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Piezoelectric Stretchable Sensor with a Vertical Wavy Structure Fabricated by Combining Dip Coating and Micro-corrugation Process

Michitaka Yamamoto[1] [ORCID] **· Naoto Tomita[1] · Seiichi Takamatsu[1] · Toshihiro Itoh[1]**

Abstract

A novel fabrication process for a vertical wavy structured stretchable piezoelectric sensor combining dip coating and micro-corrugation process is proposed. By changing the dip coating withdrawal speed, the thickness of PVDF-TrFE (poly(vinylidene fluoride-trifluoroethylene)) films deposited on metal foils was controlled; the wave shape fabricated by the micro-corrugation process was influenced by the PVDF-TrFE film thickness. By reducing the PVDF-TrFE film thickness to less than 5 μm, the wave shape exhibited a high aspect ratio (wave height divided by wave pitch). From estimations obtained by measuring the change in substrate length before and after the microcorrugation process, the predicted stretchability is expected to be greater than 30%. The fabricated vertical wavy structured piezoelectric sensor with a PVDF-TrFE film thickness of approximately 2 μm showed more than 50% stretchability. The fabricated sensor was used as a finger-bending sensor for a virtual reality system, and the proposed process is a promising method for fabricating stretchable sensors.

1 Introduction

With the recent development of soft robotics or virtual reality (VR), human–machine interfaces, such as sensors and actuators, are becoming increasingly paramount [1–4]. To ensure safety and comfort for humans, device surfaces should be soft, and sensors exhibiting not only flexibility but also stretchability are required [5, 6]. More than 30% stretchability is required because the skin strain due to human motion is up to 30%, except in some areas of great stretch.

Strain sensors are essential for human motion detection. Several strain sensors such as piezoresistive [7, 8], capacitive [9], and piezoelectric [10] sensors have been studied. Particularly, piezoelectric sensors have gained attention because of their simple sensing principle, high sensitivity, large dynamic range, and low energy consumption

[11]. PVDF (Polyvinylidene difluoride) and PVDF-TrFE (poly(vinylidene fluoride-trifluoroethylene)) are typical organic piezoelectric materials used for sensing, with better softness and flexibility characteristics than inorganic piezoelectric materials such as lead zirconate titanate and aluminum nitride [12]. However, the stretchability of organic piezoelectric films is still only around 2% [13]; therefore, additional processing to fabricate stretchable piezoelectric sensors is required.

There are two approaches for adding stretchability to nonstretchable devices: fabricating stretchable materials [14–18] and fabricating stretchable structures such as two- or three-dimensional spring-like structures [19–27]. The latter has advantages such as the possibility of using existing fabrication processes and good material properties. Among various stretchable structures, wavy structures are prominent [20–27]. Two direction types are possible when fabricating wavy structures: horizontal wavy structures in which a wave is formed in the substrate in-plane direction and vertical wavy structures in which a wave is formed in the substrate vertical direction.

Horizontal wavy structures are easy to process and exhibit a high stretchability of 100% or more [24, 25]. However, there are still problems such as requiring large surface areas.

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✉ Michitaka Yamamoto
yamamoto-michitaka@g.ecc.u-tokyo.ac.jp

1 Department of Precision Engineering, Graduate School of Engineering, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

Vertically wavy structures can produce high-density devices. Nevertheless, the prestretch method in which a metal film is placed on a prestretched substrate and a vertical wavy structure is formed using the substrate return force is the standard method for fabricating vertical wavy structures, making it difficult to stably produce them [20, 23, 26, 27].

Recently, a micro-corrugation process in which micro gears continuously bend metal foils has been proposed [28, 29]. In the micro-corrugation process, the gear shape, distance between gears, and substrate characteristics, such as material type or thickness, can control the wave structure. Thus, stable vertical wavy structure fabrication can be achieved. Furthermore, a stretchable piezoelectric sensor has been fabricated by micro-corrugating a metal foil with an organic piezoelectric film [30]. However, the fabricated sensor showed only 15% stretchability because the thickness of the metal part, which can be plastically deformed, was thin (1 μm), the thickness of the organic film, which cannot be plastically deformed, was thick (28 μm), and the height of the fabricated wave was low and unstable [30]. To improve the stretchability of a micro-corrugated sensor, control of the metal foil and organic film thickness is required.

In this study, we propose a novel fabrication process for a vertical wavy structured stretchable piezoelectric sensor by combining dip coating and micro-corrugation. Particularly, a metal foil with a dip-coated PVDF-TrFE film is vertically structured via the micro-corrugation process. Dip coating is one of the commonly employed ways to fabricate organic piezoelectric films such as PVDF-TrFE films [31, 32], and the film thickness can be controlled by the dip coating withdrawal speed. In addition, we evaluated the effect of PVDF-TrFE film thickness on the micro-corrugated structure. The stretchability of the fabricated piezoelectric sensor is also evaluated, and the fabricated sensor is demonstrated as a finger-bending sensor in a VR system.

2 Experiment Method

2.1 Sensor Fabrication Process

The proposed fabrication process for the vertical wavy structured stretchable piezoelectric sensor is shown in Fig. 1. A metal foil was prepared as the substrate, and PVDF-TrFE was deposited on the substrate via dip coating. The PVDF-TrFE film thickness can be controlled by the solution concentration and withdrawal speed. Next, the top electrode was formed by sputtering, and polling was performed. Then, a vertical wavy structure was fabricated through the micro-corrugation process in which micro gears continuously bend the substrates. Finally, the vertical wavy structured sensor was embedded in silicone rubber.
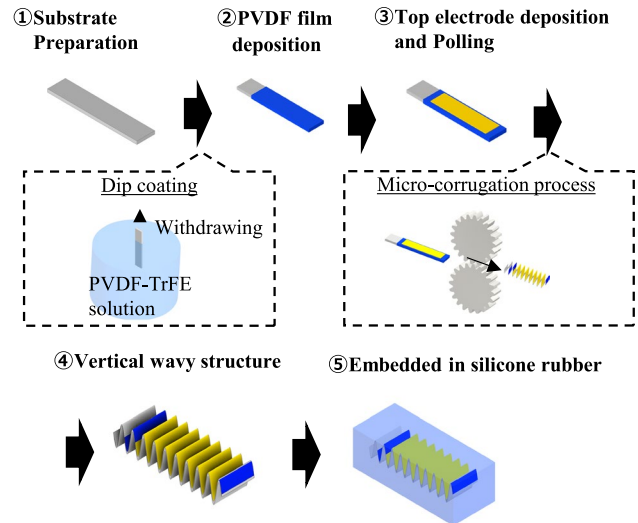

Fig. 1 Proposed fabrication process of vertical-wavy structured stretchable piezoelectric sensor

In this study, aluminum foil with an 11-μm thickness was prepared as the substrate. The solution used for dip coating was prepared by dissolving PVDF-TrFE powder (9 wt%, PIEZOTECH, FC25) in methyl-ethyl ketone (45.5 wt%) and N, N-dimethylacetamide (45.5 wt%). The dip coating withdrawal speed was varied from 10 to 250 mm/min, and the annealing process at 150 °C for 1 h was performed after dip coating. This dip coating and annealing process was repeated twice to stabilize the film deposition. The deposited top electrode was gold, and polling was performed by adding approximately 100 V/μm. The substrate was cut into 5-mm width, and the micro-corrugation process was performed. The gears used in the micro-corrugation were involute gears with module 0.15, and the gap between the gears was set to 400 μm. After the micro-corrugation process, lead wires were attached, and the fabricated sensor was embedded in approximately 800-μm thick silicone rubber (KE-1316, Shin-Etsu Chemical Co., Ltd.).

2.2 Evaluation Method

The deposited PVDF-TrFE film thickness was calculated from the measured capacitance by forming a 10×10 mm^2 top electrode. In the calculations, the PVDF-TrFE film relative permittivity was assumed to be 11, which is the nominal value of the PVDF-TrFE powder used.

The shape of the micro-corrugated samples was observed using a microscope or scanning electron microscope (SEM, Jeol Ltd., JCM-7000). The wave height and pitch of different PVDF-TrFE film thicknesses after the micro-corrugation process were measured from an observation image. There are two methods for predicting the stretchability of micro-corrugated materials: estimation from the pitch, height,

and angle of the wavy structure and estimation from the change in substrate length by the micro-corrugation process. In this study, the stretchability of each condition was estimated using the latter method. When estimating from the change in substrate length, the stretchability S (%) of a micro-corrugated sample can be estimated by measuring the initial length (L_1) and the length after micro-corrugation (L_2) as follows:

$$S = \frac{L_1 - L_2}{L_2} \times 100$$

In this study, the initial length and length after micro-corrugation of each substrate with different PVDF-TrFE film thicknesses were measured using a ruler.

The characteristics of the fabricated samples were evaluated by pulling the samples using a tensile testing machine (Aicoh Engineering Co., Ltd., FTN1-13A) and measuring the generated charge using a charge amplifier (Showa Sokki Co., Ltd., MODEL-4001B-50) and a source meter (Keithley Instruments Co., Ltd. 2450 Source Meter). The measurement system was connected to LabVIEW software, and all data were collected in batches. Notably, the data sampling rate was approximately 5 Hz. The tensile testing speed was set to 60 mm/min.

Cycle tensile tests were also performed to evaluate the sensor durability. The conditions for the cyclic tensile tests were the same as those for the tensile tests, and the durability of up to 30% stretch was evaluated.

2.3 Application as a Finger-Bending Sensor in VR

The fabricated sensor was used as a finger-bending sensor in the VR system. The sensor was attached to the second joint of the groove using glue and double-sided tape. It should be noted that both stretching and bending forces are applied to the sensor when a sensor is attached to a finger and bent. The sensor signals were captured in Unity software via a charge amplifier and data logger. Finger-bending detection was performed by setting a specific threshold and coefficient on the software.

3 Experiment Result

3.1 Experiment Result of Dip Coating

The relationship between dip coating withdrawal speed and PVDF-TrFE film thickness is shown in Fig. 2. Figure 2 shows that the PVDF-TrFE film thickness is approximately 1.2 μm at a withdrawal speed of 10 mm/min, and the thickness gradually increases with increasing speed. The film thickness was almost saturated at a withdrawal speed of
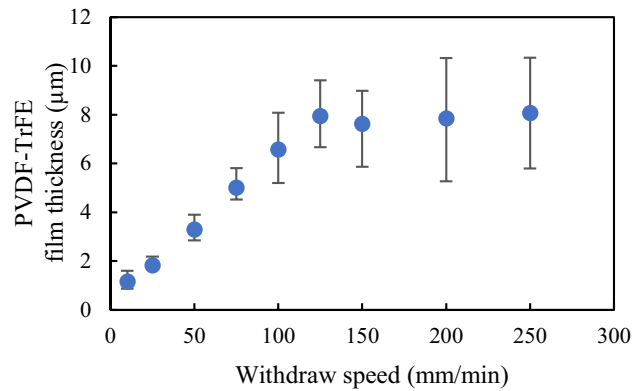

Fig. 2 Relationship between PVDF-TrFE film thickness and dip coating withdrawal speed
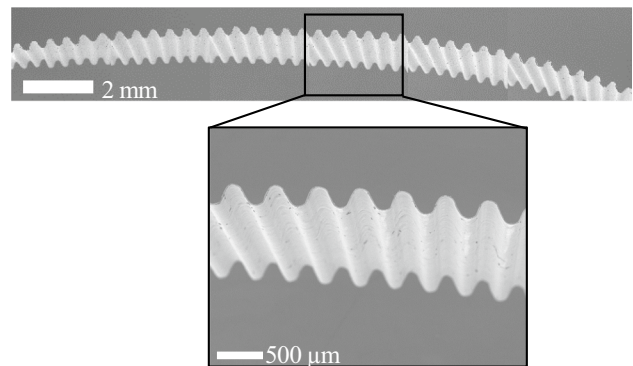

Fig. 3 Typical SEM image of micro-corrugated foils with PVDF-TrFE film

125 mm/min with a film thickness of approximately 8 μm. Notably, Fig. 2 shows the result of repeating the dip coating process twice, as aforementioned, and the measured film thickness is the result calculated from the measured capacitance.

3.2 Observation Results of Micro-corrugated Foils

A typical SEM image of micro-corrugated foils with PVDF-TrFE film is shown in Fig. 3. Figure 3 shows the sample with a dip coating withdrawal speed of 10 mm/min. It shows that a continuous wavy structure is fabricated by the micro-corrugation process.

Figure 4 shows the measured height and pitch of the micro-corrugated samples at different dip coating withdrawal speeds. Figure 4 shows that faster withdrawal speeds increase the wave pitch and decrease the wave height. Notably, the PVDF-TrFE thickness increased with the withdrawal speed, indicating that the wave shape collapsed compared with the gear shape as the PVDF-TrFE thickness increased.
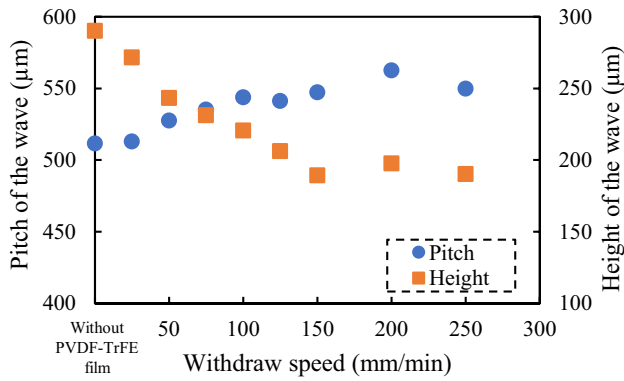
Fig. 4 Measured height and pitch of micro-corrugated samples at different dip-coating withdrawal speeds
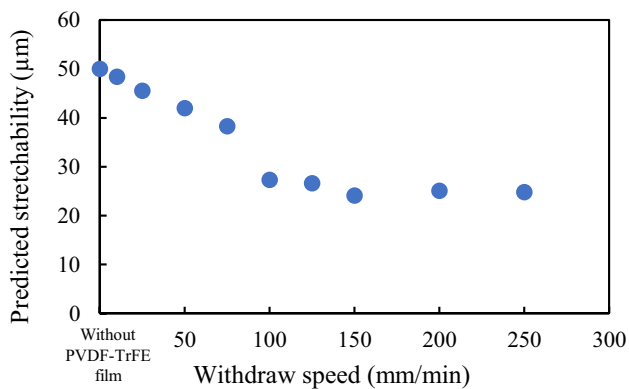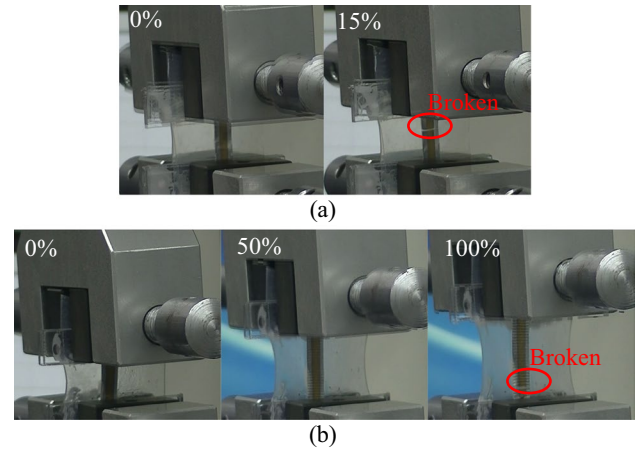

Fig. 5 Prediction result of stretchability of samples with PVDF-TrFE films deposited at different withdrawal speeds calculated from the change in substrate length
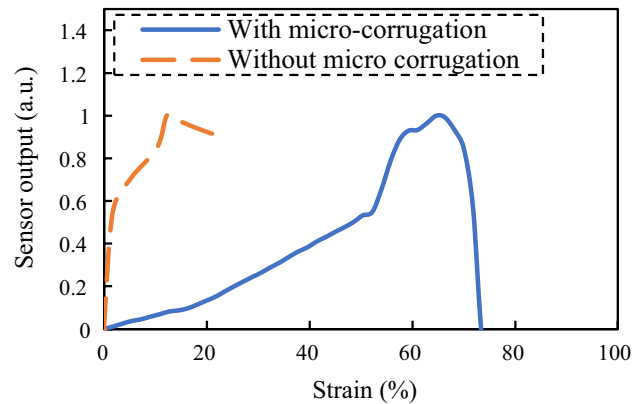

Fig. 6 The images during the tensile test of the fabricated sensor. The dip-coating withdraw speed is 25 mm/min; **a** without micro-corrugation process; **b** with micro-corrugation process

Fig. 7 Typical sensor output signal during tensile test of sample with or without micro-corrugation

The prediction result of the stretchability of samples with PVDF-TrFE films deposited at different withdrawal speeds calculated from the change in substrate length is shown in Fig. 5. The faster the withdrawal speed, i.e., the thicker the PVDF-TrFE, the smaller the predicted stretchability. This is consistent with the results shown in Fig. 4, where the wave pitch increased and the wave height decreased as the withdrawal speed increased. The predicted stretchability of the sample without PVDF-TrFE film deposition was 50%; although the predicted stretchability decreased with PVDF-TrFE deposition, it is expected to show over 30% stretchability by setting the withdrawal speed to less than 75 mm/min, which corresponds to a PVDF-TrFE film thickness of less than 5 μm.

3.3 Sensor Characteristic Evaluation Results

Tensile tests were performed on two types of samples: with and without micro-corrugation. The dip coating withdrawal speed was set to 25 mm/min. Figure 6 shows the images

obtained during the tensile test of each sample. In addition, a video of the tensile tests is provided in Supplementary Material 1. The samples without the micro-corrugation process were broken immediately after the start of the measurement, whereas stretchability was significantly improved with the micro-corrugation process.

A typical tensile test output signal is shown in Fig. 7, and the averages of three sample measurements for each condition are shown in Fig. 8. In Fig. 7, the vertical axis is normalized because the output signal varied significantly with respect to sensor installation conditions such as slight tilt. Because the main feature of the proposed process is the improvement of stretchability, normalization will not have a significant impact on the output. Without the micro-corrugation process, the sensor broke at approximately 20% strain, whereas it withstood strain up to approximately 80% with the micro-corrugation process.
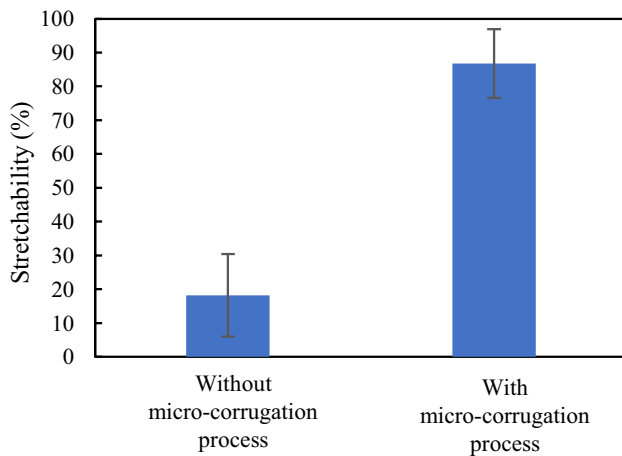
Fig. 8 Averages of three sample measurements of tensile test with or without micro-corrugation

In this case, the output signals were almost linear up to approximately 50%, indicating that the fabricated sensor can be used as a strain sensor.

The results of cycle tensile testing of up to 30% strain showed that the sensor was broken after approximately 150 cycles. Because no cracks in the substrate were observed by optical microscopic observation, the sensor seems to be broken in the PVDF-TrFE piezoelectric film or top electrode.

3.4 Application as a Finger-Bending Sensor in VR

An example of the fabricated sensor mounted on a glove and used as a finger-bending sensor on a VR system is shown in Fig. 9. The fabricated sensor succeeded in detecting finger-bending and linking it to hand movements in the VR system.

4 Discussion

In this study, the relationship between the wave shape fabricated by the micro-corrugation process and the thickness of PVDF-TrFE films deposited on metal foils was evaluated. The PVDF-TrFE film thickness could be controlled by the dip coating withdrawal speed, and the experiment revealed that when the PVDF-TrFE film thickness was increased, the wave aspect ratio (wave height divided by wave pitch) produced by the micro-corrugation process decreased. Thus, it is paramount to control the thicknesses of the metal foil and the organic film formed on it, and the thicker the organic thin film, the less stretchable it becomes. This is because increasing the ratio of organic materials during the micro-corrugation process causes a larger spring back due to the inability of the organic films to plastically deform while the metal does. The fabricated piezoelectric sensor achieved over 50% stretchability by controlling the PVDF-TrFE film and metal substrate thicknesses. Notably, the focus of this
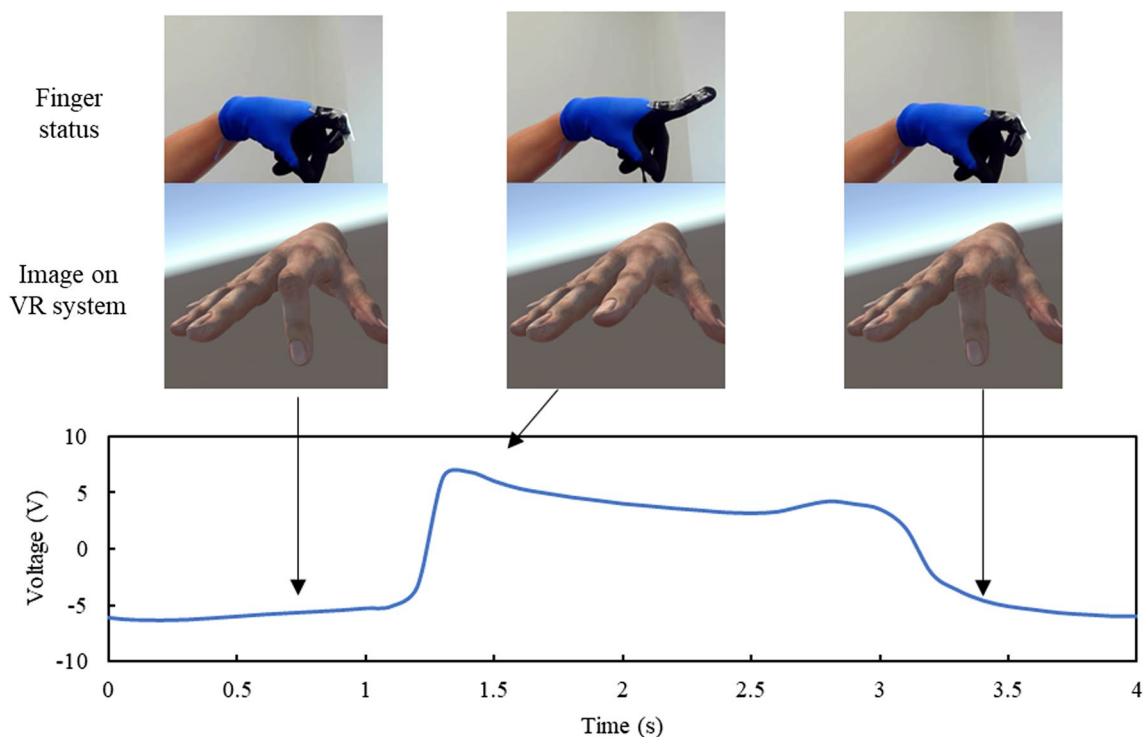
Fig. 9 An example of the fabricated sensor mounted on a glove and used as a finger-bending sensor on a VR system

study was on improving the sensor's stretchability, not its sensitivity. Further research is required to discuss the sensor's sensitivity. In addition, our evaluation setup had a low sampling rate, which was insufficient for evaluating the sensor's response time. Therefore, further evaluation of the sensor's response time is required in future work, though the response of piezoelectric sensors is more influenced by the circuitry in the subsequent stage than by the sensor itself.

It should also be noted that when the sensor is attached to the finger and bent, the sensor is both stretched and bent. Though the fabricated sensor succeeded in detecting finger bending because it has sensitivity to both stretching and bending, it is difficult for the fabricated sensor to distinguish between stretching and bending. Further approaches, such as arranging the sensors in the array or applying a bimorph structure, are required to distinguish between stretching and bending.

In the cycle testing of up to 30% strain, the fabricated sensor was broken after approximately 150 cycles. The number of 150 cycles is less than that of micro-corrugated copper interconnects with similar stretchability shown in a previous study (approximately 550 cycles) [29]. Because no substrate damage was observed using a microscope, the breakage appears to have occurred in the PVDF-TrFE piezoelectric film or top gold electrode. Because previous reports have shown that microcracks occurred on deposited metal interconnects formed on a metal substrate with an insulating layer after the micro-corrugation process [33] and inorganic materials break with less strain than organic materials, breakage in this experiment most likely occurred at the top electrode. The cycle number could be improved by changing the top electrode to an organic conductive material with a little more elasticity.

5 Conclusions

A novel fabrication process for a vertical wavy structured stretchable piezoelectric sensor combining dip coating and micro-corrugation is proposed. By changing the dip coating withdrawal speed, the thickness of PVDF-TrFE films deposited on metal foils was controlled; faster withdrawal speeds resulted in thicker PVDF-TrFE films. A vertical wavy structure fabricated by the micro-corrugation process was influenced by the PVDF-TrFE film thickness; the thicker the PVDF-TrFE film, the less stretchable the sensor. By reducing the PVDF-TrFE film thickness to less than 5 μm, the predicted stretchability is expected to be more than 30%. From the experimental results, the fabricated micro-corrugated piezoelectric sensor with a PVDF-TrFE film thickness of approximately 2 μm showed more than 50% stretchability. The fabricated sensor also functions as a finger-bending

sensor in a VR system. The proposed process is a promising technique for fabricating stretchable sensors.

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Michitaka Yamamoto received the B.E., M.E., and Ph.D. degrees from The University of Tokyo, Japan, in 2012, 2014, and 2020, respectively. He is currently a Research Associate with The University of Tokyo. His research interests include MEMS technologies and low-temperature integration.

Naoto Tomita received the B.E. degree in precision engineering and the M.S. degree in human and engineered environmental studies from The University of Tokyo, Tokyo, Japan, in 2020 and 2022, respectively, where he is currently.

Seiichi Takamatsu received the B.E., M.E., and Ph.D. degrees in mechanical informatics from The University of Tokyo, Japan, in 2003,2005, and 2009, respectively. He joined the National Institute of Advanced Industrial Science and Technology (AIST) in 2009, where he was a Researcher with the Research Center for Ubiquitous MEMS and Micro Engineering (UMEMSME), AIST, in 2016. He is currently an Associate Professor with The University of Tokyo. His research interests include wearable MEMS technologies and meter-scale electronic textiles.

Toshihiro Itoh received the B.E., M.E., and Ph.D. degrees from The University of Tokyo, Japan, in 1988, 1990, and 1994, respectively. From 1994 to 2007, he was an Assistant Professor, a Lecturer, and an Associate Professor with The University of Tokyo. From 2007 to 2015, he was a Group Leader and the Deputy Director of the Research Center, AIST, Japan. Since 2015, he has been a Professor with The University of Tokyo. His research interests include MEMS and environment monitoring systems.