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Effect of hexafluoropropylene on the performance of poly(vinylidene fluoride) polymer actuators based on single-walled carbon nanotube–ionic liquid gel

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ABSTRACT

The effects of polymer species (poly(vinylidene fluoride)) (PVdF) homopolymer or poly(vinylidene fluoride-*co*-hexafluoropropylene) (PVdF(HFP) copolymer), average molecular weight of the polymer, and the HFP content in PVdF(HFP) on the electrochemical and electromechanical properties of actuators using polymer-supported single-walled carbon nanotube (SWCNT)–ionic liquid (IL) gel electrodes were investigated. For the PVdF (Kynar 741 or 761) actuator containing 1-ethyl-3-methylimidazolium tetrafluoroborate (EMI[BF_4]), the generated strain was 0.90–1.05% for the frequency range of 0.01–0.005 Hz, which was over twice as large as that for the PVdF(HFP) (Kynar Flex 2801) actuator. Furthermore, it is considered that the HFP content should be low (or zero) for large generated strain and zero for large maximum stress. The PVdF actuator performs much better than the PVdF(HFP) actuator and has a quick response, sufficient for practical application (e.g., tactile displays).

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1. Introduction

Recently, much attention has been focused on soft materials that can directly transform electrical energy into mechanical work for a wide range of applications including robotics, tactile and optical displays, prosthetic devices, medical devices, and microelectromechanical systems [1]. Low-voltage electroactive polymer (EAP) actuators, which can function quickly and softly driven, are particularly useful, because they can be used as artificial muscle-like actuators for various bio-medical and human affinity applications [2,3]. We have previously reported the first dry actuator that can be fabricated using 'bucky-gel', a gelatinous room-temperature ionic liquid (IL) containing single-walled carbon nanotubes (SWCNTs) [4–7]. The actuator has a bimorph configuration with a polymer-supported IL electrolyte layer sandwiched by polymer-supported bucky-gel electrode layers that allow quick and long-lived operation in air at low applied voltages. ILs have low volatility and exhibit high ionic conductivities and wide potential windows, which are advantageous for quick response actuators and high electrochemical stability components [8].

We have previously reported the dependence of the IL species on the electromechanical and electrochemical properties of actuators composed of a PVdF(HFP) (Kynar Flex 2801)-supported bucky-gel electrode and the gel electrolyte layers [6,9,10]. The frequency dependence of the bucky-gel actuator displacement response was measured and it can be successfully simulated using an electrochemical kinetic model. Both the steric repulsion effects due to the transfer of ions to the electrode and the charge injection [11] results in the bending motion of the bucky-gel actuator. The generated strain of the polymer-supported bucky-gel electrode of the actuator is considered to be due to the volume change for the polymer–IL gel of the cathode and that of the anode [9]. In addition, we showed that the polymer–IL gel of the electrolyte is an important factor in the field of low-voltage electroactive polymer (EAP) actuators, and we consider that these results reveal an important design principle for low-voltage EAP actuators [12]. However, the performance of actuators using PVdF(HFP) (Kynar Flex 2801) and the EMI[BF_4] or EMI[TFSI] was not sufficient for practical applications (e.g., tactile displays).

Rechargeable Li batteries are ubiquitous energy device that is used in many types of portable electronic equipment. In the state-of-the-art technology of 4 V-class rechargeable Li batteries, a mixture of organic aprotic solvent and a conducting salt, lithium hexafluorophosphate ($LiPF_6$), is generally used as a non-aqueous

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Table 1
Young's modulus, average molecular weight and fraction for PVdF(HFP).

	Young's modulus[a] (MPa)	Chemical structure	Average molecular weight	Fraction
Kynar741	1660–2310	PVdF	269,000[b]	
Kynar761	1660–2310	PVdF	350,000[c]	
Kynar761A	1660–2310	PVdF	495,000[f]	
Kynar Flex2851	1100–1240	PVdF(HFP)	455,000[d]	$x = 0.95$, $y = 0.05$
KynarFlex2801	620–830	PYdF(HFP)	477,000[e]	$x = 0.88$, $y = 0.12$
KynarFlex2501	190–250	PVdF(HFP)	212,000[f]	$x = 0.83$, $y = 0.17$

[a] The data of young's modulus are quoted from Ref. [15].
[b–e] The data of average molecular weight are quoted from Refs. [16–19].
[f] The average molecular weight was determined by a gel permeation chromatography.

electrolyte. We are considering an electrode and electrolyte, such as an EAP actuator and electrochemical capacitor using a Li salt/IL, for a high-energy density device application. Moreover, much attention has been focused on polymer batteries using Li ion polymer gel electrolyte [13,14]. Li ion polymer batteries are commercially available mobile equipments (e.g., mobile phone).

In previous reports [4–6,9,10,12], we investigated only PVdF(HFP) (Kynar Flex 2851 or 2801) for application in the field of Li ion polymer batteries [14]; however, the effect of the polymer species (the PVdF homopolymer or the PVdF(HFP) copolymer), the average molecular weight of the polymer, and the HFP content in PVdF(HFP) on the performance of the polymer actuator was not investigated. It is considered that the affinity between PVdF and the IL is higher than the affinity between PVdF(HFP) and the IL because HFP is a perfluoro compound. Therefore, it is expected that an actuator containing PVdF will perform much better than an actuator containing PVdF(HFP).

In this paper, we investigated the effect of the polymer species (PVdF homopolymer or PVdF(HFP) copolymer), the average molecular weight of the polymer, and the HFP content of PVdF(HFP) on the electrochemical and electromechanical properties of an actuator with a polymer-supported SWCNT–IL gel electrode, i.e., the polymer-supported bucky-gel electrode.

2. Experimental

2.1. Materials

PVdF and PVdF(HFP) were used as received, and the results of characterization are shown in Table 1. The PVdF and PVdF(HFP) were used as received from Arkema Chemicals Inc. ILs, 1-ethlyl-3-methylimidazolium tetrafluoroborate (EMI[BF_4]; Fluka) and 1-ethlyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (EMI[TFSI]; Merck), were used as received and their chemical structures are shown in Fig. 1. SWCNTs were used as received (high-purity HiPco™ SWCNTs, Unidym Inc.). Other reagents, methyl pentanone (MP; Aldrich), propylene carbonate (PC; Aldrich), and dimethylacetamide (DMAc; Kishida Chemical Co. Ltd.) were used as received.

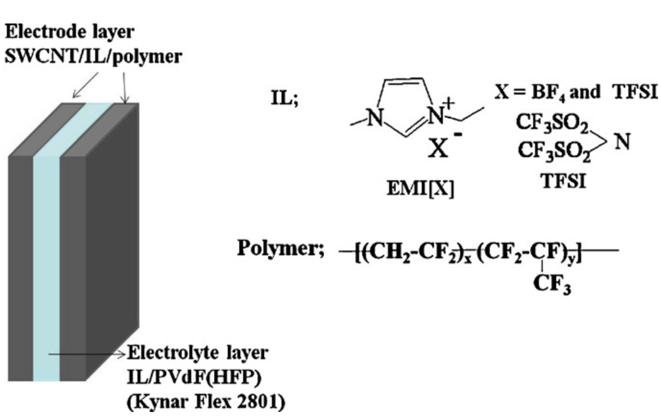

Fig. 1. Configuration of a polymer-supported SWCNT–IL gel actuator and molecular structure of the IL and polymer used.

2.2. Measurements

A fraction (x, y) of the PVdF(HFP) was determined by an elemental analysis. The elemental analysis (C, H and F) was carried out by the Center for Organic Elemental Microanalysis of Kyoto University.

2.3. Preparation of the actuator film [9]

The configuration of the bucky-gel actuator is illustrated in Fig. 1. The polymer-supported bucky-gel electrode layer was typically composed of 20 wt% SWCNT, 48 wt% EMI[BF_4] and 32 wt% polymer, and was prepared as followed. 50 mg of SWCNTs, 120 mg (0.6 mmol) of EMI[BF_4], and 80 mg of the polymer in 9 ml DMAc were dispersed using ultrasonic bath for more than 5 h. A gelatinous mixture composed of SWCNT, EMI[BF_4] and polymer in DMAc was then obtained. In the case of EMI[TFSI], the casting solution was obtained by mixing 0.6 mmol of EMI[TFSI] with the same amount of other components in 9 ml of DMAc. The electrode layer was fabricated by casting 1.6 ml of the electrode solution in a Teflon mold (an area of 2.5 cm × 2.5 cm) and evaporating the solvent. The solvents were then completely removed *in vacuo* at 80 °C. The thickness of the obtained electrode film was 70–80 μm. Gel electrolyte layers were fabricated by casting 0.3 ml of the solutions composed of each IL and the PVdF(HFP) (Kynar Flex 2801) (0.5 mmol/100 mg) in a mixed solution composed of 1 ml of MP and 250 mg of PC in a Teflon mold (an area of 2.5 cm × 2.5 cm) followed by solvent evaporation, and complete removal of the solvent *in vacuo* at 80 °C. The thickness of the obtained gel electrolyte film was 20–30 μm. The actuator film was fabricated by hot-pressing the electrode and electrolyte layers with the same IL. The typical thickness of the actuator film was 150–175 μm, which is smaller than the sum of those of two electrodes and one electrolyte layer, because the thickness of each layer decreases by being hot-pressing.

2.4. Displacement measurement

Fig. 2 shows schematic diagrams for the displacement measurement of the actuator and for evaluation of the strain from the displacement. The actuator experiments were conducted using an applied triangular voltage to a 10 mm × 1 mm actuator strip clipped by two gold disk electrodes. The displacement at a point 5 mm away (free length) from the fixed point was continuously monitored from one side of the actuator strip using a laser displacement meter (Keyence, LC2100/2220). A potentio/galvanostat (Hokuto Denko, HA-501G) and a waveform generator (Yokogawa Electric, FC 200) were used to activate the bucky-gel actuator. The electrical parameters were simultaneously measured. The measured displacement δ was transformed into the strain difference between two bucky

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