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# Fabrication and Characterization of a Polypyrrole Soft Actuator Having Corrugated Structures

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*Recently, polypyrrole has attracted much attention, because it has a high electroconductivity and can be used in a soft actuator. We have recently succeeded in increasing the expansion and contraction ratio of a polypyrrole film by fabricating a Au/polypyrrole stacked structure, whose polypyrrole film was formed on a Au thin film coated on an acrylic board as working electrode for galvanostatic electropolymerization. Here, we propose a new soft actuator with a corrugated polypyrrole film whose backside is covered by a Au film. The corrugated Au/polypyrrole actuator was fabricated by forming a polypyrrole film on a corrugated acrylic board covered by a Au thin film and successively removing the acrylic board in acetone. Consequently, the expansion and contraction ratio of the actuator increased to 11.6%, while the actuator without the corrugated structures was 8.1%.*

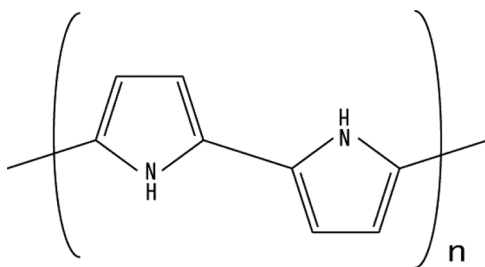
**Keywords** Actuator; conducting polymer; corrugated structures; polypyrrole; PPy

## 1. Introduction

Recently, some conducting polymers such as polypyrrole (PPy) have been extensively studied as a strong candidate for soft actuator materials. Those materials show a volume change when a voltage is given to the materials in some electrolysis solutions [1–5]. Many actuators used in the present robots and other mechanisms are servo motors. However, they are not flexible, and are usually heavy and generate phonetic noises. Thus, light-weight actuators which does not generate sound have been strongly demanded. Especially, PPy films have the strongest attentions since actuators utilizing PPy do not generate sounds during operation, and have soft natures. Important factors to use those organic conducting polymers as a soft actuator material are the expansion and contraction ratio, the generating force (stress), and the response time. PPy actuators have been regarded as a possible candidate for

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**Figure 1.** The molecular structure of PPy.

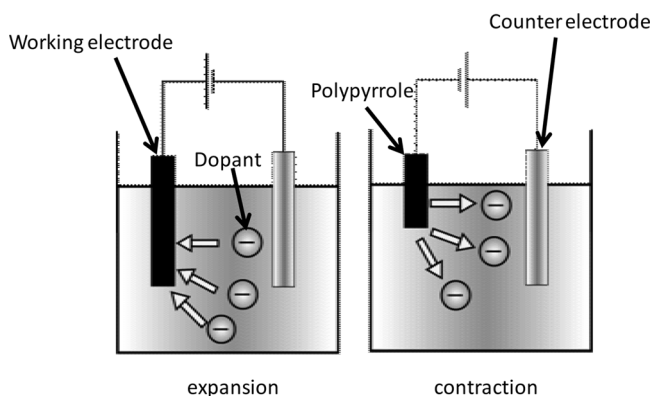
an artificial muscle because the generation of stress is nearly ten times larger than human muscles. However, the expansion and contraction ratio of a PPy actuator was only 1–3% which was far smaller than the value of human muscle, 25% [6–7]. However, Hara *et al.* recently reported that an expansion and contraction ratio of their PPy actuators exceeded 40%, which is very encouraging [6]. There are also several reports to improve the expansion and contraction ratios by modifying PPy film deposition conditions such as temperatures and modifying supporting electrolyte solutions [8–13].

In this paper, a new structure for PPy actuators to improve the expansion and contraction ratio is proposed. Here, the details of fabrication procedures and the characterization results of the corrugated PPy soft actuator are reported.

## 2. Principle of PPy Soft Actuator

Figure 1 shows the molecular structure of PPy. PPy is electro conductive since it has  $\pi$ -conjugated electrons on the polymer networks. It has been considered that the PPy polymers form sponge like networks which can expand and shrink when they absorb or desorb ions.

Figure 2 describes the conceptual description of the principle for the expansion and contraction processes of PPy actuators. A PPy film connected to the working electrode is immersed in an electrolyte solution together with a counter electrode,



**Figure 2.** The conceptual description of principle for the expansion and contraction processes of PPy actuators.

and an alternative voltage is applied between the PPy film and the counter electrode. Usually PPy actuators are driven by the anion motion. The PPy film expands by doping anions with the positive bias voltage, and the PPy film shrinks by dedoping anion with the negative bias voltage.

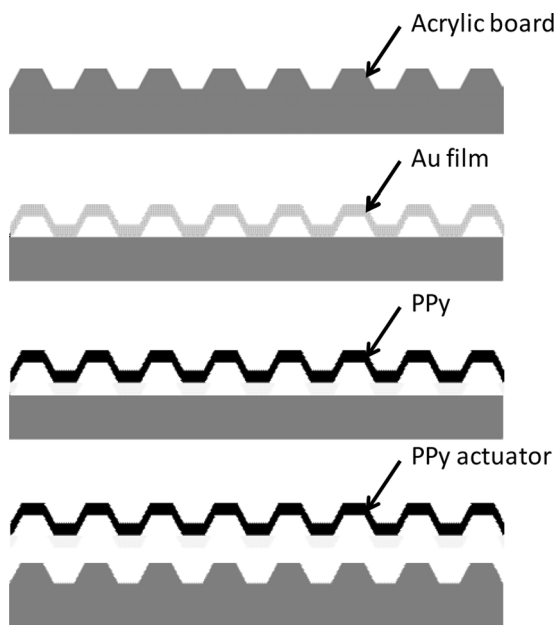
### 3. Experimentals

#### 3.1. Fabrication Processes of PPy Soft Actuator

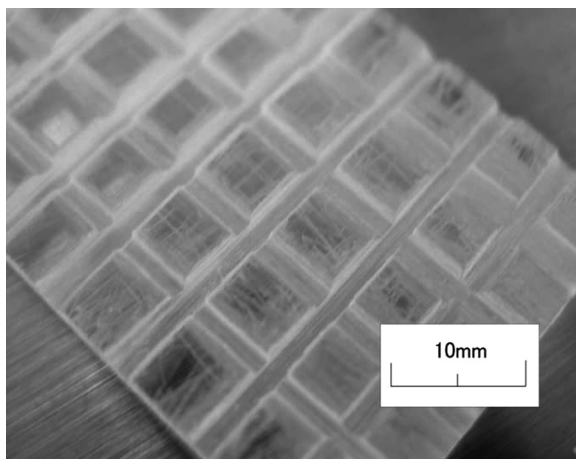
The PPy thin film was fabricated by electropolymerization. The electropolymerization of PPy was done in a methyl benzoate solution with a volume of 50 ml in which pyrrole monomers with a concentration of  $0.25 \text{ mol dm}^{-3}$  and the electrolyte (N, N-Diethyl-N-methyl-N-(2-methoxyethyl) ammonium bis (trifluoromethanesulfonyl) imide with a concentration of  $0.2 \text{ mol dm}^{-3}$  are desolved.

The working electrode was a Au thin film sputtered on an acrylic board and the counter electrode was a Ti plate. The area of the electrodes immersed in the solvent was  $30 \times 25 \text{ mm}^2$ . The polymerization was done at a constant current of  $0.2 \text{ mA cm}^{-2}$  for 4 hrs at the room temperature. The PPy thin film was polymerized with the doped anion in the supporting electrolyte. The PPy thin film was then peeled off from the electrode, and it was used as an actuator.

A working electrode with a corrugated structure and a plane structure were used for fabricating PPy thin film having the corrugated structure and the plane structure. Figure 3 describes the process to fabricate the corrugated PPy actuator. The corrugated working electrode was made by the following methods. Firstly, an acrylic board was processed to form a corrugated structure which had the ditches of 1 mm in depth, and spacing between the ditches was 5 mm as seen in Figure 4. Nextlly,



**Figure 3.** Conceptual description of the corrugated PPy actuator fabrication process.



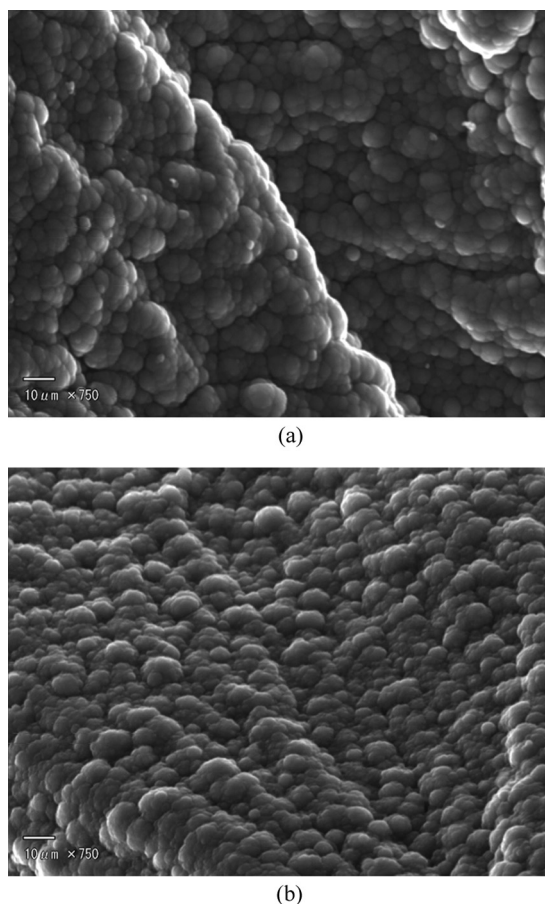
**Figure 4.** Optical microscope image of the surface of the corrugated acrylic board.

the Au thin film was coated on the processed surface of the acrylic board by sputter deposition. PPy was polymerized on the acrylic board covered by the Au thin film which worked as a working electrode. Then, the corrugated PPy thin film was peeled off from the electrode by dissolving the acrylic board in acetone. The PPy thin film was cut into slices with the width of 6 mm, and these were used as actuators. PPy films without the corrugated structure were also fabricated on the plane acrylic board with the Au film for comparison. The thicknesses of the PPy films were measured using a micrometer, and these are approximately  $14.5\mu\text{m}$  for both of the corrugated PPy film and the plane PPy film.

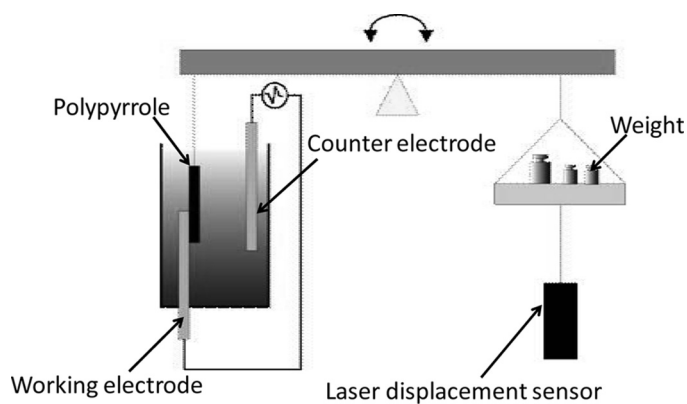
Figure 5a and 5b show the scanning electron microscope (SEM) images for the PPy films formed on the corrugated working electrode and the plane working electrode, respectively. The surfaces of the both PPy films look like a sponge surface. However, no notable differences were observed between the two films.

### 3.2. Characterization of PPy Soft Actuator

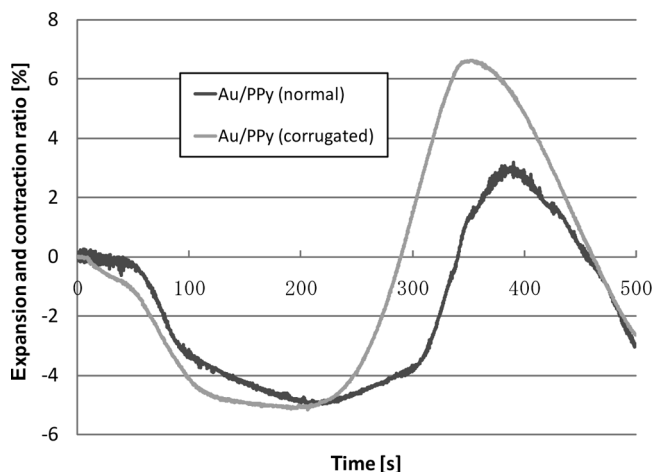
Figure 6 shows the actuator characterization system equipped with a balance to measure the expansion and contraction ratios. The PPy actuator was used as a working electrode in the LiTFSI solution of  $1\text{ mol dm}^{-3}$ , and the both of PPy actuator edges were suspended by clips. The size of the moving part of the PPy actuators was 6 mm in width, 23.8 mm in length, and  $14.5\mu\text{m}$  in thickness. The PPy actuator exhibited the expansion and contraction motions under the altering bias with the triangular wave shape applied between the PPy actuator and the counter electrode. The peak values of the bias voltage were  $-1\text{ V}$  and  $+1\text{ V}$ , and the bias voltage sweep rate was  $10\text{ mVs}^{-1}$ . The extension and contraction of the PPy actuator was measured by monitoring the displacement of the weight position using a laser displacement sensor. Moreover, an arbitrary load stress was applied on the PPy actuator by putting weights on the saucer of the balance. The weight on saucer was adjusted so that the stress to the cross section of the PPy actuator was  $0.3\text{ MPa}$ . The reason for selecting the stress of  $0.3\text{ MPa}$  was that the generated stress of human muscle is  $0.3\text{ MPa}$ .



**Figure 5.** Scanning electron microscope images of the PPy surfaces formed on the corrugated working electrode (a), and the PPy surface formed on the plane working electrode (b).



**Figure 6.** Conceptual description of experimental setup for measuring the expansion and contraction ratio of PPy actuators.



**Figure 7.** Comparison of the expansion and contraction ratios of the corrugated PPy actuator and the plane PPy actuator during one bias cycle (sweep rate  $10\text{ mVs}^{-1}$  between  $-1\text{ V}$  and  $+1\text{ V}$ ).

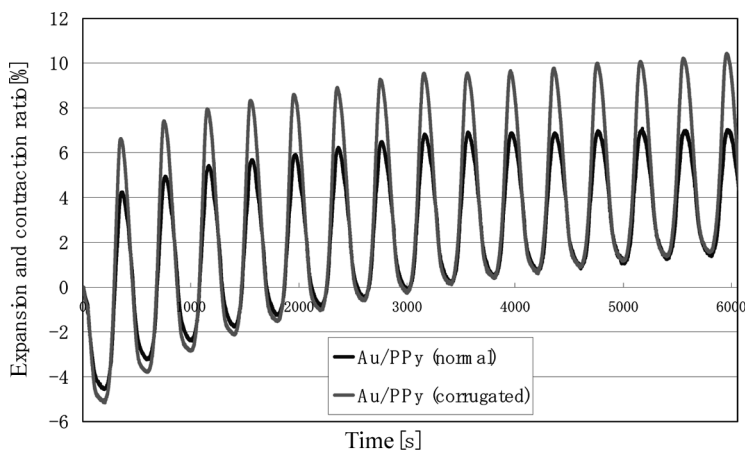
Hereafter, the expansion and contraction ratio is defined as the actuator length change divided by the initial length.

#### 4. Result and Discussion

The expansion and contraction ratios of the corrugated PPy actuator and the plane PPy actuator were compared during a bias cycle between  $-1\text{ V}$  and  $+1\text{ V}$  at the sweep rate,  $10\text{ mVs}^{-1}$  as shown in Figure 7. The expansion ratio of the corrugated actuator, 6.6% was larger than that of the plane actuator, 3.1%. However, the contraction ratios of these actuators are nearly the same. The full swing of the expansion and contraction ratio of the corrugated PPy actuator was 11.6%, while that of the plane (normal) PPy actuator was 8.1%. In addition, the response time of the corrugated PPy actuator is notably shorter than that of the plane PPy actuator.

The reason for the increase of the expansion ratio of the corrugated PPy actuator has not yet been clarified. One possible cause for that is that the corner parts of the corrugated PPy actuators expands and become more flat, and another cause is that the corrugated PPy film surface area is larger than that of the plane PPy surface. This is because the amount of the absorbed dopants in the corrugated actuator was larger than that of the plane PPy.

The behaviors of the expansion and contraction for the repeated 15 bias cycles are also shown in Figure 8. The peak and bottom values of the expansion and contraction ratios of both of the PPy actuators continued to increase as the cycle numbers increases. It is clear that the creep or memory effect is observed. In other word, the total length of the both actuators continued to increase. It should be also noted that the difference between the peak value and the bottom values becomes smaller in the plane actuator, while that of the corrugated actuator remains the same level. The mechanisms for the different behaviors of those PPy actuators are not clear. However, the corrugated PPy actuator seems to have better performances than those of the plane PPy actuator.



**Figure 8.** Expansion and contraction of the PPy actuators during 15 biasing cycles. The bias voltage range was between  $-1$  V and  $+1$  V, and the voltage sweep rate was  $10 \text{ mVs}^{-1}$ .

## 5. Conclusion

The corrugated PPy actuator was fabricated using the electropolymerization method. The PPy film was deposited on the Au film deposited corrugated acrylic board which worked as a working electrode during the electropolymerization. The acrylic board was then etched off to release the actuator structure. The fabricated corrugated PPy actuator exhibited the larger total swing of the expansion and contraction ratio of 11.6%, while that of the plane (normal) PPy actuator exhibited 8.1%. Although the creep effects was observed in the both type of the PPy actuators, the total swing of the expansion and contraction ratio remained at the similar level in the corrugated PPy actuator, and that of the plane (normal) PPy actuator continued to decrease within the voltage swing ( $-1$  V to  $+1$  V) for 15 cycles. Thus, it may be possible to conclude that the corrugated PPy actuator has better performances than those of the plane PPy actuator.

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