

Enhancement in electrochemical strain of a polypyrrole–metal composite film actuator

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A polypyrrole (PPy)–zigzag metal wire composite film actuator, prepared electrochemically from a methyl benzoate solution of tetrabutylammonium trifluoromethanesulfonate (TBACF₃SO₃), showed up to 21.4% electrochemical strain, much larger than that (12.8%) of the corresponding PPy–CF₃SO₃[−], driven in aqueous NaPF₆ solution.

Conducting polymer (CP) actuators whose electrochemical strain and stress are more than 10% and 20 MPa, respectively, will make artificial muscles practicable.^{1,2} From an engineering point of view, they require CP actuators with large displacement (mm) and force (N), not strain (%) or stress (MPa), in order to fabricate practical artificial muscle devices. Theoretically, longer and thicker actuators give larger displacement and force, respectively, but it is practically impossible to use such actuators because of the voltage drop along the actuator devices³ and the mechanism of the electrochemical stretching of CP actuators, that is, the doping–dedoping of anions.^{1,4}

We proposed PPy–metal coil composite actuators (artificial muscle fibres)³ to solve the problems mentioned above. Electrically generated displacement and force of the PPy–metal coil composite can be easily tailored by adjusting the length and the number of the PPy–metal coil composites, respectively. We fabricated some bundles of PPy–metal coil composites and confirmed the electrochemical strain remained almost constant up to a 10-unit bundle, and the electrochemical force was proportional to the number of standard units in parallel to the bundles. A 1600-unit thick bundle of PPy–metal coil composite actuators, however, did not stretch effectively due to the poor configuration and the PPy deposited badly,³ and therefore the PPy–metal coil composite actuator is now being considered in use for micro-actuators, particularly for ‘push’ devices.

In this paper, we describe another PPy–metal composite film actuator for ‘artificial muscle cells’.⁵ This novel composite actuator is an advanced form of the PPy–metal coil composite actuator, and can be piled up to create huge force. We have been fabricating ‘artificial muscle cells’ by using the PPy–metal composite film actuator for practical artificial muscle devices.

Preparation of the PPy–zigzag metal wire composite film actuator was performed in a one compartment cell by a standard galvanostatic electropolymerization with a Hokuto Denko HA-151 potentiogalvanostat. PPy–metal composite films were obtained from a methyl benzoate solution of TBACF₃SO₃ (0.2 mol dm^{−3}) at 0.2 mA cm^{−2} at room

temperature for typically 4 h on a Ti or Au sputtered accordion-like zigzag metal wire structure on a polyethylene terephthalate (PET) film electrode.⁶ Then, the PPy deposited electrode on PET was immersed in an acetone bath to peel it off the PET. The sputtered metal layer was normally collapsed, and a PPy composite film with the zigzag metal wire was obtained. The PPy–zigzag metal wire composite film was immediately rinsed with a methyl benzoate solution of TBACF₃SO₃,⁷ followed by being set in the cell for another 2 h of electropolymerization to cover the metal wire side of the PPy composite film. This procedure was to improve the mechanical strength of the PPy–metal composite film. Finally, the PPy–metal wire composite film was rinsed with acetone and air-dried. Fig. 1 shows photographs of a part of a zigzag metal wire electrode on PET without a sputtered metal layer and a PPy–zigzag metal wire composite film. The zigzag metal wire electrode on PET shown in the photo was one of the configurations used for PPy–metal composite film actuators. The holes at the end were to screw up a number of PPy–metal composite film actuators, fabricating piled up thick actuators.

Electrochemomechanical deformation (ECMD) of a PPy–metal composite film actuator (length: 10 mm, width: 26 mm) was measured in a one-compartment three-electrode cell by

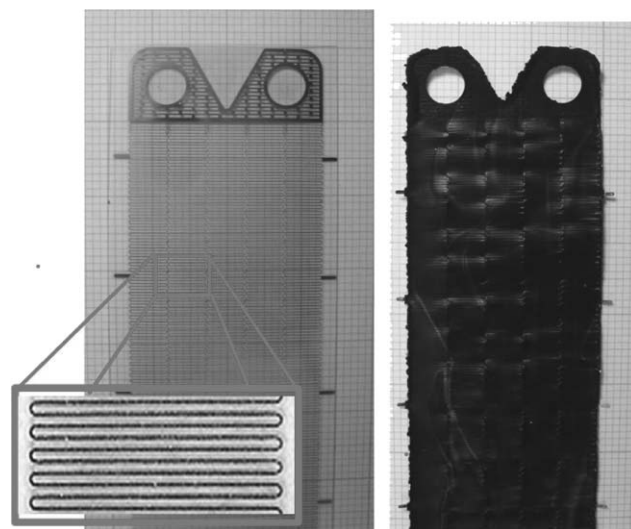


Fig. 1 Photographs of a part of zigzag metal wire electrode on PET (left) and a PPy–metal composite film (right).

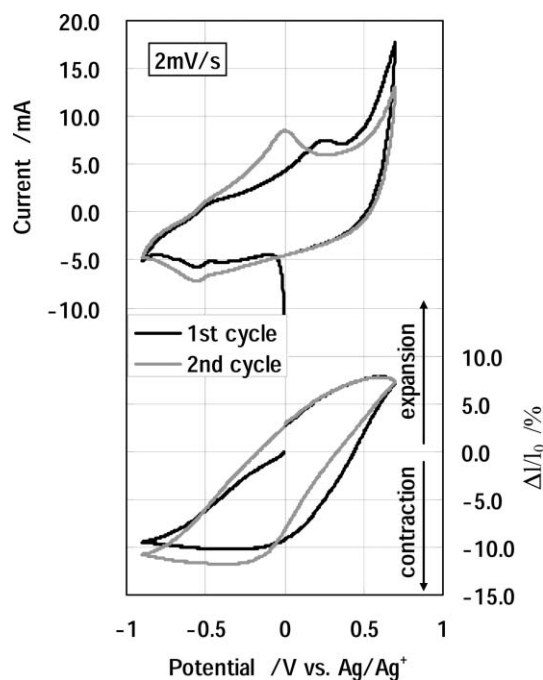


Fig. 2 A CV and an ECMD of a PPy-zigzag metal wire composite film actuator, cycled between -0.9 V and $+0.7$ V vs. Ag/Ag^+ at 2 mV s^{-1} in aqueous NaPF_6 solution.

using a KEYENCE LC-4000 laser displacement meter in aqueous NaPF_6 solution (1.0 mol dm^{-3}), driven by a Princeton Applied Research 263A potentiogalvanostat.^{1,8} Electrochemical stretching was also carried out by applying square pulses at ± 0.7 V vs. Ag/Ag^+ at 0.017 Hz by using a Hokuto Denko HA-151 potentiogalvanostat and a Wave Factory WF1946 multi-function synthesizer in aqueous NaPF_6 solution.

Fig. 2 depicts a cyclic voltammogram (CV) and an ECMD of a PPy-zigzag metal wire composite film actuator in aqueous NaPF_6 solution, cycled between -0.9 V and $+0.7$ V vs. Ag/Ag^+ at 2 mV s^{-1} at room temperature. On applying the potential toward -0.9 V, the PPy-metal composite actuator contracted by 10% with dedoping of CF_3SO_3^- . When the potential was swept anodically to $+0.7$ V, the PPy-metal composite elongated and the electrochemical strain for one redox cycle was 19.6%, much larger than that (12.8%)^{1,2} of the corresponding free-standing PPy film actuator, prepared similarly with $\text{TBACF}_3\text{SO}_3$ as the electrolyte on a Ti electrode, driven in aqueous NaPF_6 solution. The maximum electrochemical strain of the PPy-zigzag metal wire composite actuator was 21.4% when cycled at 0.2 mV s^{-1} . Note that further improvements in the electrochemical strain for PPy films doped with CF_3SO_3^- were not observed even when cycled at 0.2 mV s^{-1} .

The enhancement in the electrochemical strain of the PPy-zigzag metal wire composite film actuator was explained as follows. Although CP actuators expand and shrink three-dimensionally with doping-dedoping of anions, respectively, the deformation in the width direction was restricted by the zigzag metal wire structure and converted into the axial deformation in the PPy-zigzag metal wire composite actuator. Similar enhancements in the electrochemical strain were observed in PPy-metal coil composite actuators,³ and also proposed by De Rossi for a McKibben-like configuration.⁹

Fig. 3 shows electrochemical stretching of a PPy-zigzag metal wire composite film actuator (length: 90 mm, width: 26 mm) when square pulses were applied at ± 0.7 V vs. Ag/Ag^+ at 0.017 Hz. Potential was applied to the PPy-metal composite film actuator at 4 points (every 22.5 mm) in order to decrease the ohmic drop along the actuator. Large displacements

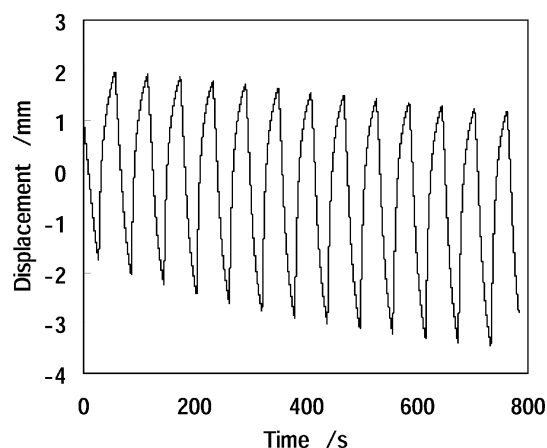


Fig. 3 Electrochemical stretching of a PPy-zigzag wire composite film actuator, operated at ± 0.7 V vs. Ag/Ag^+ at 0.017 Hz in aqueous NaPF_6 solution.

(4.1–4.7 mm) were observed for one cycle (60 s). Under moderate conditions like this (the electrochemical strain: ca. 5%),¹⁰ the repeated electrochemical stretching of PPy-metal composite actuator was very stable.

We have fabricated an encapsulated cell with a multi-layer PPy-zigzag metal wire composite film actuator (20 units) pulling up a 13 kg weight.¹¹ The encapsulated cell, ‘artificial muscle cell’, can create huge force using a number of the PPy-zigzag metal wire composite film actuators piled up. Large displacement was also achieved by using a connected PPy-metal composite actuator in series. The tailor-made artificial muscle cell can meet specifications for the applications of CP actuators such as robotics, powered suits, artificial limbs, control devices, toys, etc.

Notes and references

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- 6 The zigzag metal wire on PET, produced by Nippon Cable System Inc., was made of Inconel (Ni alloy). We named the accordion-like zigzag metal wire electrode on PET a ‘Jabara’ (snake-belly) plate. Without any sputtered layer of metal, the gap of the zigzag metal wire electrode was too wide to be covered completely with PPy, giving a limp metal wire with PPy which did not work as an actuator at all.
- 7 If a PPy-metal composite film was peeled off the PET and dried after the first 4 h of electropolymerization, the PPy-metal composite film would be considerably distorted and become difficult to be set in the cell for another 2 h of electropolymerization. As ketones such as acetone were found to be unsuitable solvents for the electropolymerization of pyrrole with $\text{TBACF}_3\text{SO}_3$ to obtain highly stretchable and powerful actuators, the PPy-metal composite film just after it is peeled off the PET in an acetone bath should be immersed in a methyl benzoate solution of $\text{TBACF}_3\text{SO}_3$ before being set in the cell to avoid contamination with acetone in the electrolytic solution for the electropolymerization.
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