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Printing Fabrication of a Bucky Gel Actuator/Sensor and Its Application to Three-Dimensional Patterned Devices

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Abstract

A bucky gel actuator is a novel electro-active polymer (EAP), which is a low-voltage-driven dry soft actuator. In addition, the bucky gel device generates electromotive force when bending and then it can also be used as a sensor. Its device has a bimorph structure with polymer-supported bucky gel electrodes and a polymer-supported ionic gel electrolyte. It can be fabricated by layer-by-layer casting to easily form any shape. In this paper, we demonstrate an automatic fabrication method of bucky gel devices based on a printing method. By using a dispensing machine, we construct the printing system and the manual forming process is replaced with automatic printing. We investigate the printing of complicated shapes and three-dimensional electrode patterns through experiments.

Keywords

Electro-active polymer, artificial muscle actuator, soft actuator, sensor, printing fabrication

1. Introduction

1.1. Electro-Active Polymers

Electro-active polymers (EAPs) [1, 2] are functional polymeric materials that respond to electrical stimulus with a shape change. EAP actuators have potential

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capabilities such as flexibility, lightweight, large deformation and quick response, and have attracted the attention from engineers and researchers in many disciplines, e.g., robotics, medical service and the toy industry. EAP actuators are sometimes called artificial muscles due to their biotic smooth motions, and have been expected to be used in practical application because of responsiveness and facility of control. There are many types of EAPs, and various research has been conducted actively from the viewpoint of activation mechanism, response, formability, etc.

An ionic polymer–metal composite (IPMC) [3] is one of the most promising EAP actuators for application, and has good properties of response and durability. Commercialized IPMC products have already been developed [4]. IPMC is produced by chemically plating gold or platinum on both surfaces of a perfluorosulfonic acid membrane, which is known as an ion-exchange membrane. However, the procedure of the chemical plating is somewhat complicated. Arbitrarily shaped actuator devices can be manufactured by shape forming of the polymer material [5, 6] or laser beam machining of electrode patterns [7]; however, there is a limitation to productivity.

From the viewpoint of miniaturization or efficient production, it is expected to develop other devices that can be fabricated by simpler processes such as printing methods. Printable electrical devices [8, 9] are attractive for the development of microelectromechanical systems (MEMS), i.e., integrated micro devices of mechanical parts, sensors, electrical circuits and actuators. In particular, printable actuators would be necessary for miniaturized mechanical devices.

Another problem with IPMC is its activation only in wet conditions. When IPMC films dried, ion conductivity vanished and IPMC could not be moved. In order to operate in air over a long period, the step of lamination is needed. It was also shown that IPMC can be operated in air by using ionic liquid as solvent [10].

1.2. Novel EAP

Recently, Fukushima *et al.* developed a novel EAP device that resolved the above problems [11]. It is a bucky gel actuator, which consists of carbon nanotubes, ionic liquids and polymers. It has a simple bimorph structure and has important advantages:

- It can be operated under dry conditions.
- It can be fabricated simply by layer-by-layer casting.
- It has good response in low applied voltages.

The bucky gel actuators are effective for miniature mechanical devices. In our previous work, it was observed that the bucky gel device also can be used as a sensor, as the bucky gel film generates electromotive force when it is deformed [12]. Since sensor and actuator functions exist in the same device, both functions can be integrated, keeping the advantages of functional polymeric materials. It can be used as a 'flexible' sensor device, and sensor–actuator systems and feedback control systems can be constructed easily; therefore, the number of possibile applications can be expanded.

The bucky gel device can be fabricated simply by the manual solution casting method; however, it is difficult to fabricate uniform films or complicated shapes. In this study, we aim to explore the possibility and the excellent formability of the bucky gel actuator/sensor, and we demonstrate an automatic fabrication method of bucky gel devices based on a printing method. In order to realize an arbitrarily shaped bucky gel device with high accuracy, the manual forming process is replaced with an automatic printing process. We construct a printing system by using a dispensing machine, and we test the printing of the bucky gel devices with complicated shapes and three-dimensional (3-D) electrode patterns.

In Section 2, we will explain the configuration of the bucky gel actuator/sensor device and the normal fabrication method. In Section 3, the printing method and verification results are demonstrated. In Section 4, we apply the proposed printing method to 3-D patterning of the electrodes. Finally, we conclude the paper in Section 5.

2. Bucky Gel Device

2.1. Structure and Responsive Property

The bucky gel actuator [11] is a novel low-voltage driven dry soft actuator composed of carbon nanotubes, ionic liquids and polymers. Figure 1 shows the basic configuration and bending behavior of bucky gel actuators. It has a simple three-layered configuration; the middle layer is an electrolyte that is composed of polymers and ionic liquids, and the outside layers are electrodes that are composed of

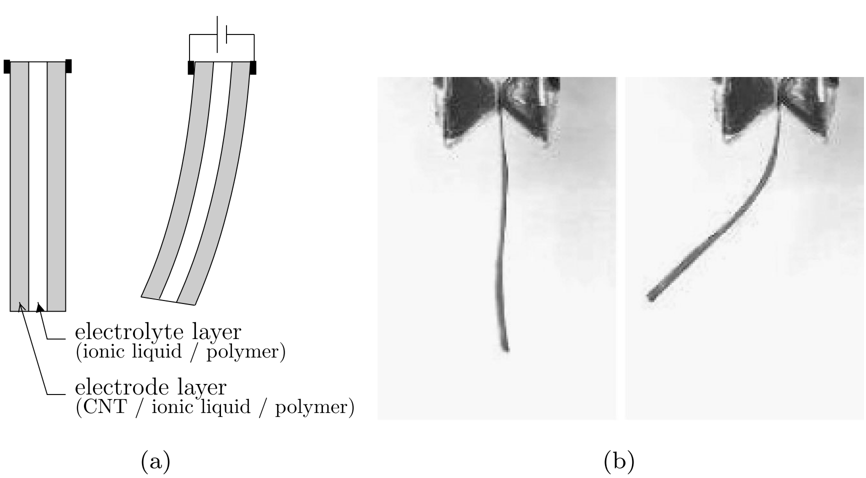

Figure 1. Bending motion of a bucky gel actuator: (a) schematic diagram and (b) bending response.

carbon nanotubes, ionic liquids and polymers. In the electrode layers, the carbon nanotubes are dispersed into the electrolyte.

Ionic liquids are liquid salts at room temperature, and have high conductivity, non-volatility and a wide potential window. These factors are advantageous for ion-conductive polymer actuators in terms of rapid response and high electrochemical stability. By using ionic liquids as the electrolyte, bucky gel actuators can be used in air for a long period without any loss of performance.

Carbon nanotubes are allotropes of carbon, which are 1-atom-thick sheets of graphite rolled up into a seamless cylinder with a diameter of the order of a nanometer. They have unique electrical and mechanical properties, and are efficient conductors of heat. Although it was not easy to process carbon nanotubes adequately for any purpose, Fukushima *et al.* discovered a processing method. By grinding single-walled carbon nanotubes in ionic liquids, carbon nanotubes are dispersed and blended materials turn into gels [13]. It was named 'bucky gel', which is soft and performs as an electrode material. It can be formed into any shape easily. By using polymer-supported bucky gels as electrodes, the actuator device was developed.

Figure 2 shows the experimental result of the bending response. In this test, a strip of the actuator was fixed in a cantilever and an input voltage of ±2 V with a frequency of 0.2 Hz was applied. The displacement at a point near the tip was measured by a laser displacement meter. The bucky gel film bends smoothly in response to low applied voltages. The bending direction is to the positive pole side. When an input voltage is applied on both surfaces of the electrodes, cations and anions of ionic liquids gravitate towards opposite poles. Then, in the electrode layers, electric double layers are formed at the interface between carbon nanotubes and the electrolyte, and the electrode layers stretch by electrostatic force. It is considered that the stretch of the electrode occurs on both sides; however, the actuator film bends to the positive pole side due to the difference in ion size. Although details of the bending mechanism are now under consideration, the main factor is considered as above. The bucky gel actuator is long-lived upon operation in air. In Ref. [11], it was reported that the actuator could be activated periodically over 2 months without any loss of performance. The bucky gel actuators are now in the development phase and research for improving the characteristics is ongoing [14, 15].

In our previous works, we observed that the bucky gel film generates an electromotive signal when it is deformed, as shown in Fig. 3. The system configuration for the measurement is very simple and the same as the case of the actuator. The surfaces of the electrode layers are connected to a measuring instrument and the voltage or current can be measured as sensor output signals. In the case of voltage sensing, the output signals are as small as about 0.1 mV; however, it is considered that the bucky gel device can be used as a sensor [12, 16]. We conducted some experiments to verify the possibility of the bucky gel actuator/sensor. Feedback control of the actuator and the sensor system for estimation of deformation was

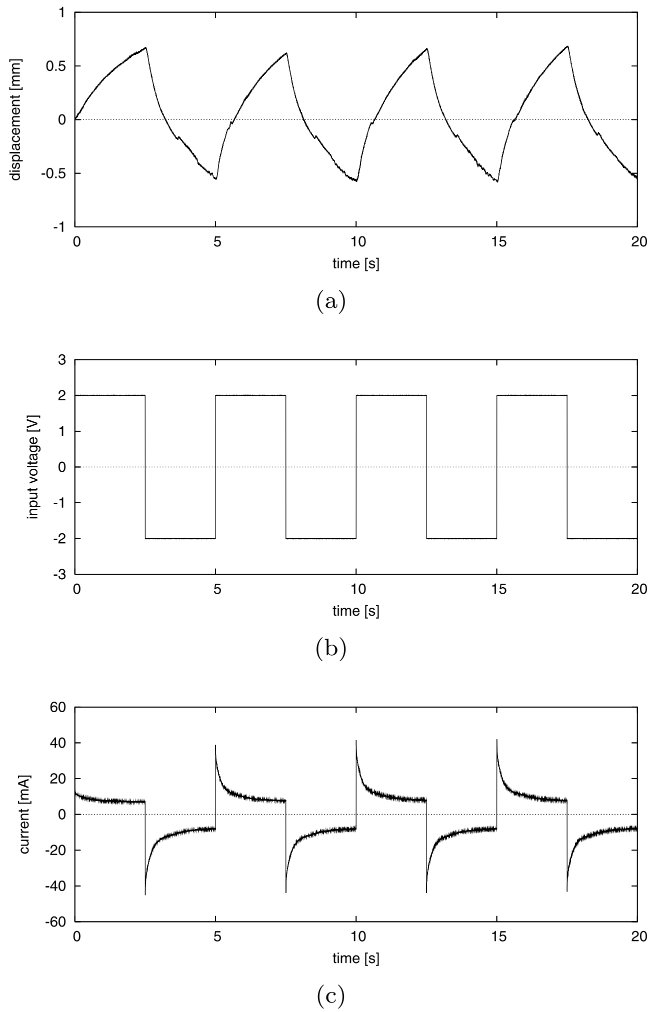

Figure 2. Bending response of a bucky gel actuator in response to square wave voltage: (a) displacement, (b) input voltage and (c) current.

demonstrated. In addition, by using both functions, feedback control based on the sensor signal was realized.

2.2. *Fabrication Method by Solution Casting*

Figure 4 shows the usual fabrication procedure of bucky gel devices. The devices can be fabricated easily by solution casting. First, each material for the electrode and electrolyte was dispersed into solvents, and their mixtures are put into an ultrasonic bath to make gelatinous mixtures. Next, the film was fabricated through layer-by-layer casting of gelatinous mixtures. Finally, the films were dried fully to

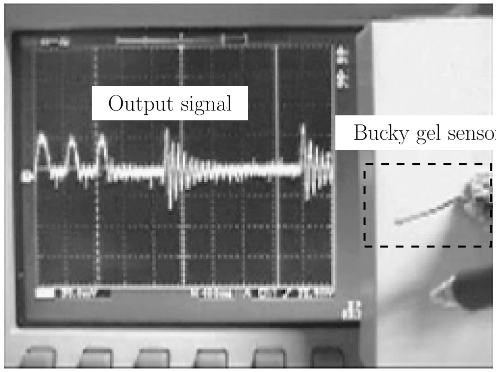

Figure 3. Output response of the bucky gel sensor.

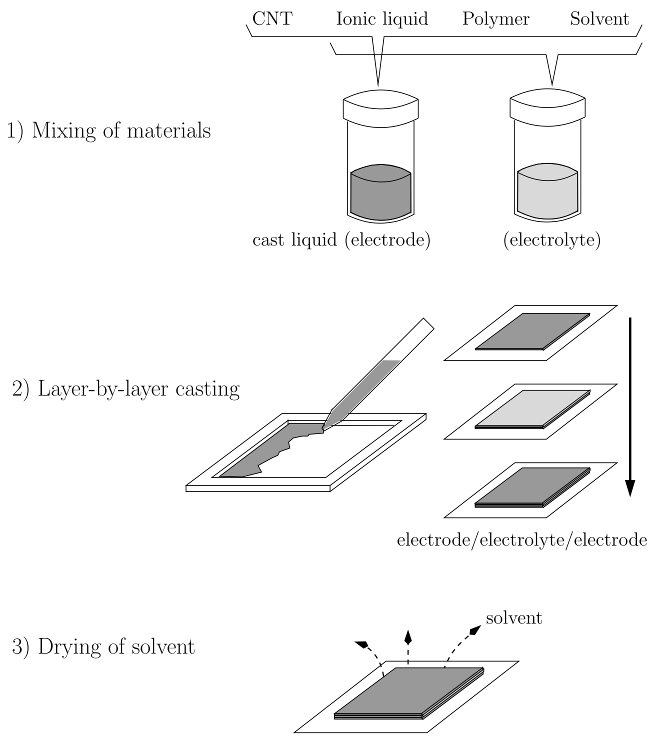

Figure 4. Fabrication method by solution casting.

remove the solvents. The devices also can be fabricated by hot-pressing the electrode and electrolyte layers that were fabricated by casting.

In the previous works, we fabricated the test sample of the bucky gel device manually based on the usual fabrication procedure. In the layer-by-layer casting procedure, we used a micro-pipette to cast the gelatinous mixture with a fixed quantity. We used a suitable form of a larger size for casting and we cut the dried sheet to an appropriate size. The fabrication procedure is very simple and the devices exhibit excellent formability; however, there exist several limitations in manual-handing procedures. It is difficult to fabricate the devices with accurate size, uniform thickness and homogeneous characteristics, and also difficult to construct complicated shapes and smaller patterns. In order to solve these problems, we apply the printing technique in place of manually casting and realize an automatic fabrication process.

3. Fabrication Method by Printing

3.1. Printing System

In order to realize the printing of the bucky gel devices, we use a dispenser machine that can discharge the materials with a fixed quantity. The mixing process of the materials and the drying process are the same as the normal procedures as in Fig. 4. The forming process is replaced with an automatic printing process.

Figure 5 shows the printing system we used in this study. The system consists of a dispenser, a three-axis positioning stage and a temperature control unit, produced by Musashi Engineering. The specifications of the printing system are summarized in Table 1. We selected a pneumatic discharge-type dispenser, and the dispenser unit consists of a syringe and the controller that regulates the discharge pressure and timing. The 3-d.o.f. positioning stage consists of a stage that can move in an anthropometry direction and a manipulator that can move the syringe widthwise and heightwise. Each axis is controlled simultaneously. The temperature control unit is connected to the heater, which is attached to the syringe and can regulate the temperature of the material.

The printing patterns are edited by using CAD software on the computer, and the traveling patterns of the 3-d.o.f. stage and dispensing intervals are described. The programming data are uploaded to the dispensing system and executed.

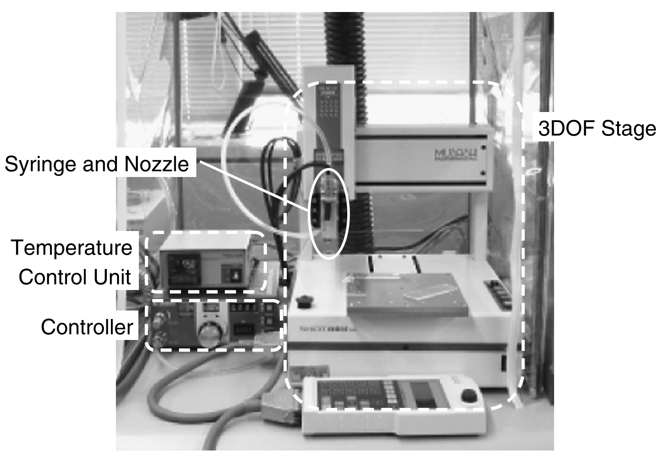

Figure 5. Printing system.

Table 1.
Specifications of the printing system

Dispenser	
control method	electronic/pneumatic system
dispensing pressure range	0.005–0.700 MPa
dispensing time range	0.010–9.999 s
vacuum pressure	0 to −20.0 kPa
Three-axis robot	
working range	200 × 200 mm
speed (X, Y axis)	0.1–500 mm/s
speed (Z axis)	1–200 mm/s
repeatability	$< \pm 0.01$ mm
resolution	0.0125 mm
Temperature control unit	
adjustable range	30–100°C
temperature precision	±0.5°C

3.2. Printing Tests

In order to investigate the proposed method, we conducted an experimental trial for printing of the bucky gel device. It is important to control the discharge conditions of materials for high-precision printing. The conditions are affected by many factors, such as the viscosity of the materials, discharge pressure and surface resistance of the nozzle. In this study, we adjusted the printing conditions through trial and error. The procedures of the adjustment are:

(i) Adjust the viscosity of the materials in the mixing process.

(ii) Select the nozzle diameter.

(iii) Adjust the discharge pressure and the speed of printing based on repeated trials.

In the first step, the viscosity of the materials depends on the ratio of the mixture, especially the quantity of the solvent. The temperature of the materials also affects the viscosity; however, the temperature should be kept fixed because of the temperature-depended properties of the polymer. If the temperature of the materials is lower than the appropriate value, then the polymer coagulates and the nozzle becomes clogged. Therefore, in the experiments, we controlled the viscosity by adjusting the quantity of the solvent. In general, it is convenient for easy adjustment that the viscosity and the discharge pressure are high. In our case, we should mix the materials fully and should keep the gel state, and then the quantity of the solvent was adjusted to a lower amount in comparison with the normal solution casting.

The test samples were made from single-walled carbon nanotubes (HiPco®; CNI), imidazolium ion-based ionic liquids and poly(vinylidene fluoride-*co*-hexafluoropropylene) (PVdF(HFP)) as a polymer, and 1-ethyl-3-methyl-imidazolium tetrafluoroborate ($EMIBF_4$) as ionic liquid. Methyl pentanone (MP) and propylene carbonate (PC) were used as solvents. For the gel solutions of the electrode layer, a mixture of 50 mg carbon nanotubes, 80 mg PVDF(HFP) and 120 mg $EMIBF_4$ in 240 mg PC and 4 ml MP was dispersed in the ultrasonic bath to form a gel solution, and kept in a hot stirrer for about 24 h. The solution for the electrolyte layer was obtained by mixing 200 mg PVDF(HFP), 200 mg $EMIBF_4$, 360 mg PC and 2 ml MP. After preparing these solutions, each solution was set into syringes and placed in the dispensing machine. Then the printing program for each layer was executed. As previously mentioned, the adjustments of the printing condition are very important. In the tests of the paper, we selected a nozzle of 100 μm diameter. The printing speed was 50 mm/s, and the distance between the nozzle and target plate was 300 μm. The discharge pressure was adjusted from 0.005 to 0.010 MPa by repeating the trials. For the electrode layers it is necessary to repeat the multiple printings to achieve sufficient thickness. After printing, solvents in the printed films were perfectly evaporated.

Figure 6 shows the results of the printing tests, where the materials for the electrode were printed on a glass plate. Figure 6a shows the printing result of a straight line. From the result, it can be seen that the width of the printed line is uniform. Figure 6b shows the printing result of a strip of film, where five straight lines of 20 mm length at 0.5-mm intervals were printed twice. It was confirmed that arbitrarily-sized films can be fabricated by repeating the printing of thin lines. Furthermore, the thickness of the film can be adjusted by overprinting little by little.

It was also confirmed that the electrolyte layer can be printed by the same procedures. Thus, the bucky gel devices can be fabricated by layer-by-layer printing of

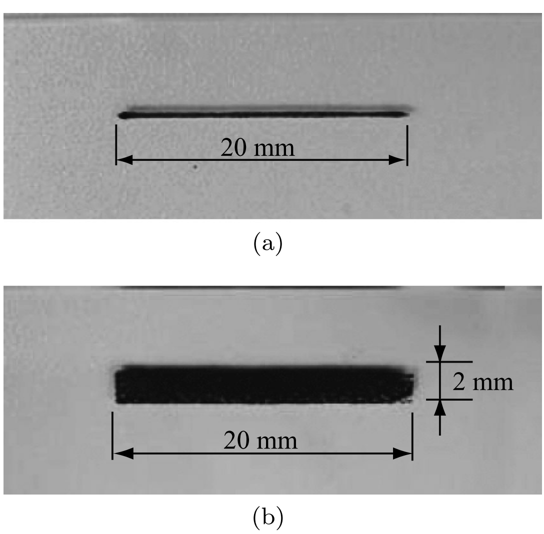

Figure 6. Printing test: (a) straight line and (b) strip shape.

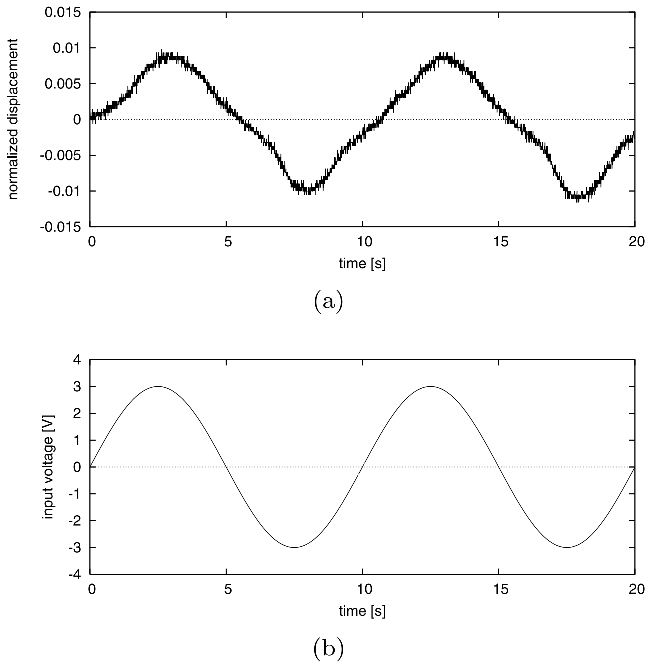

Figure 7. Experimental result of the printed bucky gel device: (a) normalized displacement of the actuator and (b) input voltage.

the materials. Figure 7 shows the experimental results of a printed bucky gel device as an actuator. In this test, a sinusoidal signal was applied as an input voltage and the displacement at a point near the tip measured by a laser displacement meter. The size of the film was 14 mm × 7 mm and 380 μm thick. From the result, it can be seen that the printed actuator was activated the same as actuators that were made by the solution casting method.

In the case of manual fabrication, it was difficult to form arbitrarily-shaped devices with a precise size. By using the printing system, arbitrarily-shaped bucky gel devices, not only of rectangular shapes but also curved shapes and complicated shapes, can be fabricated directly. As an example of curved shapes, we conducted the printing of a handshape pattern. Figure 8 shows sequential photographs of the printing process (each photograph is captured at intervals of 0.5 s). Figure 9 shows the printed film of the handshape. The electrode layers were printed in the outer shape and the electrolyte layer was printed in the whole area. As shown in the result, arbitrarily-shaped devices can be fabricated easily. In Ref. [17], a spiral-type actuator was also fabricated and its linear motion was demonstrated experimentally.

3.3. Discussion

In this section, we elaborate on the fabricated devices and consider the results of the printing method. On the printing of the straight line of Fig. 6a, we used a nozzle

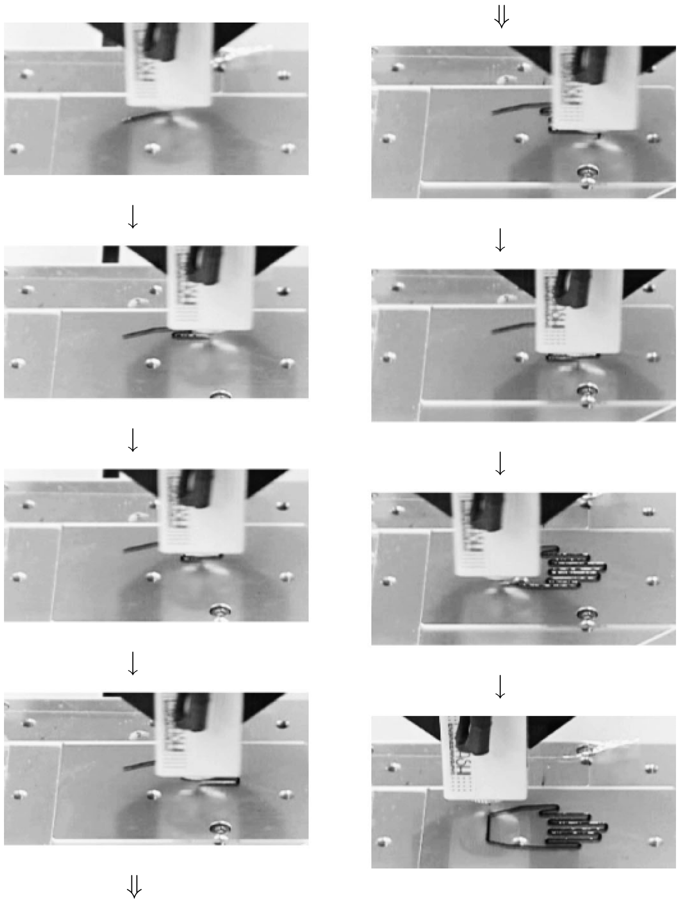

Figure 8. Sequential photographs of the printing of a handshape.

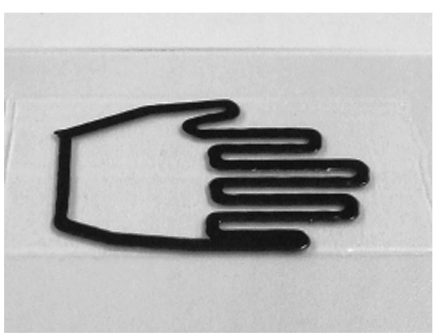

Figure 9. Printing result of a handshape.

of 100 μm diameter, and the distance between the nozzle and target plate was set to 300 μm. Under digital microscopy (Keyence VHX-900), the width of the line was about 330 μm and the error in the test was within 20 μm. The thickness of the printed materials was 300 μm just after printing and thinned to about 30 μm after drying of the solvents. The shapes of the printed materials at the starting and end points were not uniform. At the starting point, the material spread out in a circle to a size of 500 μm. At the end-point, the material spread out in an acuate shape. Although constant discharges were difficult in the transient state of 'on' and 'off' of the discharge, it is considered that the accuracy can be improved by controlling the speed the printing.

The positioning accuracy of the three-axis robot is sufficient for our experiments; instead, the accuracy of the printed materials depends on accuracy of the discharge rate. The surface roughness and the width of the printed materials depend on the discharge rate. To realize uniform discharge of the materials, it is important to adjust of discharge conditions, such as the viscosity of the materials, discharge pressure and size of the nozzle. Quantitative evaluation of the surface roughness, and the accuracy and the optimization of the discharge condition are future investigations. There was no problem in repeating the printing and the surface roughness was considered to be small. It is considered that the downsizing of the printed devices can be realized by a using smaller nozzle and adjustment of the printing conditions. In addition, we can print the materials not only on a flat target but also on a curved object.

By using the printing technique, we can control the shape, size and thickness of the printed devices. It is very useful to fabricate samples for characteristic verification. Comparison of the same condition or various parameters can be easily conducted.

In the previous works, we have investigated the integrated design of the actuator/sensor of the EAP device [16, 18]. By using the printing method of the bucky gel device, it is easy to construct the parallel or stacked device of the sensor and actuator. Actually, these devices have already been fabricated by way of trial. Although we have verified the simultaneous utilization of actuators and sensors, the problem was determined as the electrical interference of the activation input to the sensing signal occurs. A similar problem is present in another high-polymer gel actuator/sensor such as the IPMC [19, 20] and we have engaged in research to solve the problem.

4. Three-Dimensional Patterning

The bucky gel device has a three-layer configuration of the electrodes and electrolyte. In the case of the normal configuration of each layer, the bucky gel actuator bends into a circular arc. By using the printing system, a 3-D pattern of the electrodes can be constructed and various actuator devices that bend in particular shapes can be developed. As an example of the 3-D patterning, we propose the actuator that bends into an S-shaped curve.

Figure 10 shows the configuration of the actuator that bends in an S-shaped curve. This pattern has a reversing connection of the electrodes [21]. As shown

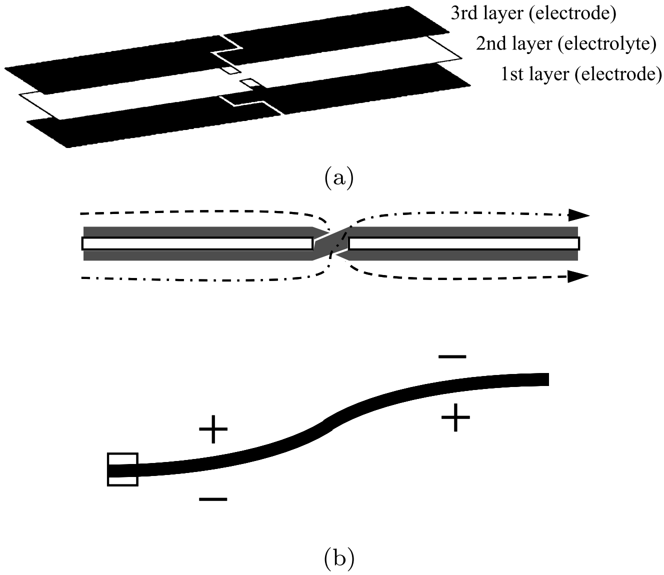

Figure 10. Three-dimensional patterning (reversing connection): (a) printing pattern and (b) bending image.

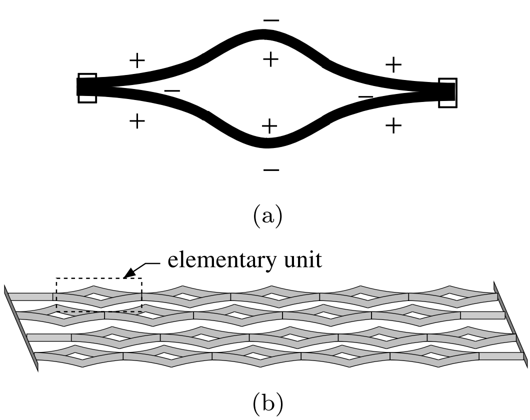

Figure 11. Application of reversing the connection: (a) elementary unit and (b) image of the linear actuator.

in Fig. 10b, the right side of the upper electrode is connected to the left side of the lower electrode in the central part and the opposite side has the same configuration. These patterned devices can be realize by the S-shaped bending motion. By combining the reversing connection, a linear actuator can be constructed as in Fig. 11. This structure is realized by combining the parts of different bending directions and then switching of the electrode is required. By utilizing the reverse connection, additional connection of electrodes is not necessary.

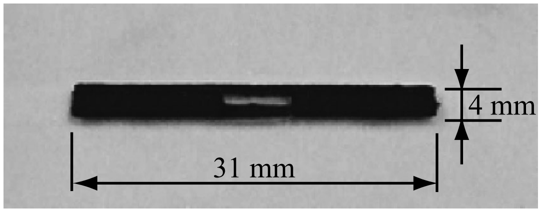

Figure 12. Reversing connection film.

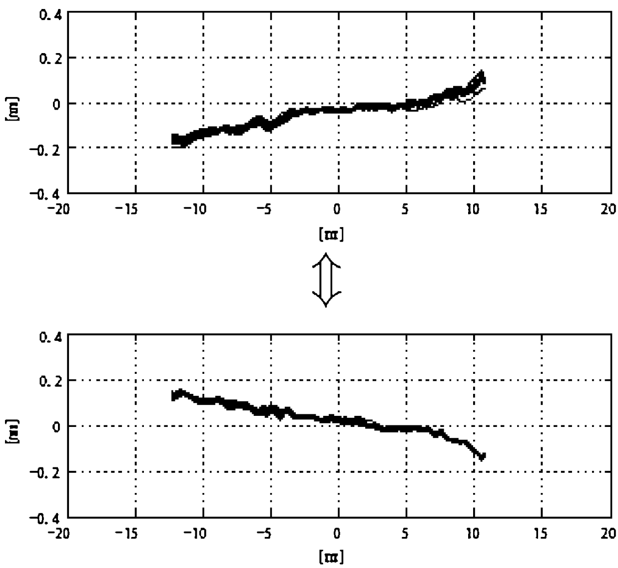

Figure 13. Test result of reversing connection film.

In order to investigate the 3-D patterning, the reversing connection film was fabricated. The pattern of Fig. 10a was printed on the glass plate. The materials and printing procedure were the same as the printing tests in Section 4. Figure 12 shows the printed result; its size is 31 mm × 4 mm and 60 μm thick. Figure 13 shows the experimental result of the actuation test. The electrode was fixed on the point 3 mm from the center, and the deformation of the film was measured by a 2-D laser displacement sensor (Keyence LJ-G200). In this test, input voltages of ±3 V were applied. Since each layer was printed only once and the film was very thin, the deformation was small. However, the S-shaped curves were generated on both sides and it is confirmed that the 3-D patterning can be constructed by using the proposed printing system. It is considered that the 3-D patterning was impossible by manual fabrication and that the printing system realized the development of devices with a new function.

5. Conclusions

In this paper, we demonstrated the fabrication of the bucky gel devices with a printing method. By using the dispensing machine, the printing system was constructed and the manual forming process was replaced with automatic printing. Printing tests of the bucky gel films were conducted, and the printing of complicated shapes and 3-D configurations was demonstrated. It was indicated that the printing system has the possibility to realize devices with new functions. The bucky gel device can be activated by low voltages; however, it cannot output large forces since the output force depends on the stiffness of itself. Furthermore, high polymer materials have good formability and it is easy to form various shapes. To take advantage of the bucky gels, small devices such as MEMS, micro-robots and micro-sensors are important application areas.

We will investigate the optimal conditions of the printing method, and then various designs and miniaturization of the actuator/sensor integrated system should be realized. The printing system is very attractive since there is the possibility of printing of the whole system, including actuators, sensors, power cells, electrical circuits, signal lines, etc. Although improvements in performance are needed for practical use, we consider that the bucky gel actuator/sensor has great potential for application in soft robotics and MEMS.

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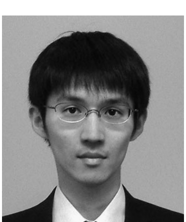

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