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Preliminary Study on Piezoresistive and Piezoelectric Properties of a Double-layer Soft Material for Tactile Sensing

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This paper describes a double-layer simplified sensor unit with interesting electromechanical properties, which consists of a film made from multiwalled carbon nanotube (MWNT) mixed by polymer composite and a thin film of polyvinylidene fluoride (PVDF). It is envisaged to imitate the distributed tactile receptors of human hands so as to help the disabled to recover the basic tactile perception. This paper shows the fabrication and performance research of such a new piezoelectric-piezoresistive composite material, which indicates a promising application in prosthetic hand.

Keywords: double-layer; piezoelectric-piezoresistive composite material; multiwalled carbon nanotube; PVDF.

1. INTRODUCTION

Tactile sense is one of the basic perceptions of human beings. However, traditional artificial limbs lack the capability of tactile perception, which deteriorates the rehabilitation and disability repairment. Since late 1990s, intelligent artificial limbs have been studied to develop tactile systems to help the disabled to reproduce the ability of sensing. Tactile sensors implanted in artificial limbs have an important influence and a wide application.

Most past work studying tactile sensors mainly involved silicon element by introducing either piezoresistive effect [1 – 3] or capacitive one [4 – 6]. There were also compliant robotic tactile sensors fabricated from piezoelectric polymer [7 – 9]. Another category of tactile sensor is made by integrating rigid silicon elements into a compliant polymer substrate, such as implanting silicon sensor units into polymer skin [10, 11]. Due to the application requirements of flexibility/compliance, recently researchers are moved to the array fabrication and integration of total polymer formed tactile sensors [12 – 14].

The mechanical perception stimulation of human hands is achieved by tactile corpuscles, which can be divided into four main layers as the increase of depth of tactile elements in epidermis [15]. They are Meissner's corpuscles, Merkel's disks, Ruffini's corpuscles and Pacinian corpuscles. They are distributed at different depth in epidermis with enough distance from rigid bone, between the receptors and the bone filled by soft tissues, which endows them receiving good strain even at minor mechanical stimulation [16, 17]. The dimension of the receptive field of a receptor is different from each layer, which increases with the depth of the epidermis. Besides, the response frequency range of signal reception of each layer is also different. Merkel's disks and Ruffini's corpuscle mainly sense static deformation (static pressure), while Meissner's corpuscles and Pacinian corpuscles mainly sense tiny contact and vibration (dynamic simulation).

Most of the tactile sensors mentioned above have the capability to perceive multivariate force, but rare of them are able to perceive steady strain and dynamic stimulation simultaneously like human hands. It is obviously benefit to developing a tactile sensory system if a sensory element can reliably provide both static and dynamic response to mechanical stimulations. It is well known that piezoresistivity is suitable to steady measurement and piezoelectricity is essential for dynamic sensing to mechanical stimulation. There are reliable doped silicon piezoresistors and piezoelectric ceramic sensors with good sensitivity existed and extensively used in many fields. However, their rigid and brittle characters lead us to look for the flexible/compliant substitutes, which are feasible to bond together and meet the static/dynamic sensing.

H. Kawai found strong piezoelectric capacity in one kind of polarized fluorine-containing compound – PVDF. PVDF piezoelectric films have the properties of broadband, high dielectric strength, high tenacity and flexibility, which make them ideal materials for piezoelectric sensors in tactile sensing. In this paper PVDF film is also used as one layer and mimicking the tiny contact and vibrations sensibility of human hand. Many research groups are working at new compliant conductive material called conductive polymer matrix composites (CPMC), which shows interesting piezoresistivity with flexible/compliant properties [18, 19].

With this train of thought and by analysis of the four types of receptor working frequency and consideration the availability well-developed signal processing technologies, the authors shift on trying to realize a novel double-layer piezoelectric-piezoresistive sensing element, which is able

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to gather both static and dynamic mechanical stimulation information so as to imitate the tactile corpuscles functions.

Fig. 1 shows the envisaging design for this combined compliant tactile sensor array. The combined compliant functional material consists of upper layer with piezoelectricity and lower layer with piezoresistivity, which relates to tiny contact / vibration and static deformation detection respectively so as to perceive dynamic mechanical stimulation and static pressure. Designed thin electrodes are deposited directly on the piezoelectric material surfaces in order to separate the material into different meshes so as to enable the capability of contacting position detection. The middle electrode layer can be used both for the piezoelectric and piezoresistive elements and no any adhesive glue is used between the electrode layers and functional layers in order to maintain the whole composite with good mechanical property immune to the drawbacks possible caused by adhesive gluing process such as hardening and non-uniform of the glue layer.

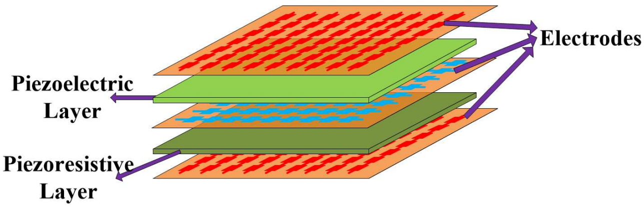


Fig. 1. Concept of tactile array with the piezoelectric-piezoresistive double-layer composite

In order to reach the envisaged tactile system, we need to firstly investigate the multilayer composite fabrication feasibility, validate its sensibility of the multifunctional composite. This paper presents the first trial investigation on related fabrication process, its sensibility analysis instead of development the sensor array itself though it is closer to applications. Based on these considerations, Section 2 deals with a simple molding technology from multilayer functional films fabrication to packaging processing, Section 3 illustrates the experiment setup, results and discussions and Section 4 as conclusion.

2. FABRICATION

2.1. The functional materials selection

As indicated in Section Introduction, PVDF thin film can be a good selection as the compliant functional piezoelectric material due to its good mechanical properties and piezoelectricity.

The conductivity of a conductive polymer matrix composite (CPMC) depends on the content of the conductive particles in the base polymer matrix governed by the percolation theory. Its piezorestivity is closely related to the volume ratio of doping conductive particles to polymer base, the conductive properties of the conductive particles and the mechanical property of the entire composite. The authors choose polyurethane (PU) as the polymer base due to its easy availability with variety mechanical properties. The carbon nanotube (CNT) has been extensively in different domains such as electrode materials and nanoelectronics devices, due to its unique mechanical properties and electrical properties since the carbon nanotube (CNT) was first discovered by Iijima [20], which leads us to choose multiwalled carbon nanotube (MWNT) as the doping conductive particles. The bottom piezoresistive layer consists of MWNT/PU CPMC.

2.2. Fabrication of Piezoelectric Layer – PVDF film

A simple procedure is used to fabricate the PVDF film. 0.71 g PVDF (-TrFE) (Piezotech S.A.S) is solved in 5 ml DMF and sonicated over 2 h. Then the solution is poured into 30 mm × 30 mm copper mould following several time distillations under a reduced pressure chamber, which is necessary step to eliminate possible tiny bulbs generating in the final piezoelectric film. After that, the mould with the solution is stewed under 70 °C for about 1 h. The obtained PVDF film with thickness of 75 μm is shown in Fig. 3, a.

2.3. Fabrication of Piezoresistive Layer – MWNTs/PU film

Modified MWNTs, MWNTs-OH is used as doping conductive particles in order to improve its dispersing capability into the PU matrix while both mixing in a proper solvent. The main physical parameters of MWNTs-OH are listed in Table 1.

A bunch of experiments have been done to define the best doping density of MWNTs in order to obtain the good piezoresistive performance of composite. The fabrication process of MWNT film presents in following flow chart (Fig. 2). The MWNTs are beforehand dissolved in DMF with the weight percentage of 2 %.

Table 1. Some physical parameters of MWNTs-OH

OD (nm)	Purity (wt%)	Length (microns)	SSA (m^2/g)	Tap density (g/cm^3)	–OH content (wt%)
>50	>95	0.5-2	>40	0.18	0.71

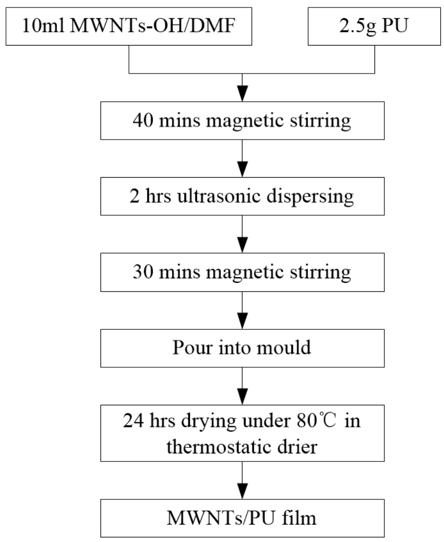


Fig. 2. Flow chart of MWNT/PU film fabrication process

The simple molding technology is applied again with the same mould. Final obtained MWNTs/PU film is shown in Fig. 3, b, whose dimension is 30 mm × 30 mm with the thickness of 12 μm.

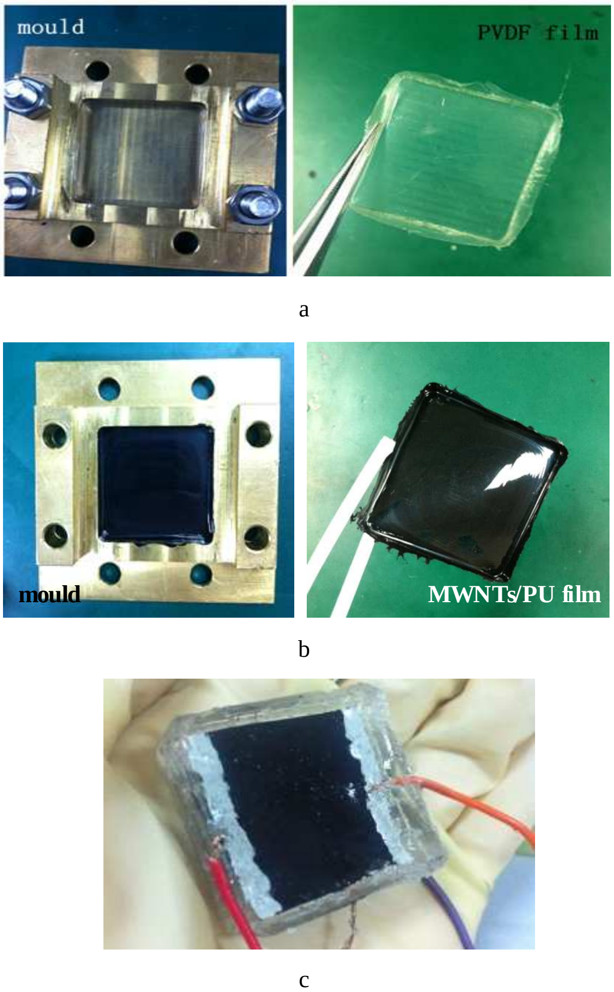


Fig. 3. The fabrication of piezoelectric-piezoresistive unit: a – the mould and obtained PVDF film; b – the mould and obtained MWNTs/PU film; c – compliant packaged double-layer piezoelectric/piezoresistive composite unit

2.3. Packaging

In order to get rid of the drawbacks of the adhesive gluing processing as mentioned in Section Introduction, Dowcorning Sylgard184 filling glue (PDMS) is selected to package the whole multilayer functional composite into a compliant tactile sensing unit. PDMS has been extensively used in compliant package due to its easily processing capability and variety hardness selection. Appropriate Young’s Modulus needs to be chosen to produce similar hardness as the human being skin. After several trials, the mass ratio of host crystal and curing agent has been defined as 7.5 : 1. With this ratio, the package PDMS material reaches proper compliance. Firstly, the ready PVDF film and MWNT film aligned and stacked carefully and then restored into the mould; Secondly, well prepared mixture according defined ratio gently poured into the mould and put into vacuum chamber for 30 min to get rid of tiny bubbles; Thirdly, the miture was then moved into a drying oven under 80 °C for 3 h. The compliant multifunctional composite unit by simple molding technology is shown in Fig. 3, c.

3. EXPERIMENTS AND RESULTS

3.1. Piezoelectric-piezoresistive composite testing setup

3.1.1. Setup for piezoelectric testing

The construction of the dynamic testing system for PVDF film is shown in Fig. 4. A dynamic exciter is used to provide mechanical stimulation to the PVDF film so as to generate charges on the film’s surfaces. The generating charges are then fed into a charge-amplifier, at which the tiny charge signal is transformed into voltage signals and collected by a DAQ card (NI Instrument) and displayed on a monitor. The mechanical stimulation frequency is 1 kHz so as to cover tactile dynamic sensing range. The dynamic load is monitored by a standard pressure sensor embedded in the exciter.

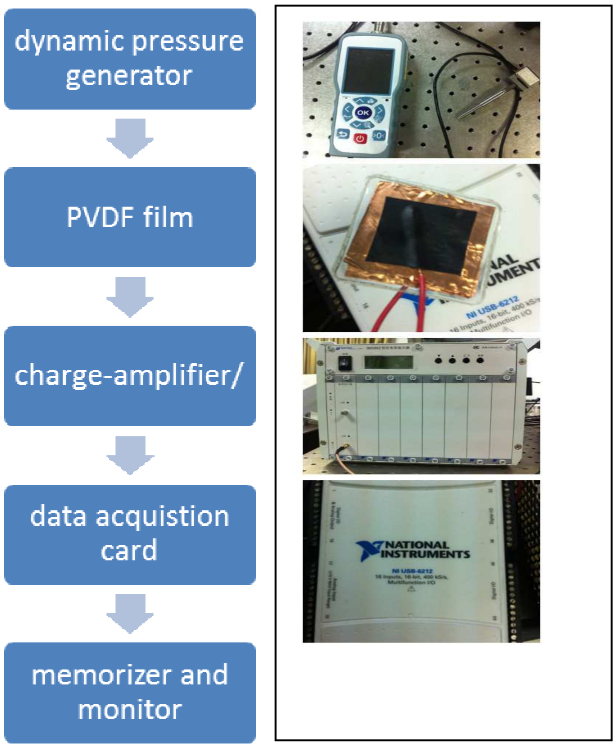


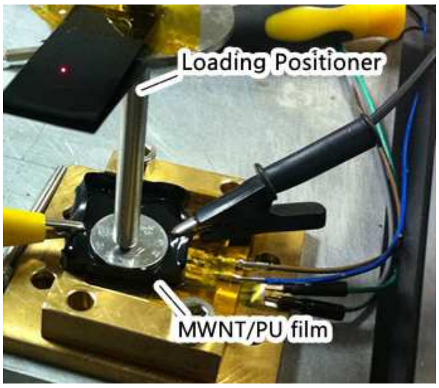
Fig. 4. Setup of the dynamic testing for PVDF film

3.1.2. Setup for piezoresistive testing

As to piezoresistivity testing of the MWNTs/PU film, the silver painted electrodes of the piezoresistive film are connected to a high voltage source (HB-621) and a high resistance test box (HB-600A) is used to measure its initial resistance (Fig. 5, a). The high voltage source works at a constant voltage. The resistance can be calculated according to the added voltage and generated current through the piezoresistive film simply through Ohm’s law.

After the initial resistance testing, the investigation of MWNTs/PU film piezoresistivity must be proceeded at the situation of different loads (Fig. 5, b). A compliant layer of PDMS with thickness about 1 mm is formed in the mould, which mimics the natural tactile receptor’s compliant base. The piezoresistive film is moved onto the PDMS layer, then aligned and carefully spreaded to eliminate gas between the contacting surfaces. In order to avoid

a



b

Fig. 5. Setup of the piezoresistive film testing system: a – initial resistance test of MWNTs/PU film; b – resistance test under the static pressure.

destroying the thin film, the load is gradually added through a ten cents coin center via a probe. The working procedure is first loading, waiting for several seconds and then unloading. Motion of the probe is controlled by a three-dimensional positioner (VP-25XYZL, Newport), which guarantees applied load vertically and slowly onto the piezoresistive film. The load value is monitored by a force gauge (EVT-10F-2kg) inserted in the loading positioner. A simple electric circuit structure is used for the testing. One selected constant resistor is serial connected with the piezoresistvie film; the other terminal of the constant resistor is connected to the high voltage source, and the other one is serial connected to one terminal of the piezoresistive film; the left terminal of the piezoresistive film is then connected to the signal ground. With this simplified electric circuit structure, the voltage on the piezoresistive film reduces while it is deformed by increasing its resistance. The same data collection device (NI Instrument) as in the setup of PVDF film testing is used to monitor the voltage changes, which reflects the resistance change.

3.2. RESULTS AND DISCUSSION

3.2.1. Electrical properties of PVDF film

Ten times testing data averaged value is shown in Table 2, at which contains 10 outputs vs each effective pressure-load.

From the statistics, the authors obtain a fitted approximate equation that indicates its piezoelectric response with good sensitivity and linearity of the PVDF film during dynamic stimulation (Fig. 6, a). The fitted equation is $y = 1383x + 8.458$ with relative coefficient is 0.997, which shows good linearity and stability.

Table 2. Pressure and output charges of PVDF film

No.	Pressure (MPa)	Output charges (pC)	Sensibility (pC·MPa^{-1})
1	1.02	1398	1370.59
2	1.62	2357	1454.94
3	2.02	2667	1320.3
4	3.05	4401	1442.95
5	3.98	5568	1399
6	4.08	5782	1417.16
7	5.11	7238	1434.05
8	6.11	8186	1339.77
9	6.34	8398	1324.61
10	7.13	9838	1379.8
11	7.68	10637	1385.03
12	8.15	11269	1382.7
13	8.73	12305	1409.51
14	9.18	12388	1349.46
15	10.2	14475	1419.12

Another important parameter is sensibility which is defined as [21]:

$$K = \frac{Q_{\max}}{P_{\max}} \text{ (pC/MPa)},$$

where $Q_{\max}$ and $P_{\max}$ are the peak value of output charge and pressure, respectively. The average sensibility of the PVDF film is 1388.60, which is basically up to the common pressure sensors in current market.

3.2.2. Electrical properties of piezoresistive film

The initial resistance is $5.56\times10^{10}\,\Omega$, which is indicated as R_0 in following piezoresistive film response testing. During the loading /unloading testing, generally, it shows clearly that the piezoresistive film response can follow the mechanical stimulation well either in loading and unloading period except 2 abnormal step sections (Fig. 6, b). And the response curve slopes maintain well the trends before and after each abnormal step. A time-delay of response is clearly observed which should be due to the compliance of the film also the underneath base softness. The abnormal step sections are appeared regularly during cycle testing both in loading and unloading periods.

After checking the testing setup in detail, the authors believe that the abnormal step responses relate to space confine of the mould wall, which limited the film elongation during the testing. When a load applied onto the piezoresistive film, the film under the coin undergoes stretching and thinning which leads its resistance increasing (let us call it positive effect); meanwhile, the other part is driven and approaches to the mould borders so as to be compressed, which leads it to be thicker and gives negative effect of the film resistance changing. Both the resistance changing effects contribute to the testing response. During the loading period, the whole film resistance increases following the load increment at the beginning due to also the underneath base deformation and the negative effect is small. The negative effect becomes stronger and stronger when the load continuous increased.

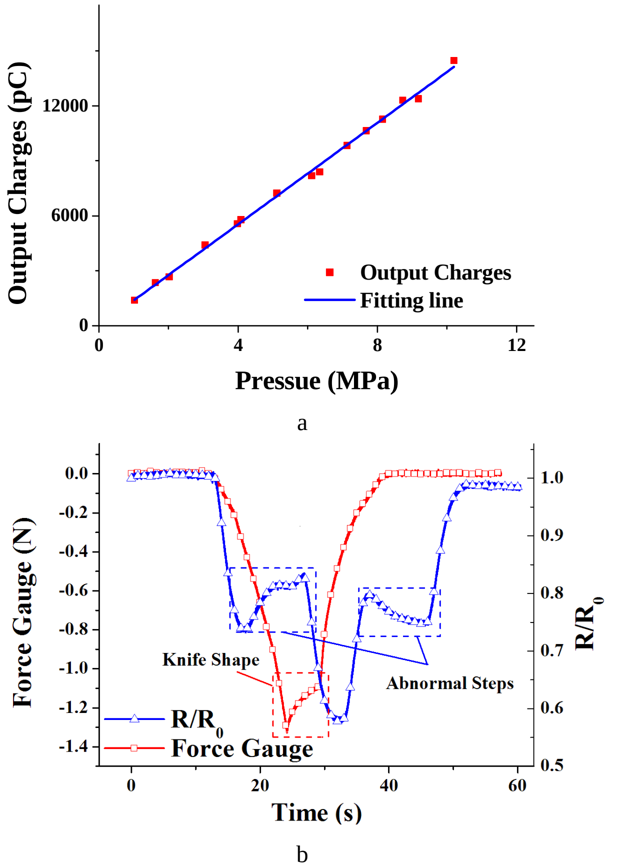


Fig. 6. Piezoelectric-piezoresistive film properties: a – relationship between pressure and output charges of PVDF film; b – relationship between stimulation gathered by the force gauge and normalized resistance of the piezoresistive film

The abnormal step response appears till it becomes dominant. However, the negative effect cannot increase endlessly due to compliant property limitation. The composite will become curved instead of continuing compressed. That is why the positive effect overcomes the negative one when load continues increased and the response follows the stimulation again. During the unloading period, similar situation happens only at opposite sequences. The input (the mechanical stimulation in force) and output (the normalized resistance of the piezoresistive film) relationship is shown in Fig. 7, a. It is should be noted that the authors cut off the data of the abnormal steps during the fitting process because this space confine is not the real application situation. The fitted equation obtained between the mechanical stimulation and the normalized resistance is $y = 0.98703 + 0.24687x$ with relative coefficients 0.858. The gauge factor calculated from the curve is about 3.5, which is better than the available piezoresistors made from the metal wires. Another important property, the hysteresis characteristics has also been investigated and shown in Fig. 7, b.

It is observed that during the waiting time the force reduction forms a sharp knife shape response due to the relaxation of the soft material properties. Comparing to the force gauge sensor, the response of developed piezoresistive film has shown smoother response, which hints the possibility better imitating the slowly adapting function commonly happened in human hands.

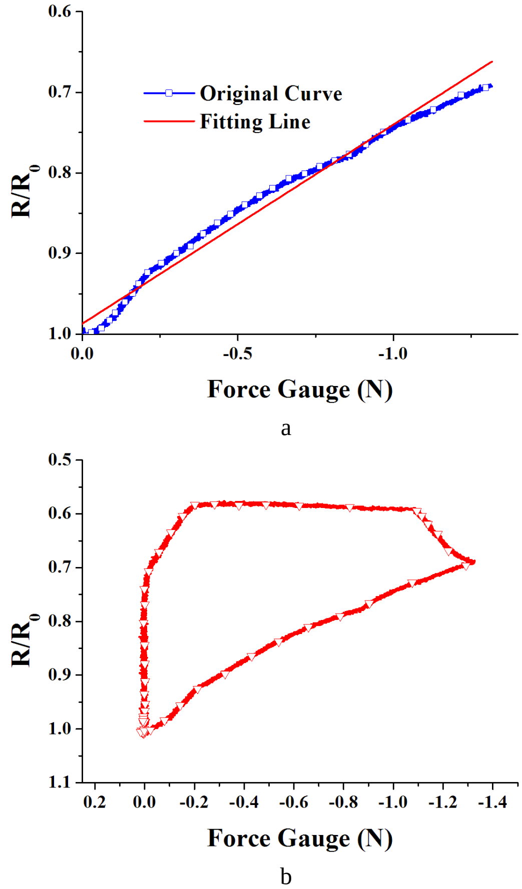


Fig. 7. Piezoresitive response of composite unit: a – relationship of load and output; b – hysteresis of piezoresistive film

4. CONCLUSIONS

This paper presents the development of a new piezoelectric-piezoresistive composite unit envisaged for tactile sensory applications, which is integrated from MWNT/PU composite and PVDF layers through a simple molding technologies. The molding fabrication procedure of the PVDF layer, the MWNT/PU layer and the adhesive glue drawbacks avoiding package method has been explained in detail. The sensing unit piezoelectric and piezoresistive properties have been investigated respectively. The sensitivity of the formed piezoelectric film is comparable to commercial piezoelectric one with 1388.60 pC/MPa and good linearity among the tactile stimulation range. The gauge factor of the compliant MWNT/PU composite is about 3.5 and better than current piezoresistors made from the metal wires. The preliminary testing results show the feasibility of the novel double-layer compliant sensing unit promising application in tactile field, at which dynamic mechanical stimulation and static pressure simultaneous detection is preferable. However, it can only been seen as the first step in the trials of the development of this double-layer novel compliant sensing composite. The piezoresistive film initial resistance needs further reduced in order to facilitate simple measuring interface instead of using specific instrument such as high voltage source, which enables the real applications. Considering the size of the natural sensory receptors is quite different from each other, the thickness of the piezoelectric-piezoresistive film and also the other dimensional parameters optimization has to be further investigated in future work. After that, the authors will focus on development of the novel double-layer array fabrication method including the molding technology for sensory array fabrication, the deposition technologies for the multilayer electrodes, and modeling technologies for suppress signal interfering which is quite possible from it intrinsic soft property.

Acknowledgments

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