

Soft Actuators Prepared by Electrochemical Copolymerization of Pyrrole and Methyl 1H-pyrrole-3-carboxylate

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### 1. Introduction

Organic soft actuators made of conducting polymers such as polypyrrole (PPy) films are of special interest for applications in microelectromechanical systems (MEMS) because they generate large electrochemical stresses and electrochemical strains.<sup>1-3)</sup> However, PPy actuators usually exhibit notable electrochemical creep (swelling) after repeated electrochemical actuations. The actuating strain corresponds to the actuator's dynamic length change that is dependent on the repeated potential sweeps, while the electrochemical creep was defined as the continuous change in the length (swelling) during the actuation process. Electrochemical creep is a serious problem in practical use and needs to be minimized for precise positioning of the actuators. Tominaga et al. have recently reported that a chemical cross-link in PPy with dipyrrole alkanes suppressed the creep; however, the actuating strain also decreased.<sup>4)</sup> Ogihara et al. also reported that actuator films prepared by electrochemical polymerization of a pyrrole (Py) and diethyl 3,4-pyrroledicarboxylate showed a significant reduction in the electrochemical creep with minimal reduction in the actuating strain.<sup>5)</sup>

In this study, we attempted to modify a PPy by introducing Py derivative units into its framework and to control its actuator performances. In this study, actuator films were prepared from solutions with different mole ratios of pyrrole (Py) and methyl 1H-pyrrole-3-carboxylate.

### 2. Experimental procedures

The electrochemical polymerization of Py and methyl 1H-pyrrole-3-carboxylate was carried out. A counter electrode (Ti), a reference electrode (Ag/AgCl), and a working electrode (Ti) were immersed in methyl benzoate solutions of 0.25 M Py and methyl 1H-pyrrole-3-carboxylate in 100:0, 98:2, 95:5, and 80:20 mole ratios and 0.2M N,N-diethyl-N-methyl-N-(2-methoxyethyl)ammonium bis(trifluoromethanesulfonyl)imide. The thickness of the prepared films was measured using a micrometer and was found to be approximately 100  $\mu\text{m}$ . The PPy copolymer films were cut into  $20 \times 5 \text{ mm}^2$  strips to form the PPy actuators. The PPy actuator was used as the working electrode in 1 M LiTFSI aqueous electrolyte solutions. Both PPy actuator ends were clipped with two metal plates. The PPy actuator exhibited expansion and contraction motions under an alternating potential with a triangular wave shape applied between the actuator films and the counter electrode. The potential voltage applied between the PPy actuator and the electrolyte solution was monitored using the Ag/AgCl reference electrode. The peak values of the potential voltage were -1 and +1 V, and the potential sweep rates were  $10 \text{ mV s}^{-1}$ . A load stress of 0.3 MPa was applied on the actuator films.

### 3. Results and discussion

Figure 1 shows the time dependences of the strain of the actuators as measured by the displacement of weight as a function of time under the load stress of 0.3 MPa. The strain was defined as the change in the length of the PPy/Py copolymer actuators divided by the length prior to deformation. The characteristics of the actuators were measured in the aqueous solutions of LiTFSI.

The PPy actuator showed an actuating strain of 8%. However, the actuating strain reduced as the actuations continued, and the electrochemical creep increased after 10 cycles of actuation. In contrast, the Py copolymer film, fabricated from the Py solution containing 2 and 5 mol% of methyl 1H-pyrrole-3-carboxylate showed a reduced actuating strain of approximately 2%, but the creep strain reduced to less than 1%. Thus far, there are no plausible explanations for the improved mechanical characteristics of the actuators. However, improvement in electrochemical creep for better positioning of PPy soft actuators is identified to occur as a result of the modification of PPy structures by adding suitable chemicals.

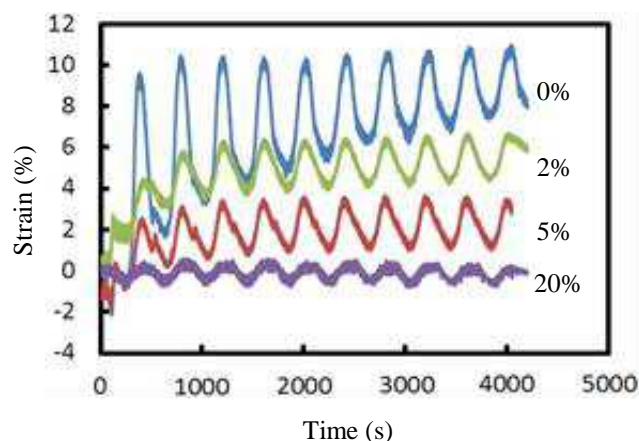


Fig. 1 Relationship between the strain and time of actuators under repeated actuations in the LiTFSI aqueous solution. The actuators fabricated from pyrrole solutions containing 0, 2, 5, and 20 mol% of methyl 1H-pyrrole-3-carboxylate were compared.

### 4. Summary

Copolymer films were prepared by electrochemical polymerization of pyrrole and methyl 1H-pyrrole-3-carboxylate with different mole ratios. The film actuators prepared from a solution of pyrrole and methyl 1H-pyrrole-3-carboxylate in a 2–5% mole ratio showed a significant reduction in the electrochemical creep with minimal reduction in the actuating strain.

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