

## MPTMS Treated Au/PDMS Membrane for Flexible and Stretchable Strain Sensors

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### Abstract

Au/PDMS membranes are widely used to fabricate strain sensors which can detect input signals. An interfacial adhesion between metal films and polydimethylsiloxane (PDMS) substrates is one of the important factors determining the performance of strain sensors, in terms of robustness, reliability, and sensitivity. Here, we fabricate Au/PDMS membranes with (3-mercaptopropyl) trimethoxysilane (MPTMS) treatment. PDMS membranes were fabricated by spin-coating and the thickness was controlled by varying the spin rates. Au electrodes were deposited on the PDMS membrane by metal sputtering and the thickness was controlled by varying sputtering time. Owing to the MPTMS treatment, the interfacial adhesion between the Au electrode and the PDMS membrane was strengthened and the membrane was highly transparent. The Au electrode, fabricated with a sputtering time of 50 s, had the highest gauge factor at a maximum strain of ~0.7%, and the Au electrode fabricated with a sputtering time of 60 s had the maximum strain range among sputtering times of 50, 60, and 120 s. Our technique of using Au/PDMS with MPTMS treatment could be applied to the fabrication of strain sensors.

**Keywords:** Gold (Au), Polydimethylsiloxane (PDMS), Flexible strain sensor, (3-mercaptopropyl) trimethoxysilane (MPTMS)

### 1. INTRODUCTION

Recently, the development of flexible strain sensors has received a lot of attention and various strain sensors with high sensitivity have been developed. These electrically conductive sensors capable of bending and stretching are used for flexible displays [1-3], field-effect transistors [4,5], energy-related devices [6,7], smart clothing [8], and actuators [9-11]. However, the development of mechanosensors having ultra-high mechanosensitivity, flexibility, and durability still remains a challenge.

In previous studies, cracks in conducting materials were regarded as traits of failure for electrical interconnections and their potential for application in strain sensing was neglected. However, in the nature world, spiders have crack-shaped slit organs which are extremely sensitive to surrounding vibrations [12]. Recently, several studies showed that micro [13,14] or nanocrack [15]-based

mechanosensors are ultrasensitive to external strain.

Polydimethylsiloxane (PDMS) is widely used as the membrane material fabricate flexible devices because it is optically transparent, biocompatible, flexible, and easy to process. For flexible strain sensors, a strong interfacial adhesion between the thin metal film and the PDMS substrate is very important to fabricate a robust, reliable, and highly sensitive device [14]. This is because a thin metal film with good interfacial adhesion on a flexible membrane can experience significant structural changes upon stretching without any sliding or delamination. Several methods to enhance the interfacial adhesion have been introduced using titanium (Ti) or chromium (Cr) as an adhesion interlayer [16,17] and thermally curing a prepolymer of PDMS on gold (Au) electrodes with a Ti interlayer [18]. However, Ti or Cr layers can affect the optical and electrochemical properties of the device and these materials are not suitable for bioapplications [19-21]. However, a molecular adhesive of (3-mercaptopropyl) trimethoxysilane (MPTMS), as a self-assembled monolayer (SAM) can avoid the problems caused by using metallic interlayers. However, until now, no study has tried to exploit the electromechanical characteristics of MPTMS treated Au/PDMS membranes for use as a strain sensor.

In this study, flexible strain sensors were developed using PDMS membrane coated by a thin Au film. Au was deposited on the PDMS membrane using metal sputtering and the interfacial adhesion between Au film and PDMS was enhanced by MPTMS

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treatment. The electromechanical characteristics that the fabricated Au/PDMS membranes show can be used as a sensing mechanism for strain sensors. The results show that the Au/PDMS strain sensors had a superior transparency and a very high gauge factor.

## 2. EXPERIMENTAL

### 2.1 Chemicals

The following chemicals were used in this paper:

- (1) PDMS: Sylgard 184 (Dow Corning, USA)
- (2) Si substrate: <100> bare Si wafers (Silicon Technology Corp., USA)
- (3) Au sputter: MCM-100 (SEC Co., Ltd, Korea) (Deposited thickness per minute: 20–25 nm)
- (4) PTFE: Teflon AF 601S1-100-6 (Dupont, USA)
- (5) DC-75: Acros Organics, Belgium
- (4) MPTMS:  $\text{HS}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$ , 95% purity (Sigma-Aldrich, USA)

All chemicals were used without further purification.

### 2.2 Fabrication of Au/PDMS membrane

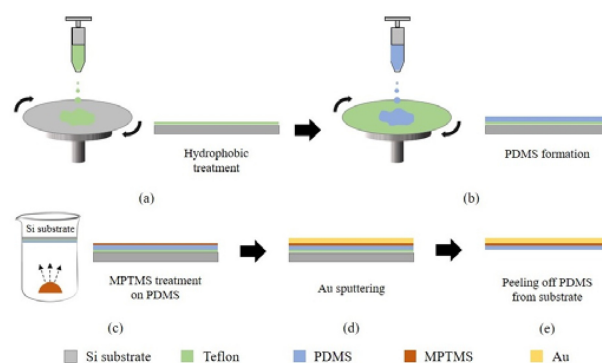
Manufacturing of the stretchable Au/PDMS membrane was carried out in 5 steps (Fig. 1). The fabrication process included hydrophobic treatment, PDMS formation, MPTMS treatment, Au sputtering, and peeling off the PDMS, as follows:

(1) A Si substrate was cleaned in acetone, rinsed in isopropanol, distilled (DI) water and dried with  $\text{N}_2$  gas. To give the Si surface hydrophobic characteristics, the Si substrate was coated with a 2% PTFE solution diluted in FC-75. The PTFE solution was spin-coated on the Si substrate for 50 s at a spin rate of 750 rpm and soft baked at  $100^\circ\text{C}$  for 10 min. Then, the substrate was fully dried for 1 day at room temperature.

(2) PDMS was prepared by mixing base silicone gel with a curing agent in a 10:1 ratio (by weight). The mixture of PDMS base/curing agent was poured on the Si substrate and spun at speeds between 400 and 700 rpm for 50 s to achieve the desired thickness.

(3) A 20  $\mu\text{L}$  MPTMS solution was placed in a beaker. The MPTMS was deposited on the PDMS surface by vacuum deposition.

(4) Au was deposited by sputter coating on the MPTMS surface.



**Fig. 1.** Sequential steps for fabricating flexible strain sensors. (a) PTFE solution spread on a Si substrate using spin coating, (b) PDMS solution spread on the substrate using spin coating, (c) MPTMS deposited on the PDMS surface by vacuum deposition, (d) Au deposited on the substrate using sputtering, (e) The fabricated flexible strain sensor peeled off from the substrate.

(5) Finally, the PDMS with the thin Au film was manually peeled off from the Si substrate. The conducting wires were connected with the Au film using a silver paste to make electrical contact.

The surface of the PDMS layer with a thin Au film was investigated by field-emission scanning electron microscopy (FE-SEM, JMS-7400F, JEOL, operating at 5 keV). The electrical resistance of the thin Au film on PDMS was measured by a voltage source/measure unit (B2902A, Keysight, minimum resolution of 100 fA, under a constant voltage of 1 V).

### 2.2 Thickness measurement and failure strain test of pure PDMS membrane

The PDMS membrane was fabricated by a spin-coating process and the membrane thickness was controlled by varying the spin rate between 400 and 700 rpm. The thickness of the fabricated PDMS membrane was measured with Vernier calipers, and the failure strain was measured by extending the PDMS membrane. The failure strain was calculated by the following equation (1). The final length was the length of the PDMS membrane when the membrane failed during extension.

$$\text{Strain}_{\text{Failure}} = \frac{L_{\text{Final}} - L_{\text{Initial}}}{L_{\text{Initial}}} \times 100 \quad (1)$$

### 2.3 Transmittance measurement of the Au/PDMS membrane

The transmittance of the fabricated Au/PDMS membrane was measured by a UV-vis spectrophotometer (2120us, OPTIZEN). The transmittance spectra were acquired at wavelengths ranging from 400 to 1,000 nm.

## 2.4 Conductance measurement of the Au/PDMS membrane under an applied strain

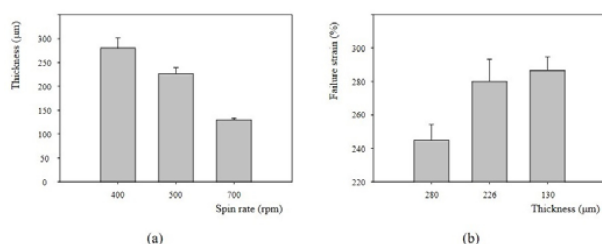
The strain applied to the Au/PDMS membrane was controlled with the help of micro-translation stage (M-112, Physik Instrumente). The micro-translation stage had a travel range of 25 mm with high resolution and the minimum moving distance was 0.05  $\mu\text{m}$ . The resistance change was measured using a voltage-source-measure equipment (B2902A, Keysight, minimum resolution of 100 fA, constant voltage of 1 V) using the current across the membrane.

## 3. RESULTS AND DISCUSSIONS

### 3.1 Thickness and failure strain of pure PDMS membrane

The thickness of the flexible membrane determines the transmittance and the failure strain. In this study, the flexible membrane was produced by a spin-coating process and the thickness of the fabricated membrane was controlled by varying the spin rates.

Fig. 2 shows the thickness and the failure strain of the pure PDMS membrane. With increasing spin rate, the PDMS membrane became thinner. The thickness was 280  $\mu\text{m}$  when the spin rate was 400 rpm, 226  $\mu\text{m}$  at 500 rpm, and 130  $\mu\text{m}$  at 700 rpm. Then, the failure strains of the three membranes were analyzed. The membrane having 280  $\mu\text{m}$  thickness could endure up to 245% strain without breaking, while membranes with thicknesses of 226  $\mu\text{m}$  and 130  $\mu\text{m}$  could endure up to 280% and



**Fig. 2.** Characteristics of as-is pure PDMS membrane. (a) The thickness of pure PDMS membrane vs. the spin rates, (b) The failure strain of pure PDMS membrane vs. the thickness of the pure PDMS membrane ( $n = 5$ , mean  $\pm$  standard error).

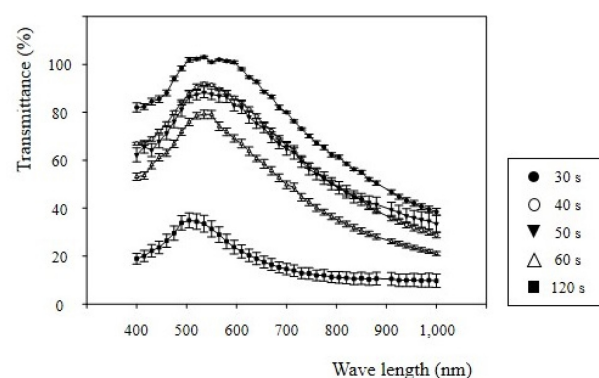
286% strain, respectively. The membranes with 226  $\mu\text{m}$  thickness were adopted to fabricate subsequent Au/PDMS membranes because of their high failure strain.

### 3.2 Transmittance of the fabricated MPTMS treated Au/PDMS membrane

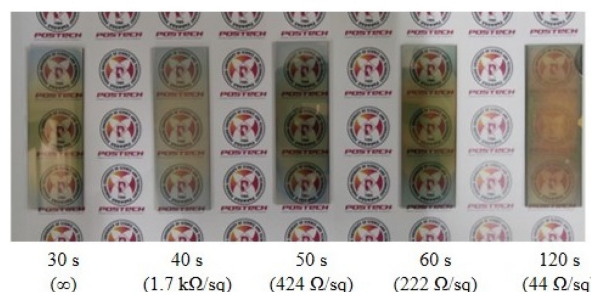
As described before, a strong interfacial adhesion between the



**Fig. 3.** Photograph of Au/PDMS membranes without MPTMS treatment. Blue arrow indicates that Au was fully detached from PDMS membrane. Red arrow indicates that Au film partially detached from the PDMS membrane.



(a)



(b)

**Fig. 4.** Transmittance characteristic of Au/PDMS membranes. (a) Transmittance spectra in the range of 400–1,000 nm for five Au sputter conditions ( $n = 3$ , mean  $\pm$  standard error), (b) Photographs of five Au/PDMS membranes with their corresponding sheet resistance specified.

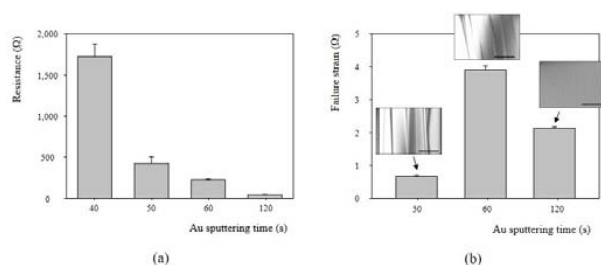
thin metal film and the flexible membrane is very important to have a robust, reliable, and highly sensitive sensing performance. Metallic interlayers are widely used for enhancing the interfacial adhesion, but they can decrease the optical and electrochemical properties of the device. MPTMS as a SAM can solve these problems caused by the metallic interlayers.

Fig. 3 shows a photograph of the Au/PDMS membranes (sputtering time: 30, 40, 50, 60, 120 s) without MPTMS treatment. Under all conditions, Au films were either fully (blue arrow) or partially detached (red arrow) from PDMS because of poor adhesion between the Au film and PDMS. Figure 4b shows Au/PDMS membranes with MPTMS treatment. Au films strongly adhered to PDMS because MPTMS enhanced the interfacial adhesion between Au films and PDMS.

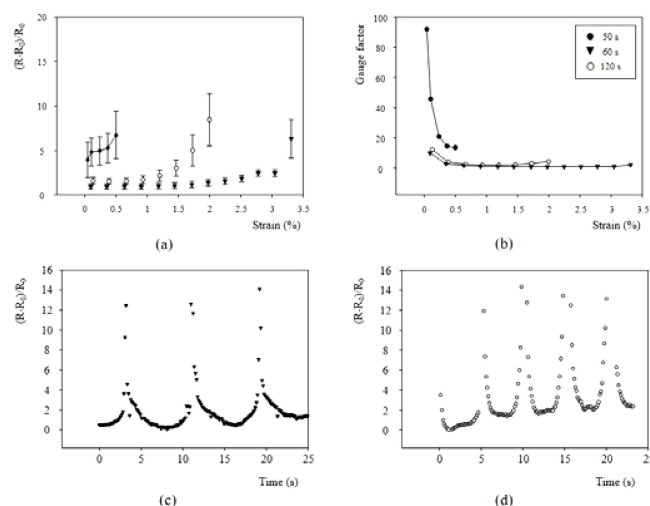
Fig. 4 shows the transmittance spectra and photographs of the fabricated Au/PDMS membranes under five different sputter conditions. The transmittance of the Au/PDMS membranes at 550 nm approached 100.9% when the sputter time was 30 s, 91.4% at 40 s, 87.5% at 50 s, 79.1% at 60 s, and 31.3% at 120 s (Fig. 4a). The Au/PDMS membranes of 30, 40, 50, 60 s conditions show good optical properties. Hence, the underlying image could be clearly observed through these membranes (Fig. 4b).

### 3.3 Electromechanical characteristics of Au electrodes

In general, electromechanical characteristics are very important for strain sensing. The electrical properties of the Au electrodes were controlled by varying the sputtering time. Fig. 5a shows the electrical properties of fabricated Au electrodes according to the sputtering time. The initial resistance was 1727, 424, 222, and 44  $\Omega$  for sputtering times of 40, 50, 60, and 120 s, respectively. The initial resistance was decreased as the Au sputtering time



**Fig. 5.** Electromechanical characteristics of Au/PDMS membranes according to Au sputtering time (a) Initial resistance of Au/PDMS membrane vs. Au sputtering time ( $n = 4$ , mean  $\pm$  standard error), (b) Failure strain of Au/PDMS membrane vs. Au sputtering time (Scale bar indicates 100  $\mu\text{m}$ ).



**Fig. 6.** Resistance variations vs. strain of Au/PDMS membranes. (a) Normalized resistance vs. strain ( $n = 4$ , mean  $\pm$  standard error), (b) Gauge factor vs. strain, (c, d) Normalized resistance vs. time change (sputtering time: 60 s and 120 s respectively, moving velocity: 100  $\mu\text{m/s}$ ).

increased. The resistance was inversely proportional to the square of the sputtering time ( $R^2 = 0.99$ ). Fig. 5b shows the failure strain of Au electrodes and inset images show the surface of Au electrodes. The failure strain was 0.7, 3.9, 2.13% at sputtering times of 50, 60, 120 s, respectively. The failure strain was the highest at a sputtering time of 60 s among the three conditions.

### 3.4 Resistance variations of Au electrodes with strain changes

Fig. 6a shows the normalized resistance variations with strain changes and Figure 6b shows the gauge factor with strain changes. The gauge factor of the Au electrode ranges around 13–92, 0.8–9.9, and 1.9–12 for sputtering times of 50, 60 and 120 s, respectively. The Au electrode prepared with a sputtering time of 50 s had the smallest maximum strain ( $\sim 0.7\%$ ) but the highest gauge factor. The gauge factors of the samples sputtered for 60 and 120 s were similar, but the maximum strain range for the 60 s sample was larger than that of the 120 s sample. Figures c and d show the change in the normalized resistance vs. sputtering time for Au sputtered for 60 and 120 s.

## 4. CONCLUSIONS

In summary, Au/PDMS membranes were fabricated for strain sensors. PDMS membranes were produced by a spin-coating

process and the thickness of the PDMS membrane was controlled by spin rates. The thickness of the fabricated PDMS membranes ranged from 130 to 280  $\mu\text{m}$  and the membrane with 226  $\mu\text{m}$  thickness was adopted to fabricate subsequent Au/PDMS membranes because of their high failure strain. To solve the problems caused by the use of metallic interlayers, the interfacial adhesion between Au film and PDMS membrane was enhanced through MPTMS treatment. Thanks to MPTMS treatment, the interfacial adhesion and the optical properties were enhanced. The electrical property of Au electrodes was controlled by sputtering time, and the resistance was inversely proportional to the square of the sputtering time. The Au electrode with a sputtering time of 50 s had the highest gauge factor with  $\sim 0.7\%$  strain, and the Au electrode with a sputtering time of 60 s had the largest maximum strain range among the three conditions studied (50, 60, and 120 s of sputtering time). We anticipate that our techniques using Au/PDMS with MPTMS treatment will be widely used for fabricating strain sensors.

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