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RESEARCH ARTICLE

Pre-Strain Method for Stretchable Strain Sensors With Linear Sensitivity Using Acrylic Elastomer and CNTs Powder

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ABSTRACT Stretchable sensors can measure large strains generated in the joints of the human body. Generally, in the characteristics of carbon nanotubes based stretchable resistive sensors, the reference resistance increases with repeated strain, and the gauge factor which indicates sensitivity is not constant. These characteristics are related to the rupture of the CNT networks. This study proposes a pre-strain method to produce the resistive strain sensor with linear sensitivity. In this method, since the CNT network is formed on a strained elastomer, we expected that the CNT networks withstand the strain. By observing the electrodes, we discovered that this method aligned the agglomerated CNTs, changed the density of the CNT, and strengthened the networks. The pre-strain method is simple because it requires just a brush and stretching jig without special equipment. The sensor is completed by applying CNT powder to an elastomer that has been pre-strained 47% in the uniaxial direction and then returning it to its initial state. In the strain from 0% to 100%, the GF of the sensor is 1.46 constantly, and the variance of the normalized change in resistance of the sensor is less than 0.06. Therefore, the sensibility of the sensor has strong linearity. This result solves the problem of non-linear sensitivity of sensors using CNT powder in previous research. These features of constant gauge factor and reference resistance are expected to simplify the system's data processing.

INDEX TERMS Stretchable sensors, carbon nanotubes, pre-strain, normalized change in resistance, gauge factor, coefficient of variation.

I. INTRODUCTION

Wearable devices are attracting researchers' attention in the fields of nursing care and sports [1], [2]. The body movements of athletes and people who need care can be monitored to improve the efficiency of exercise and rehabilitation methods [3], [4]. Stretchable sensors can withstand a large strain and effectively measure body motion [3], [4], [5]. Recent studies have improved the gauge factor (GF), responsibility, and flexibility of wearable sensors [6], [7], [8], [9].

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In this study, we focus on stretchable resistive sensors. At its simplest, a stretchable resistive sensor consists of a flexible electrode on an elastomer. When an external force is applied to the elastomer, the flexible electrodes deform according to the magnitude of the force. Because the resistance of the electrodes is correlated with the strain of the elastomer, the strain can be measured electrically [10]. The GF , which is the sensitivity parameter of stretchable resistive sensors, is expressed as the normalized change in resistance with respect to strain. Equation (1) defines the GF :

$$GF = \frac{\partial \Delta R}{\partial \varepsilon R_0}, \quad (1)$$

where ε , ΔR , and R_0 are the uniaxial strain, change in resistance, and reference resistance, respectively. An ideal sensor should be able to measure large strains exceeding 100% and should exhibit a high GF value; in addition, the relationship between the normalized change in resistance ($\Delta R/R_0$) and the strain should be linear [6], [7], [9]. This linearity contributes to the reduction of data correction in the system. The GF of resistive and capacitive sensors have been compared [6], [9]. The GF of stretchable capacitive sensors is theoretically 1 and is constant [6]. The GF of stretchable resistive sensors varies with the material and shape of the electrodes, enabling better sensitivity than capacitive sensors [8], [11], [12]. In addition, when integrating a system, the circuit layout using a voltage divider of a resistive sensor is simpler than that of a capacitive sensor.

Our previous work developed a stretchable resistive sensor by applying carbon nanotubes (CNTs) powder to elastomers using a brush [8]. The reference resistance of the CNT powder electrode increases with repeated strain [8]. This characteristic has also been observed for electrodes created by mixing CNT powder with polymers and has been attributed to the rupture of the CNT networks [13], [14]. As shown in Fig. 1(a), when the sensor is subjected to strain, the CNT networks rupture and do not recover when the sensor returns to its initial shape. We speculated that increasing the density of CNTs would suppress the rupture of the CNT networks. Therefore, we searched for a method to strengthen the CNT network. In previous research, carbon black (CB) powder was applied to elastomers under pre-strained conditions to fabricate flexible electrodes [15]. CB powder acts as an electrode for a dielectric elastomer actuator (DEA) under large strains of the elastomer. We expected that, if CNT powder is applied to a pre-strained elastomer, networks would be strengthened, as shown in Fig. 1(b). Since the CNT network is formed on a strained elastomer, the CNT network adapts to the strain applied to the sensor. This is an expected mechanism of the stretchable resistive sensor that is less prone to rupture of the CNT networks.

This study proposes a pre-strain method to produce a sensor with a constant GF greater than 1. The sensor is completed by applying CNT powder to an elastomer under pre-strain and returning it to its initial state (0% strain). This solves the problem of the sensor with nonlinear sensibility in our previous work [8]. We indicate optimal pre-strain levels by investigating the relationship between pre-strain levels and the GF . As a demonstration, we show that mounted sensors onto a wearable device for the knee joint. In the static experiments, we confirm the GF to be constant when the device is attached to a human body. In the dynamic experiments, we derive the responsiveness of the sensor.

II. MATERIALS AND FABRICATION

A. MECHANICAL CHARACTERISTICS

The elastomer materials used in stretchable sensors are primarily acrylic rubber [16], [17] and silicone rubber [6], [8], which are polymerized by heat [6], [8] or ultraviolet

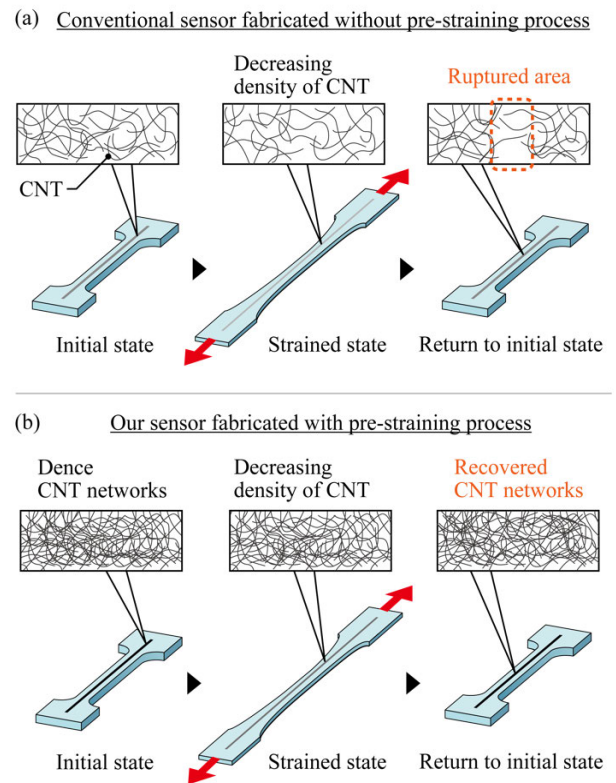


FIGURE 1. The state of the CNT networks: (a) the conventional sensor fabricated without the pre-straining process; (b) our sensor fabricated using the pre-straining process.

(UV) light [18], [19]. Typical elastomers exhibit mechanical hysteresis characteristics [20], [21]. When a dynamic load is applied to a sensor, such as during the measurement of body movements, a large hysteresis will result in the sensor value not returning to its reference value (at 0% strain) when the load is removed [22]. Therefore, elastomers with low hysteresis are suitable materials for sensors that capture dynamic strain [23]. With respect to integration into devices, acrylic rubber, which exhibits strong adhesion with common-oriented polypropylene (OPP) tape, is desirable. We evaluated the mechanical hysteresis properties of two acrylic rubbers: Suave-10F100 (Osaka Organic Chemical Industry, Kashiwara, Osaka, Japan) and VHB (VHB Y-4905J, 3M, Maplewood, MN, USA). The elastomer for the mechanical experiment follows dumbbell-shaped specimens (JIS K6251 Dumb-bell test pieces No. 6). The tensile test apparatus used in the present study is the same as that we previously reported [8], [24]. To investigate stress-strain characteristics, the experimental setup consisted of a tensile test apparatus with a speed of 5 mm/s and a displacement of 50 mm; the test was conducted for 50 cycles. Fig. 2 shows the stress-strain characteristics of Suave-10F100 and VHB. In Fig. 2(a), the strain value of Suave-10F100 at a stress of 0 MPa drifted from 1% to 9% between the 1st and 50th cycles. In Fig. 2(b), when VHB was under 0 MPa stress, the strain value of VHB drifted from 32% to 56% between

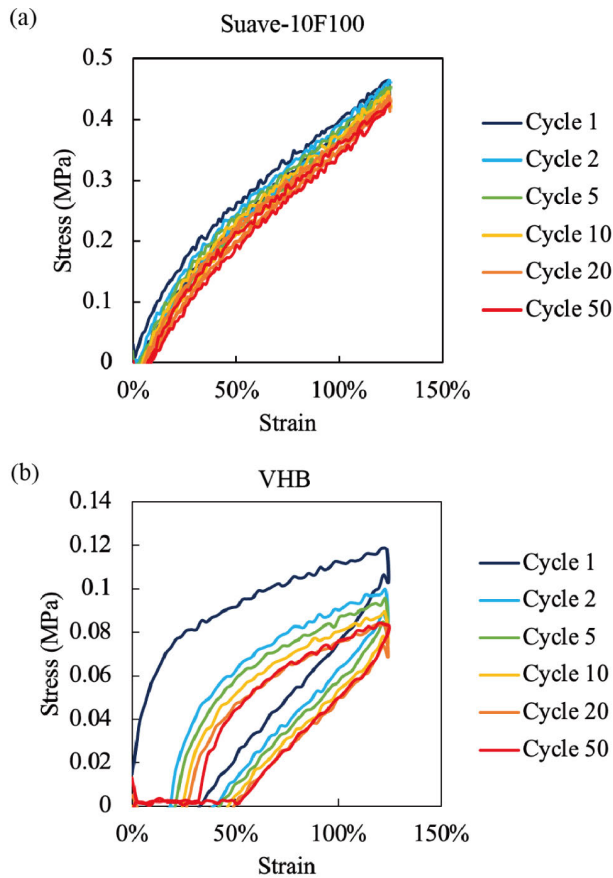


FIGURE 2. Strain-stress characteristics of (a) Suave-10F100 and (b) VHB.

the 1st and 50th cycles. Suave-10F100 shows less change in stress-strain curve characteristics with cyclic strain than VHB in the 0-125% strain range. This characteristic difference suggests that Suave-10F100 requires a shorter relaxation time to recover its initial state than VHB. Hence, Suave-10F100 has less hysteresis than VHB. Therefore, we conclude that Suave-10F100 is more suitable for use in stretchable strain sensors than VHB, especially at higher strain levels (in the strain range of 0-125%).

B. ELECTRICAL CHARACTERISTICS

The surface stickiness of the elastomer, which leads to strong adhesion of electrode material and elastomer, is also related to the sensor’s electrical properties [8]. We show the difference in electrical properties of the sensor when using Suave-10F100 and VHB respectively. In this study, we selected multiwalled CNTs (MWCNTs, No. 724769-25G, Sigma-Aldrich) with an outer diameter of 6–9 nm and a length of 5 μm as an electrode. This CNT is the same that we adopted in previous works [8]. For the cyclic electrical tensile test, we prepared the specimens. We applied CNT powder to the Suave-10F100 and VHB elastomers under 0% pre-strain. The shape of the elastomer for the electrical experiment also follows dumbbell-shaped specimens (JIS K6251 Dumb-bell

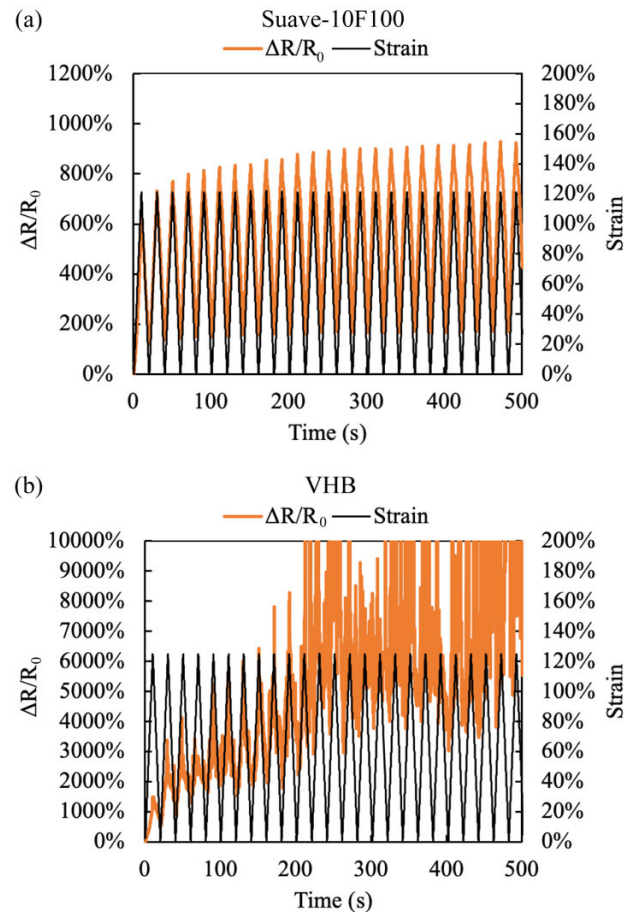


FIGURE 3. Normalized change in resistance and strain of the sensors with 0% pre-strain: (a) Suave-10F100 and (b) VHB.

test pieces No. 6). The length and width of the electrode were 50 mm and 2 mm, respectively. We then investigated the relationship between the strain and the normalized change in resistance ($\Delta R/R_0$) in a cyclic electrical tensile test. We set the experimental conditions as follows: the tensile speed was 5 mm/s, the maximum tensile displacement was 50 mm, and the number of tensile testing cycles was 25. Fig. 3 shows the relationship between strain and the normalized change in resistance during the cycle tensile test. As shown in Fig. 3(a), the normalized change in resistance of the Suave-10F100 sensor was synchronized with the strain during the 25 testing cycles. However, in Fig. 3(b), the VHB sensor showed poor synchronization between the normalized change in resistance and the strain. In this case, VHB exhibited poor performance as a strain sensor. We assumed that the stickiness of VHB made the CNT networks more prone to rupture. Based on these results, Suave-10F100 is more suitable than VHB for use in a CNT-based stretchable strain sensor.

C. FABRICATION FOR THE SENSOR

We here explain the elastomer processing method and the brushing process used to prepare electrodes for the tensile

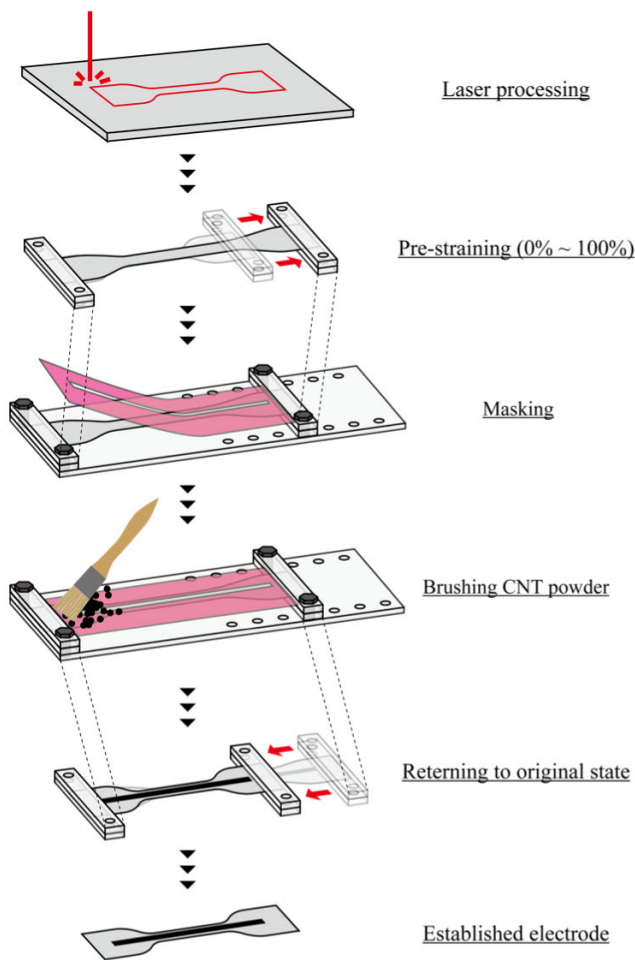


FIGURE 4. Schematic of the fabrication for the stretchable resistance sensors with the pre-straining process.

test to investigate the effects of pre-strain on the sensor’s electrical properties. Fig. 4 shows the fabrication process. We fabricated sensors that adjusted the density of the CNTs by cutting the elastomer with a laser cutting machine (Trotec, Speedy 100), pre-straining the elastomer in the uniaxial direction with a jig, masking the elastomer with a release film (silicone separator PET77 × 1-CRD-A3, Nippa), applying CNT powder with a brush, and then restoring the elastomer to its initial state. In the case of conventional sensors, 0% pre-strain is applied [8].

III. EXPERIMENT

A. PRE-STRAIN AND CNTS NETWORK

We measured the reference resistance of the nine sensors in their initial geometry (0% strain) to determine the pre-strain process’s effect on the electrodes’ electrical properties. Fig. 5(a) shows nine sensors fabricated with 0% (conventional type), 7%, 20%, 33%, 47%, 60%, 73%, 87%, and 100% pre-strain. As shown in Fig. 5(b), the reference resistance of the sensor at 0% pre-strain was 1.8 MΩ, and at 100%

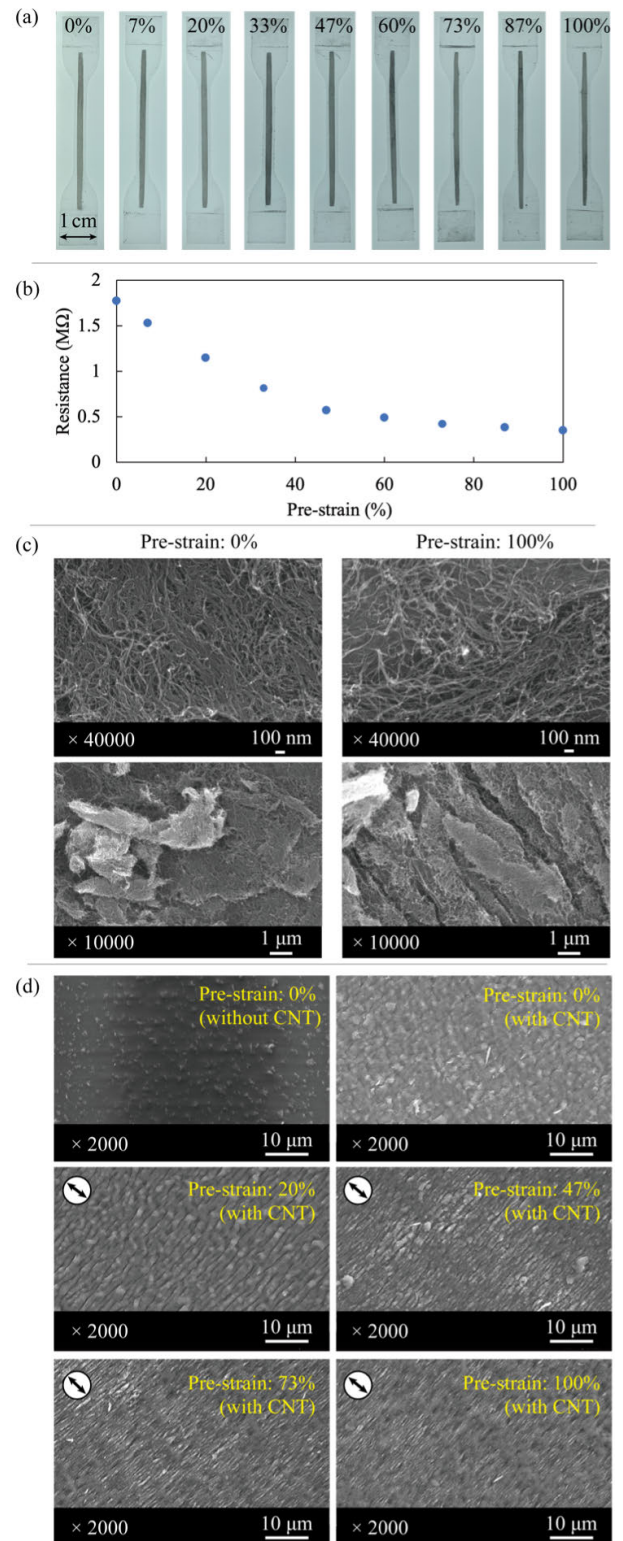


FIGURE 5. Relationships between reference resistance and pre-strain: (a) nine sensors fabricated 0-100% pre-strain; (b) reference resistance and pre-strain; (c) CNT networks at the 0% and 100% pre-strain; (d) the state of the agglomerated CNTs from 0% to 100% pre-strain.

pre-strain was 0.4 MΩ. From 0 to 47%, the reference resistance decreases rapidly; and over 47%, it decreases gently.

When the CNT network is strengthened, the number of CNTs per unit area should increase. To confirm the state of the CNTs on the elastomer, we observed the surface layer of the sensor using field-emission scanning electron microscopy (FE-SEM JSM-7610F, JEOL) in Fig. 5 (c) and (d). Compared to the image of 0% and 100% pre-strain at the $\times 40000$ magnification, the CNT network is randomly connected, and the density difference cannot be confirmed. We then observed each electrode at the $\times 10000$ magnification and discovered agglomerated CNTs are placed at 0% pre-strain and are aligned at 100% pre-strain. Fig. 5(d) shows the electrode surface fabricated with 0-100% pre-strain at $\times 2000$ of the magnification. Agglomerated CNTs are randomly placed throughout the image in the area coated with CNT at 0% strain. Pre-strained directions are shown on the left-top side in the 20, 47, 73 and 100% pre-strain images. In each image, the streaks that are perpendicular to the pre-strained direction are generated. Since these streaks become finer as the pre-strain increases, we presume that the amount of CNT applied per unit area increases.

Based on these results, we expected that the pre-strain method would adjust the alliance of the agglomerated CNTs, thereby strengthening the CNT networks at the connected points between agglomerated CNTs. Therefore, the pre-strain method in previous research solves the problem of non-linear sensor sensitivity using CNT powder.

B. OPTIMIZATION OF THE PRE-STRAIN

We investigated the relationship between the repeated strain and the electrical properties of three sensors fabricated with 0%, 47%, and 100% pre-strain. The stroke applied to the sensor was 50 mm (125% maximum strain), the velocity in the z-axis direction was 5 mm/s, and the number of cycles was 20.

Fig. 6 shows (a) the relationship between the number of cycles and the reference resistance, (b) the normalized change in resistance as a function of strain, (c) the coefficient of variation (CV: the ratio between the standard deviation and the mean) of the normalized change in resistance as a function of strain, (d) GF as a function of strain.

As shown in Fig. 6(a), the sensor with 0% pre-strain did not converge to a constant value, and 47% and 100% pre-strain converged to 0.6 M Ω and 0.4 M Ω after the second cycle, respectively.

Conventional sensors with 0% pre-strain showed large variations in the normalized change in resistance, with a maximum of 1189% and a minimum of 663% at 125% strain (Fig. 6(b)). On the same strain, the data of the 47% and 100% pre-strain appear to be less variability than that of the 0% pre-strain.

To investigate variability, we derived the CV of the normalized change in resistance (Fig. 6(c)). The CV of the 47% pre-strained sensor was consistently less than 0.1 in the strain range from 0% to 120%. This characteristic means that it is more reliable than the sensors with 0% and 100% pre-strain. The CV of the 0% pre-strained sensor was higher than 0.1 in

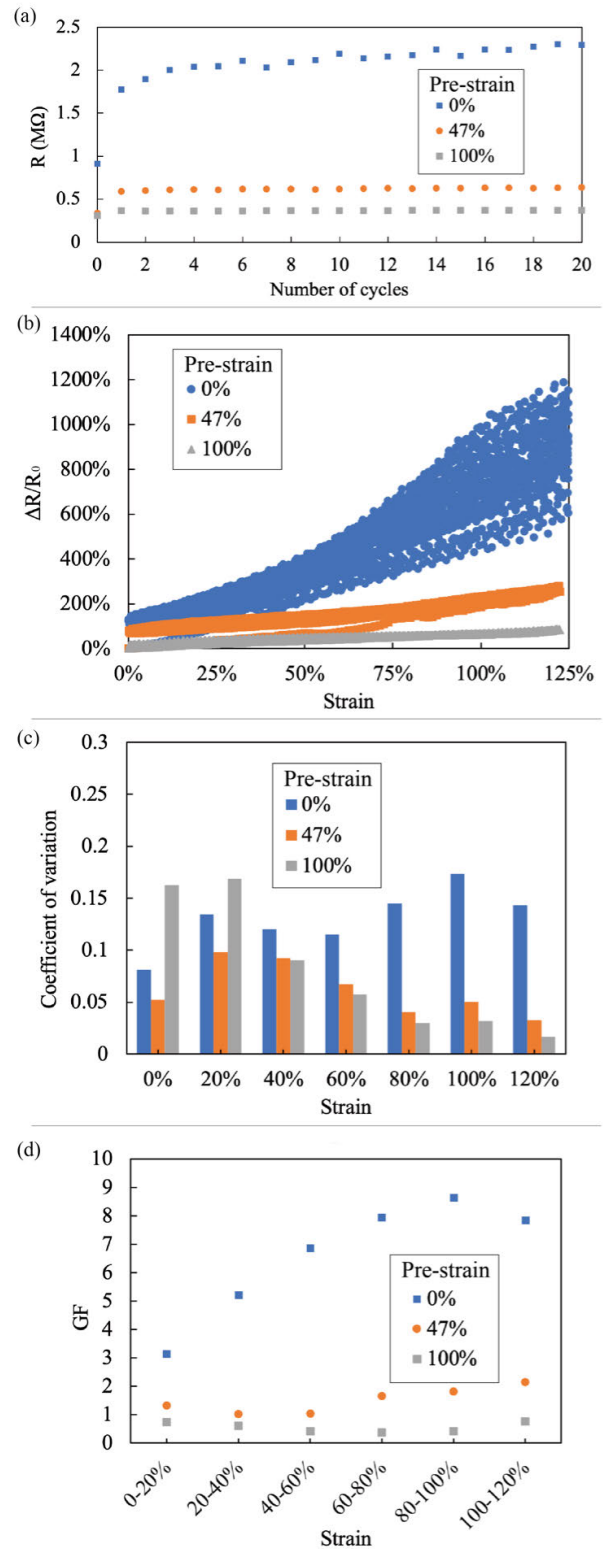


FIGURE 6. Electrical properties of the sensors: (a) the number of cycles and the reference resistance; (b) resistance as a function of strain; (c) coefficient of variation as a function of strain; (d) gauge factor as a function of strain.

the strain range from 20% to 120%. We assumed that this phenomenon is similar to that observed for sensors prepared

TABLE 1. Performance comparison of the resistive strain sensor.

Ref.	Coating method	Materials	GF	Linearity
This study	Brushing	CNT powder/ Acrylic rubber	1.46	Linear
[8]	Brushing	CNT powder/ PDMS	6.2- 8.2	Nonlinear
[6]	Bar coating	CB solution/ Ecoflex	1.62- 3.37	Nonlinear
[24]	Spray coating	CNT solution/ Ecoflex	1.75	Linear
[25]	Soak in solution	CNT solution/ TPU	1.24, 1.67	Nonlinear
[26]	3D printing	Carbon grease/ Ecoflex	3.8	Nonlinear
[27]	Coating	CNT solution/ PDMS	1.8	Nonlinear
[28]	Spray coating	CNT film/ polyurethane	6.89	Linear

using conductive polymers [15], [16] and was caused by the rupture of the CNT networks. Even for the 100% pre-strained sensor, the CV was higher than 0.1 at 0% and 20% strain. We assumed that this phenomenon was due to an excessively high density of CNTs, which over-strengthened the CNT networks. The over-strengthened CNT networks on the elastomer led to the stretchable sensor’s low sensitivity, and the sensor’s resistance change became smaller than the measurable range of the measuring instrument.

We subsequently calculated the GF by averaging the normalized change in resistance for each 20% strain (Fig. 6(d)). The GF values were 3.1 minimum and 8.7 maximum for the 0% pre-strained sensor, 1.0 minimum and 2.1 maximum for the 47% pre-strained sensor, and 0.36 minimum and 0.77 maximum for the 100% pre-strained sensor. Because the GF was calculated by averaging the normalized change in resistance, the sensor with a smaller CV has a more reliable GF.

Based on these results, we expected that a sensor with 47% pre-strain would be promising as a sensor in the strain range from 0% to 120% because the CV was less than 0.1, and the GF was greater than 1. Therefore, we assumed that 47% is the optimal pre-strain.

C. LAMINATION PROCESS OF CNT ELECTRODES

When a CNT electrode is exposed to the atmosphere, collisions with objects might change its properties. Therefore, covering the electrode with elastomers prevents physical external interference. As the sensor’s electrical properties with 47% pre-strain were promising, we, therefore, used two sensors with 47% pre-strain and assembled two sensors with the electrodes facing each other. Fig. 7 shows (a) the structure of the stretchable sensor with the protected CNT electrode, (b) the photo of the stretchable sensor, (c) the normalized change in resistance and strain for the three samples, and (d) the average of the normalized change in resistance and strain for three samples.

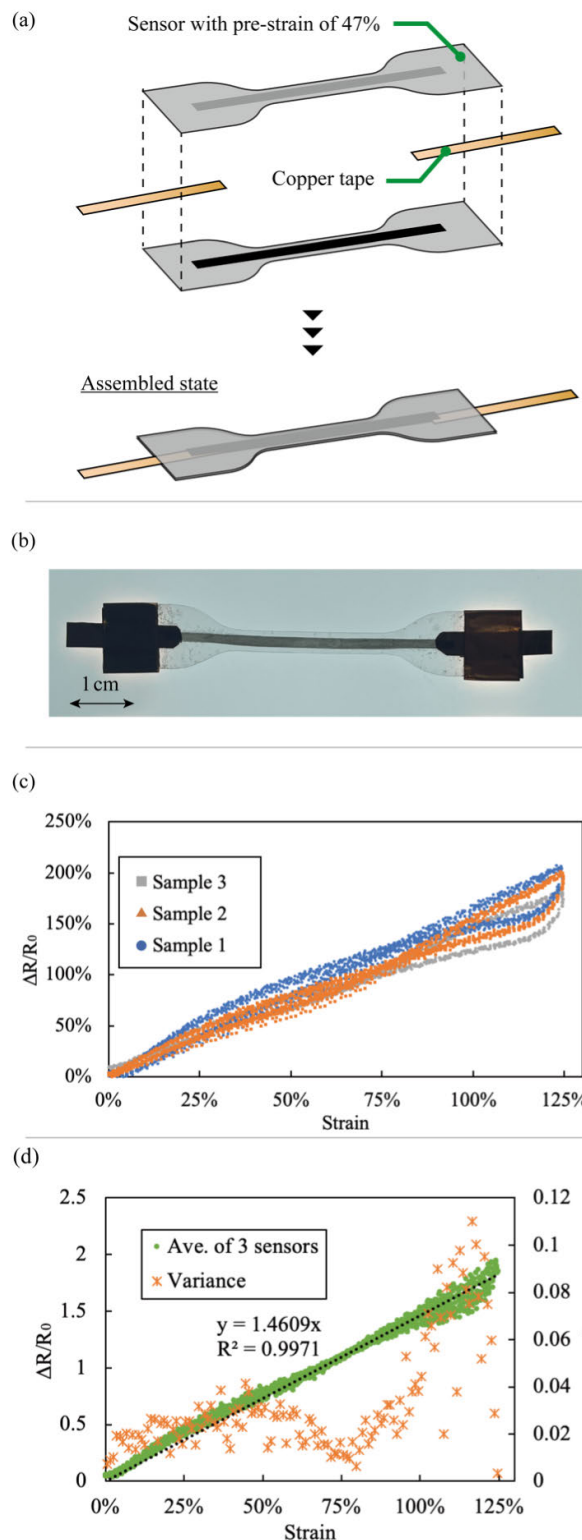


FIGURE 7. Sensor with protected CNT electrode and characteristics: (a) structure of the sensor; (b) photo of the sensor; (c) normalized change in resistance as a function of strain; (d) average normalized change in resistance and variance as a function of strain.

When laminating the two sensors, we sandwiched copper tape between the two sensors to transmit electrical

information from the internal CNT electrode to the exterior (Fig. 7(a)). Both ends of the electrode overlap the copper tape by 5 mm. As applying a strain of 125%, the normalized change in resistance gradually increased (Fig. 7(c)). The approximate line derived from the average of the three sensors was $y = 1.4609x$ (Fig. 7(d)). Thus, the GF of the sensor with a protected CNT electrode is 1.46. In this data, the variance is less than 0.06 in the strain from 0% to 100%, so the characteristic of the sensor shows strong linearity in this range of strain. Furthermore, as the value of GF (1.46) is greater than a capacitive sensor, our sensor has great sensitivity.

We compare our sensor and other study's sensor in terms of material, electrode fabrication, and linearity of sensibility. Table 1 shows the performance comparison of the resistive strain sensor. As a result of investigating previous research, there were few studies in which electrodes were formed using powder, and most of them were formed using solutions. We realized a sensor with linear sensitivity using the same CNT as the previously reported sensor [8]. Applying CNT powder directly to the elastomer with a brush is simple and does not require a spray gun or 3D printing machine like other methods. In addition, since acrylic rubber is used for the base, our sensor can be easily attached to the application with general OPP tape. Furthermore, to realize constant GF , our pre-stretching method can easily strengthen the CNT networks without the process of dispersing the electrode material in a solvent or mixing it with a polymer.

IV. WEARABLE DEVICE

We developed a wearable device to measure knee joint motion to evaluate the performance of the sensor. Fig. 6(a) shows the circuit diagram for obtaining the value from the stretchable resistive sensor. We used a 470 k Ω resistor for the voltage divider circuit. We also used a 470 μ F electrolytic capacitor and a 100 nF ceramic capacitor for noise reduction. The microcontroller was a Tiny PICO, which could be operated remotely. The total size of the electric circuit (including the Tiny PICO, the voltage divider, and the capacitor) was 30 mm \times 30 mm. The battery was a 3.7 V Li-po battery with a capacity of 600 mAh. The sensor was magnetically connected to the supporter, which facilitated sensor mounting. The contact resistance between the sensor and the sensor mounting was less than 10 Ω . In this case, we expected low noise in the sensor reading. The weight of the wearable device with a sensor unit is 93.6 g.

Since the sensor may be damaged if excessively large strain is applied, we designed the wearable device so that when the knee angle is 90, the strain applied to the sensor is approximately 75%. Therefore, in the demonstration, a strain of less than 125% was applied to the sensor, and the experiment was performed within a range where the sensor performance was stable.

We performed a static experiment in which the knee angle of a person wearing the device was changed from 0 $^\circ$ to an arbitrary angle and, after 7 s, was returned to 0 $^\circ$. The knee

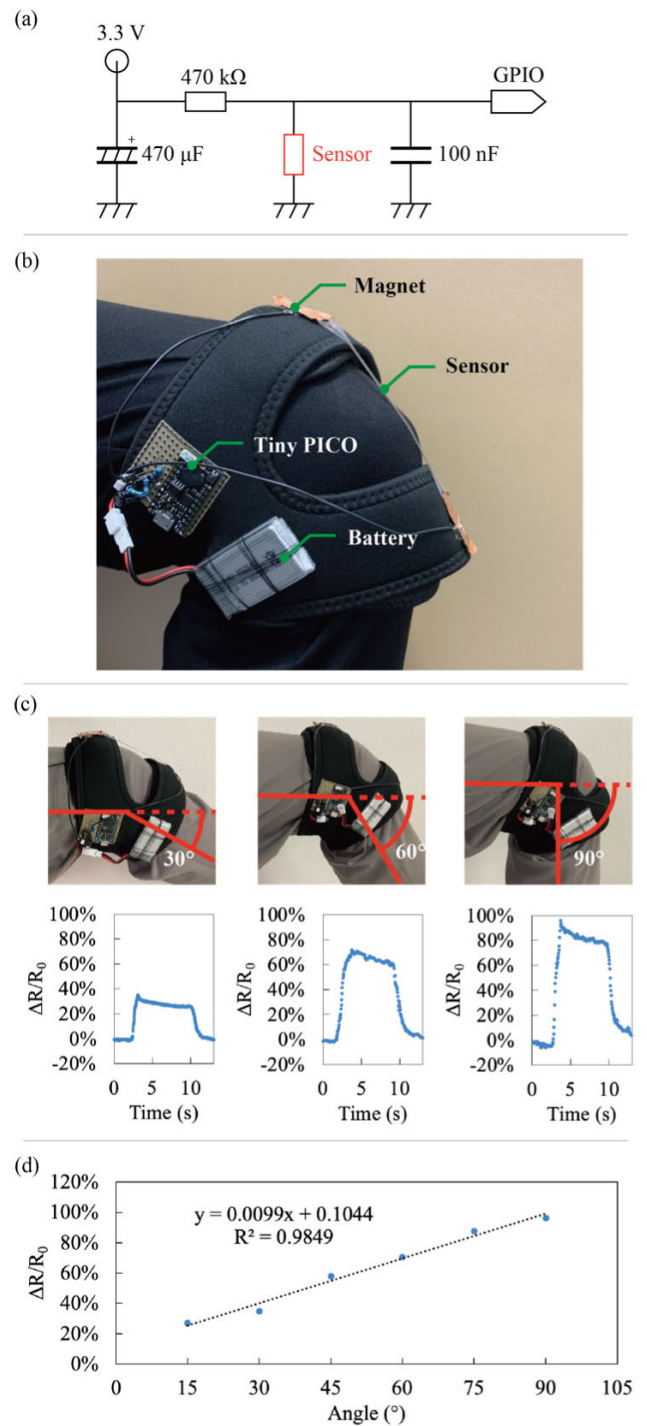


FIGURE 8. The static experiment of the wearable device for knee joint: (a) circuit diagram; (b) attached wearable device; (c) static experiments of bending the knee joint (30 $^\circ$, 60 $^\circ$, and 90 $^\circ$); (d) normalized change in resistance as a function of the static knee angle.

angles were 15 $^\circ$, 30 $^\circ$, 45 $^\circ$, 60 $^\circ$, 75 $^\circ$, and 90 $^\circ$ (Video S1). As shown in Fig. 6(c), when the knee was bent, the data showed transient characteristics. Such transient properties were observed in previous studies involving conductive polymer electrodes and were attributed to the recombination of

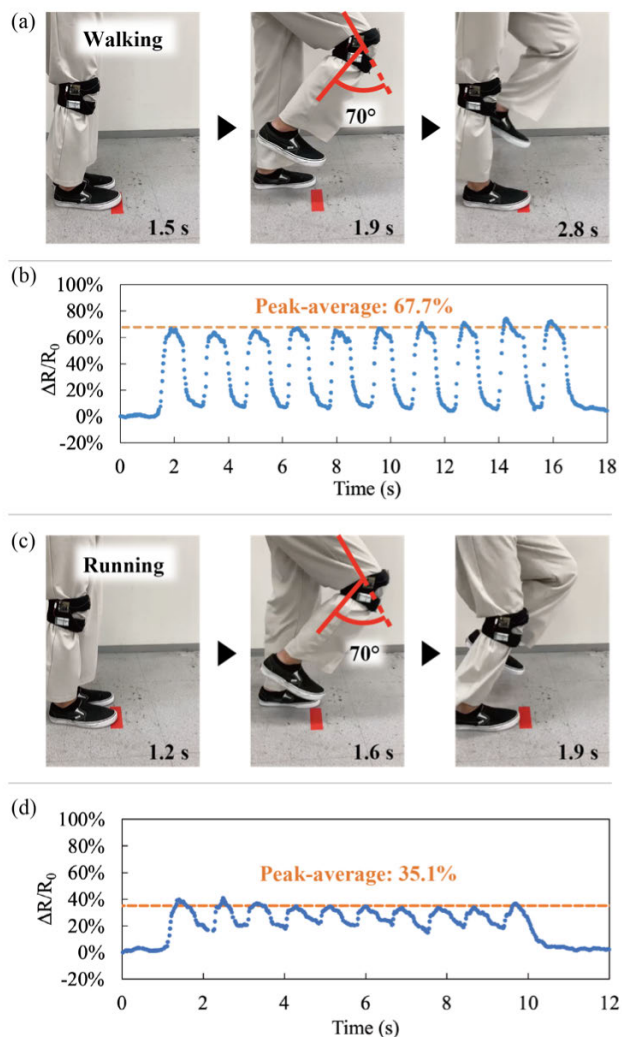


FIGURE 9. The dynamic experiment of a wearable device for knee joint: (a) walking appearance; (b) walking data; (c) running appearance; (d) running data.

the CNT networks [13]. This phenomenon caused a gradual decrease in the normalized change in resistance as a function of time. We assumed that a similar recombination of the CNT networks occurred in our sensor. When the knee angle was returned to 0° , the curve of the normalized change in resistance gradually returned to its reference value. This gradual returning characteristic is caused by the mechanical hysteresis of the elastomer. The experimental data in Fig. 6(c) show that the knee joint angle was changed by 15° , and the peak value of the normalized change in resistance increased linearly as the joint angle increased. This means that the GF is constant since the joint angle is proportional to the strain of the sensor.

In the dynamic experiment, we obtained sensor values as a person wearing the sensor walked with a 1.6 s cycle and ran with a 1.0 s cycle. The knee bending angle for both experiments was 70° , as shown in Fig. 7(a) and (c). The number of bending cycles was 10. As shown in Fig. 7(b),

during walking, the average cycle peak of the normalized change in resistance was 67.7%, and the CV of the peak was 0.051. As shown in Fig. 7(d), during running, the average cycle peak value of the normalized change in resistance was 35.1% and the CV of the peak was 0.084. According to the results of the static experiments, the normalized change in resistance at a knee angle of 70° is 80%, as indicated by the approximate line in Fig. 6(c). In the dynamic experiments, the normalized change in resistance was lower than expected by 12.3% for walking and 44.9% for running. Therefore, the peak of the standard resistance change tends to be lower as the period becomes shorter. In the running experiment, when the knee bending angle was changed from 70° to 0° , the normalized change in resistance decreased to 18.6% and did not reach 0%.

These sensor characteristics are mainly attributable to the next movement being initiated before the elastomer's residual stresses were relaxed. In the dynamic experiment, the CV of the normalized change in resistance was less than 0.1. After 10 cycles, the normalized change in resistance returned to 0%.

We inferred that the effect of CNT networks' rupture was small even when the sensor was attached to the body. We can clearly confirm the 10 bending cycles of the knee joint by observing the data during walking and running. In the running experiment, the sensor deformed at 122.1 mm/s, which is the maximum deformation rate confirmed in this work.

V. DISCUSSION AND CONCLUSION

In this study, we propose a pre-strain method to produce the stretchable resistive sensors with a constant GF of 1.46 in the strain from 0% to 100%. In this method, CNT powder was applied to 47% pre-strained elastomer that was subsequently returned to its original state (0% strain). By this fabrication, the agglomerated CNTs are aligned, and their density increase, and consequently, the CNT network is strengthened. Hence, rupture of the CNT networks is less prone to applied strain with the sensor. The pre-strain method is simple because it requires just a brush and stretching jig without the splay, bar-coater, or 3D printer. Using the same CNT as the previously reported sensor with nonlinear sensitivity [8], we realized a sensor with linear sensitivity. In addition, since acrylic rubber is used for the base, our sensor can be easily attached to the application with general OPP tape. As the demonstration, we mounted the sensor to a wearable device that can measure knee joint motion and performed static and dynamic experiments. In the static experiment, the normalized change in resistance was found to increase linearly as the joint angle increased. This means that the GF is constant. In dynamic experiments, the sensor clearly captured 10 bending cycles of the knee joint during walking and running. In especially, the running experiment revealed that the sensor tolerates deformation velocities of up to 122.1 mm/s. Because the CV of each peak value of the normalized change in resistance was small, and the normalized change in resistance returned to 0% after 10 cycles, we inferred that the effect of CNT networks' rupture was small. The results show that

our sensor is suitable for incorporation into wearable devices, especially those for capturing knee joint motion.

By using stretchable resistive sensors with a constant GF and with a small variance, we can simplify the wearable device system and eliminate the cumbersome process of calibration when the device is attached to a person's body. Our method can contribute to the improvement of powder-based stretchable electrodes. In addition, it is believed that our stretchable electrode can be incorporated into other stretchable devices such as dielectric elastomer actuators [29], [30], [31], EHD pumps [32], [33], [34], [35], and electrostatically adhesive pads [36]. In the future, our sensors could contribute to the soft robots equipped with these soft devices.

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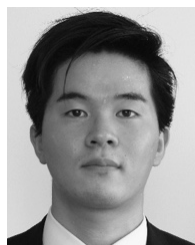
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