. Obviously, the bi-directional bending of the actuator was observed. However, as seen in the Figure 3a, the actuators edge area close to the counter electrode shows a curl, and the actuator bending was not uniform. Similar phenomenon was also seen when the PPy1 ($138.8 \mu\text{m}$)/adhesive tape actuator as shown in Figure 4. This phenomenon may be due to the fact that the electric field between the actuator and the counter electrode increases resulting in the increase of the ionic current when the actuator comes closer to the counter electrode. Therefore, it was considered that some techniques to prevent this phenomenon were necessary. It was considered that if PPy1/adhesive tape/PPy2 with different extension/contraction ratios or different thickness may prevent non-uniform bending of the actuators. Figure 5 exhibits the bending characteristics of the PPy1 ($138.8 \mu\text{m}$)/both-side adhesive tape/PPy2 ($38.8 \mu\text{m}$). Please not that the actuator shown in Figure 5 shows pretty uniform bending independent of the relative positions from the counter electrodes with some expense of the bending angle. The experimental results are summarized in Table 1. As seen in Table 1, the bending angle increases



Figure 3. Bi-directional bending organic actuator of PPy1 ($68.8 \mu\text{m}$)/adhesive tape.

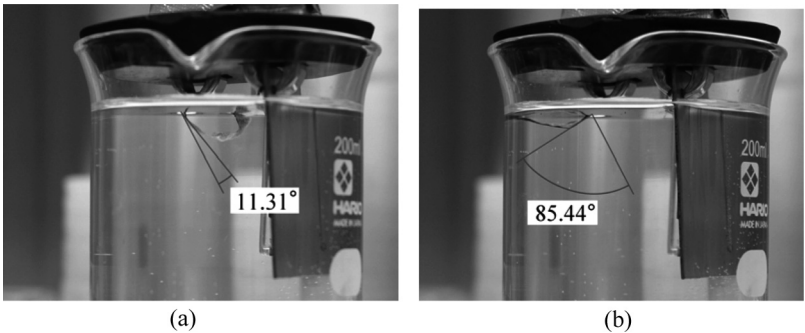


Figure 4. Bi-directional bending organic actuator of PPy1 (138.8 μm)/adhesive tape.

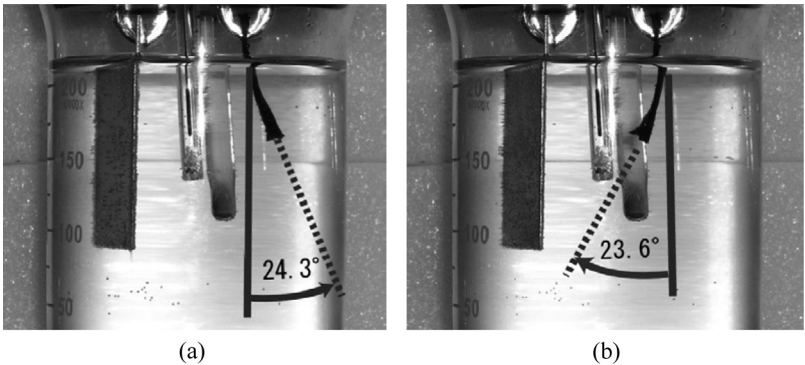


Figure 5. Bi-directional bending organic actuator of PPy1 (138.8 μm)/adhesive tape/PPy2 (28.6 μm).

when the PPy film thickness increases, and the bending angle can be adjusted by changing the thicknesses of the PPy films on both sides.

The reason for the uniform vending has still not yet been clarified. However, it might be speculated that when a part of the actuator comes closer to the counter electrode, the amount of dopants penetrating into the part will be larger, which may cause the nonuniform bending of the PPy/adhesive actuator. On the other

Table 1. Results of bending angles of the PPy actuators

Thickness [μm]				
PPy1 (counter electrode side)	PPy2 (Rear side)	Bending angle [°]		Total [°]
68.8	None	+42.4	−47.8	90.2
138.8	None	+11.3	−85.4	96.7
138.8	28.6	+24.3	−23.6	47.9

■ Positive (+) bending angle means that the actuators bend towards the counter electrode, and negative (−) bending angle means that actuators bend towards the counter electrode.

hand, this nonuniform bending may be canceled in the PPy1/adhesive tape/PPy2 structure, because the amounts of the dopants of the PPy1 and the PPy2 films near the counter electrode become larger simultaneously.

5. Conclusion

Two kinds of polypyrrole films were Galvanostatically fabricated, and the polypyrrole/adhesive bending actuators were fabricated. The actuators exhibited a large bending angle range of nearly 90°. However, when the actuators approached the counter electrode, the bending angle increased which resulted in non-uniform bending of the actuators. Then, the three layered actuator of PPy1/adhesive tape/PPy2, whose PPy films have different expansion/contraction ratios, was fabricated and characterized. The three layered actuator showed pretty uniform bending deformation with some expense of the bending angle.

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