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SELF-DEFORMABLE FLEXIBLE MEMS TWEEZER MADE OF POLY (VINYLIDENE FLUORIDE) / IONIC LIQUID GEL WITH ELECTRICAL MEASUREMENT CAPABILITY

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ABSTRACT

We demonstrated a self-bending flexible tweezer capable of simultaneous mechanical handling and electrical measurement. A soft cantilever of 2mm × 8mm × 75μm bent by itself, and electrical signal observation through touched surface was successful. The working voltage (1.5V) was the lowest, and normalized strain was one of the highest values as compared to world's similar devices, to our best knowledge. Unique fabrication process of flexible cantilever with novel ionic polymer-metal composites (IPMCs) was developed. The material was poly (vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE) and ionic liquid (IL) by use of acetone solvent, and silver nanowire for electrodes. The IL of 50wt% gave the largest 7mm bending displacement at +1.5V_{DC}. The cantilever with the above actuator touched on a gold test surface, then electrical observation of alternating voltage of 700mV (1 kHz and 4 kHz) was confirmed.

KEYWORDS

Self-deformable cantilever, flexible actuator, soft gripping, electrical observation, PVDF-TrFE, ionic liquid, silver nanowire

INTRODUCTION

To study living organisms, it is necessary to stimulate it and observe the response. So far, in some research a part of organism has been locally stimulated by chemical substances or observed electrically [1, 2]. The devices in these studies had however some problems, such as tissue damage and lack of adhesion due to planar nature [1]. To solve these problems, we propose a flexible MEMS device. If we can realize a tweezer by a self-deformable cantilever (Fig. 1), the device will be able to observe a target sending electrical signal while gently gripping the target.

To realize such cantilever, we propose to employ an Ionic Polymer-Metal Composites (IPMCs) actuator. IPMCs actuators attract attentions with their flexibility, large strain, good conductivity and low operation voltage [3]. An IPMC is composed of polymer and mobile ions with electrodes. When voltage is applied, cations and anions move to opposite direction [3]. The actuator bends to either side of electrodes because of the size difference of the mobile anions and cations. As the polymer, Nafion has commonly been used up to present [3]. However, recent studies have often applied poly (vinylidene fluoride) (PVDF) [4, 5] and

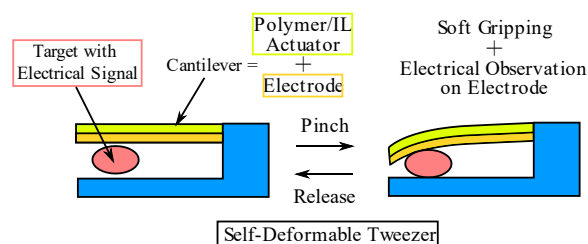


Figure 1. Self-deformable tweezer enables mechanical gripping and electrical observation simultaneously.

its co-polymer [6, 7, 8, 9] to IPMCs. PVDF is known as a ferroelectrics and piezoelectric material that has as low Young's modulus (~500 MPa) as that of Nafion [8]. PVDF in IPMCs is occasionally used as co-polymer structure such as poly (vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP [6, 7, 8]), poly (vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE [8]), and poly (vinylidene fluoride-co-chlorotrifluoroethylene) (PVDF-CTFE [8, 9]) with ionic liquid (IL) as fluid ions. IL is salt that exists as fluid stably at wide ranged temperature, having low vapor pressure, incombustibility and good conductivity [4]. Above these PVDF co-polymers with IL, the maximum displacement was 3.5 mm of for a 10 mm length actuator at 100 mHz 5.0 V_{pp} made of PVDF-TrFE including 1-ethyl-3-methylimidazolium bis (trifluoromethanesulfonyl) imide (EMIMTFSI) as IL with gold sputtered electrode [8].

As discussed above, IPMCs actuator composed of PVDF co-polymers exhibit big movements at low voltage and is suitable for the MEMS soft tweezer handling delicate materials, such as organic tissues. Though a lot of research has investigated a variety of actuator with electrode [4-8], little has applied such actuators to some practical application [3]. Based on above consideration, we fabricated PVDF-TrFE/EMIMTFSI composites actuator with silver nano wire electrodes for MEMS tweezer (Fig. 1). The silver nanowire of the electrodes is an assembly of silver wires that are nm scales, expecting higher flexibility and conductivity than silver, gold and platinum in sheet form electrodes. The combination of IPMCs of these polymer, IL, electrodes, and solvent is unique in the world, to our best knowledge. The fabrication process of this PVDF-TrFE/IL composites was established by use of the acetone solvent, with the silver nanowire coated as the electrodes. The mechanical characteristics of the actuator with such electrodes was verified whether it had the appropriate performance to be adopted to the Fig. 1

tweezer. Finally, a tweezer with a cantilever of the actuator was confirmed to observe the electrical signal that is sinusoidal wave voltage, on a gold target electrode.

FABRICATION OF IPMCs TWEEZER

Materials

Ionic liquid was EMIMTFSI manufactured by TCI, Tokyo Chemical Industry (Japan,) whose purity is 98.0%. It was stored and used in glove box under nitrogen atmosphere in order not to absorb moisture or carbon dioxide. It was heated up to 110 °C in 5 minutes on hotplate to evaporate moisture when used. As a polymer, we used PVDF-TrFE, PIEZOTECH® FC25 produced from Arkema Piezotech (France.) It contains 25mol% trifluoroethylene (TrFE.) The solvent was acetone (99.5% purity,) obtained from Kanto Chemical Holdings (Japan.) All the above substances were stored at 25 °C, atmospheric pressure. The electrode was silver nanowire that was made in Merck (Germany.) The concentration was 5mg/mL in water. It was kept at about 2 to 6 °C, under atmospheric pressure.

Fabrication process of PVDF-TrFE/Ionic liquid membranes

First, we fabricated three types EMIMTFSI/PVDF-TrFE composites, in which mass proportion of EMIMTFSI was 30, 50, 70wt%. From now on, this polymer/IL composites are called as “ionic gel actuator.” The weight of PVDF-TrFE was either 150mg or 300mg, hence 6 actuators membranes were made in total. These actuators were named as 30a, 30b, 50a, 50b, 70a, 70b. ‘30’ and ‘50’ refers to weight proportion of EMIMTFSI, and ‘a’ and ‘b’ refer to 150mg and 300mg of PVDF-TrFE, respectively.

The process flow was as follows. Appropriate amount of PVDF-TrFE was put in glass bottle and acetone was added to dissolve solute to 5wt%. The solution was stirred on 50 °C hotplate for more than 24 hours. EMIMTFSI was added and dissolved in the previous solution. This liquid was also stirred at 25 °C (room temperature) for more than 24 hours. After the agitation, the solution was spread into a Petri dish of size $\phi 80 \times 17$ mm and dried covered with a lid at 25 °C for more than 24 hours to form film by evaporating acetone. All the process was conducted in the glove box.

As a result, among six membranes, four (30b, 50a, 50b, 70b) was obtained and two (30a, 70a) was broken because the two membranes were too thin to be peeled off from the Petri dish. The next process of coating electrodes was applied to these four membranes.

Coating of silver nanowire electrodes

The membranes were cut about 20×20mm squares before coating. They were put in a Petri dish and coated with silver nano wire of water solvent by a syringe. In order to evaporate solvent completely, the dish was heated on hot plate at 110 °C for 8 minutes. The membrane was cooled down for 5 minutes at 25 °C (room temperature.) This process was done to the other side of the membrane. The film was peeled off and cut into 2×10mm square by UV laser cutter (LPKF ProtoLaser U4).

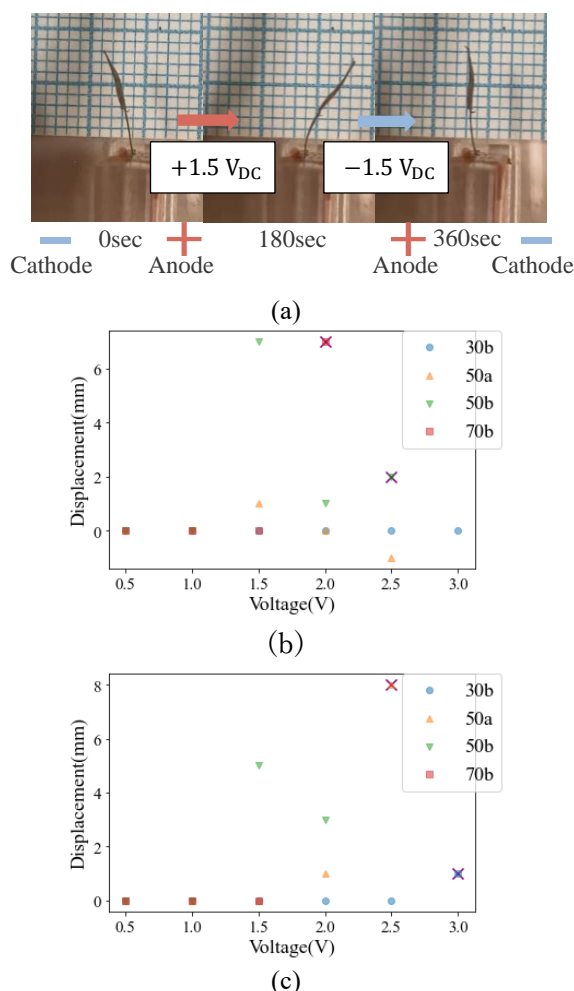


Figure 2. Measurement of mechanical features. (a): Bidirectional bending motion of 50b at $\pm 1.5V_{DC}$. (b, c): Displacements of four actuators at 0.5 to 3.0V_{DC}, (b) At positive voltage, (c) at negative voltage.

Thickness of obtained membranes with or without electrodes were observed by laser microscope OLYMPUS LEXT OLS5000. Thickness of the membranes 30b, 50a, 50b, 70b after electrodes coated was 52.6, 65.6, 75.3, 87.7 μ m, respectively. This result shows that the more IL weight ratio was, the thicker the membranes were. The ionic gel contained IL and swelled as the weight ratio of IL. The thickness of electrodes layers was measured to vary from 0.2 to 11.2 μ m.

RESULTS AND DISCUSSIONS

Mechanical Characteristics of Ionic Gel Actuator

The 2×2mm part of the 2×10mm IPMC actuator was fixed between copper tape electrodes. With DC supply Hewlett Packard E3640A, the DC voltage was applied for 3 minutes, and more 3 minutes in the opposite direction, from 0.5V to 3.0V in 0.5V increments. The displacement was read in 1mm unit using a graph sheet.

As shown in Fig. 2 (a), the actuator always bended to the anode side (positively charged electrode) because the cation was bigger than the anion in EMIMTFSI. Fig. 2 (b) and (c) are displacements obtained in 3 minutes after the voltage was applied, in the forward (b) or reverse (c)

Table 1: Performance Comparison

Ref.	Material	D [μm]	V_{DC} [V]	ϵ [%]	FoM: ϵ/DV_{DC} [ppm/ $\mu\text{m}\cdot\text{V}$]
This work	PVDF-TrFE	75.3	1.5	0.93	82.3
[3]	Nafion	300	5	0.67	4.5
[4]	PVDF	75	10	0.53	7.1
[5]	PVDF	150-175	2	1.05	34.9 - 29.9
[6]	PVDF-HFP	280	3.5	2.24	22.9
[7]	PVDF-HFP	106-162	2	2.0	94.2 - 61.6
[8]	PVDF-TrFE	50	5	0.49	19.8
[9]	PVDF-CTFE	31-33	5	1.35	82.1-87.4

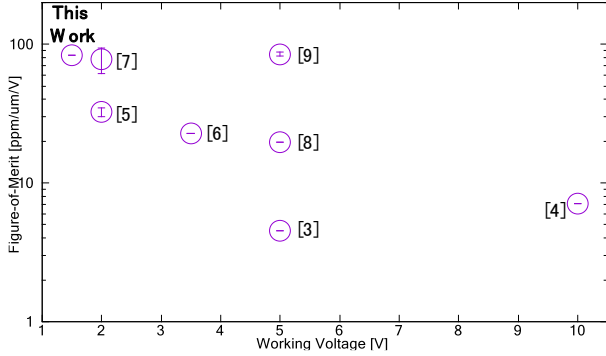


Figure 3: Figure-of-Merit and Working Voltage

directions, respectively. Four cantilevers made at certain conditions became conductor during measurement, supposedly, the electrical double layer is destroyed. The current rapidly increased and aborted at 3A, limit current of the used DC supply. Such case is also plotted with cross marks on the Figs. 2 (b) and (c), 50a and 50b had significant displacement, though 30b and 70b did not.

State-of-the-art Performance Comparison

The largest bending was obtained by of 50b actuator at 1.5 V_{DC} . The values were 7 mm in the forward direction and 5 mm in reverse direction. This maximum displacement of 7 mm at 1.5 V was compared with the ones of previous research works that employ Nafion [3] and PVDF composites [4-9]. The following equation (1) was used to evaluate the mechanical performance of actuators:

$$\epsilon = \frac{2D\delta}{L^2 + \delta^2} \left(= \frac{D}{R} \right), \quad (1)$$

where ϵ , D , δ , L , R is normalized strain, thickness, displacement, free length of actuator, and curvature radius, respectively. Because ϵ is in proportion to D and δ ($\delta > \sqrt{L}$) and supposed to follow applied voltage V_{DC} , we can define figure-of-merit (FoM) as ϵ/DV_{DC} . The values are summarized in Fig. 3. By comparing FoM plotted in Fig. 3, we can conclude that our developed actuator realized the lowest working voltage V_{DC} and high FoM comparable to the state-of-the arts [7] [9].

Demonstration of Tweezer as Electrical Probe

We have made a demonstration of our self-deformable cantilever actuator as an electrical probe. The cantilever was held by a jig composed of printed circuit. The jig had

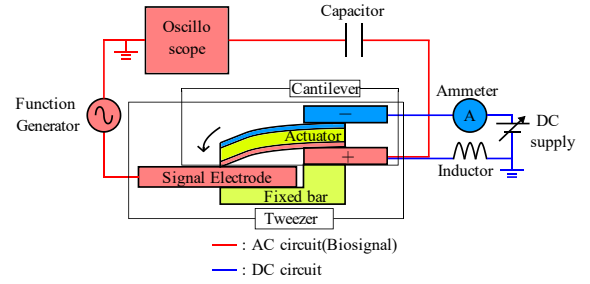


Figure 4. Schematic of the tweezer circuit

Table 2. v_{AC} observed at 1kHz and 4kHz.

Frequency	Contact voltage	Released voltage
1kHz	43mV	105mV
4kHz	36mV	60mV

two electrodes, one fixed bar, a gold signal electrode. In Fig. 4, blue circuit was direct current (V_{DC}) for cantilever operation, and red circuit was alternating current (v_{AC}) mimicking a biosignal. They were electrically separated by using capacitor and inductor (known as Bias-T). When V_{DC} was applied, the cantilever moved to and touched the gold signal electrode. After the red circuit was closed by actuation of IPMCs cantilever, the AC signal could be observed at function generator.

The cantilever repeated contact and release to the signal electrode for 5 times in a row. In Fig. 4, the function generator, ammeter, DC supply was respectively RIGOL DG4202, the ammeter mode of HIOKI DT4253, 1.5V alkaline battery. The v_{AC} was 700 mV of 1 or 4 kHz. The 10pF ceramic capacitor and 47mH lead inductor were used. The oscilloscope GW instek GDS-2072A was used as Math mode, which could pick up the wave of target frequency in the observed voltage wave by Fast Fourier Transformation. The v_{AC} in contact and release, movement of the actuator and the transition of current was recorded by video camera SONY HDR-PJ680.

As shown in Table 2, in both 1 and 4kHz frequency the voltage when the actuator touched the signal electrodes was more than 1.5 times as large as when the actuator was open. Therefore, the PVDF-TrFE/EMIMTFSI gel composites actuator with silver nanowire electrodes was able to observe the sine wave voltage imitating biosignal. Figs. 5 (a), (b), (c) show the movement, v_{AC} of the actuator and current. When the actuator touched the signal electrodes like middle of Fig. 5 (a), v_{AC} increased abruptly at a purple point Fig. 5 (b). After 40 seconds of voltage application, the DC current stayed constant at 3 μA (Fig. 5 (c)). The v_{AC} stayed around 100mV, when the actuator and the signal electrode are thoroughly conducted.

Estimation of Equivalent Circuit of Tweezer

Based on the measurement we have estimated an equivalent circuit as shown in Fig. 6. A DC circuit of the actuator of bending operation is drawn with blue lines. The input resistance of ammeter is 1M Ω . R_1 is a resistor which represents the leak current of the actuator, R_2 is triggered by ionic conduction of IL movement, and C_1 is electric double layer capacitor of IPMCs. These

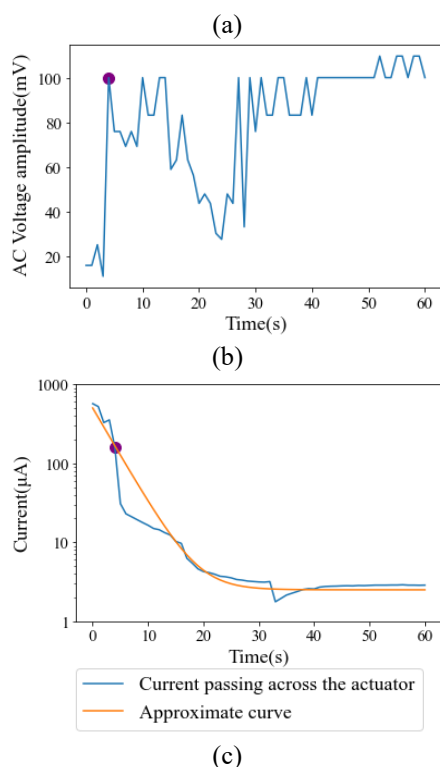
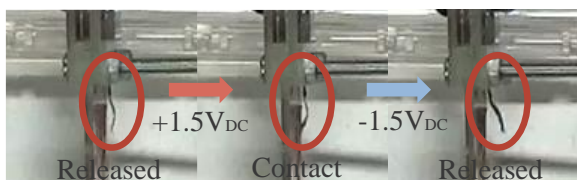


Figure 5. Characteristics of the tweezer. (a): movement of tweezer, (b): v_{AC} observed, at third time operation. (c): current when $+1.5V_{DC}$ was applied.

parameters were determined with the current of Fig. 5 (c), the inductance ignored because the buffering current of capacitance was much larger than that of inductance. By estimation from the data, $R_1 = 6 M\Omega$, $R_2 = 2 k\Omega$ and $C_1 = 1.2mF$, from approximate curve in Fig. 5 (c). The PVDF-TrFE/EMIMTFSI membrane has a huge insulation resistance to DC and some ionic conductive resistance to AC. Moreover, it was shown that ionic gel actuator has very large capacitance, generally called “supercapacitor.”

The equivalent circuit for AC signal was a red circuit in Fig. 6. R_c is a series resistance, in which a resistance between copper electrodes of the jig and silver nanowire electrodes of the actuator, and a resistance of the silver nanowire electrodes. R_p and C_p are each contact resistance and capacitance between signal gold electrodes and actuator electrodes. When the actuator was open R_p was infinite. The estimated values from table were $(R_c, R_p, C_p) = (500k\Omega, 1k\Omega, 1 \text{ to } 100pF)$. This result explains that R_c was the dominant factor for the signal decay from 700mV to 100mV.

CONCLUSIONS

Towards biological MEMS devices that manipulate and observe biological tissues, we could fabricate new PVDF-TrFE /EMIMTFSI composites gel actuators with the silver nanowire electrodes by the acetone solvent.

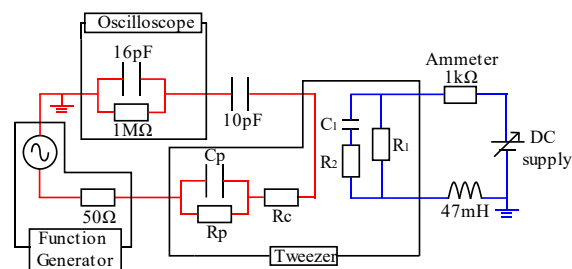


Figure 6. DC (blue) and AC (red) equivalent circuits

The actuator of 50wt% EMIMTFSI had the largest 7mm bending at $1.5V_{DC}$. The actuator was applied to the cantilever of the tweezer, which could observe 700mV, 1 or 4kHz sine wave signal, first in the world. The circuit elements of the tweezer were also estimated, which indicated that this actuator had huge capacitance and unexpected large resistance of electrodes. The authors will examine remaining issues for demonstration with cells with better S/N ratio: actuator reliability improvement, resistivity decrease of nanowire electrode, dedicated signal wiring introduction.

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