

## Enhancing the electrical conductivity of stretchable silicone composite textiles using ethanol solvent treatment

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**Abstract**—The electrical conductivity and mechanical properties of a stretchable conductive composite textile (CCT) were simply enhanced by solvent treatment with ethanol (EtOH). The proposed flexible conductive composites were prepared using a conductive silicone polymer and a stretchable fabric, wherein the EtOH solvent effectively eliminates the unreacted silicone components between the textile fibers to form a mechanically interlocking structure between silicone polymer and fabric. As a result, mechanical failure between the silicone matrix and the textile layer after repeated tensile testing was prevented to result in a high strain recovery rate of silicone-embedded CCT. After six cyclic strain recovery tests, the EtOH-treated CCT exhibited an excellent recovery rate of 79.3% and continued to maintain a high recovery rate of 70.4% even after ten cycles. Notably, even after continuous tensile loading, an extremely low electrical resistance of 3.1  $\Omega$  was also retained.

Keywords: Conductive Composite Textile, Elasticity, Electrical Conductivity, Ethanol Treatment, Silicone Composite

### INTRODUCTION

Wearable systems have attracted increasing attention as next-generation electronic devices for application in smart clothing, electronic skins, and smart devices [1-4], and research is being actively conducted to combine smart textiles with cutting-edge technology [5,6]. Smart textiles, which exhibit the appearance and touch of general textile materials, can respond to the external environment by sensing environmental changes and variations in performance parameters [7]. To obtain such smart textiles, the electronic components must possess favorably elastic and conductive properties. In general, polydimethylsiloxane (PDMS) is commonly employed for flexible electrodes and, to date, various studies have been conducted using silicon-based flexible electrodes [8-11]. PDMS has received particular attention because it exhibits almost no permanent deformation and can stably adhere to large and uneven surfaces [12-15]. As a result, PDMS electrodes have the potential for application in wearable flexible electronic sensors for signal detection, with examples including smart clothing [16,17], brain wave detection [18], chemical sensors [19], and thermoelectric devices [20].

However, PDMS-based composite textiles tend to exhibit poor electrical conductivity due to mechanical failure between the silicone matrix and textile layer, or the formation of cracks upon the application of repeated tensile stress [21]. In addition, the recovery rate has been shown to decrease owing to the strain factor when cyclic tension is applied. Therefore, to address these deformation issues related to conductive composite textiles (CCTs), solvent treatment has been employed to improve performance [22,23].

In this study, we employed ethanol (EtOH) as a solvent to improve the electrical conductivity and tensile recovery rate of a stretchable silicone-embedded CCT [24-26], and investigated the mechanism of improvement. The overall manufacturing process for the EtOH-treated CCT is depicted in Fig. 1. After a typical additive-type silicone polymer and conductive copper filler are homogeneously mixed using a planetary mixer, the conductive silicone mixture is screen-printed on a stretchable fabric, and subsequently thermally cured. Finally, the fabricated silicone-embedded CCT is chemically treated with EtOH solvent, leading to the formation of a highly conductive and stretchable silicone composite textile. The penetrated EtOH solvent can effectively eliminate the uncured silicone components inside the CCT to induce a mechanically interlocking structure between silicone polymer and fabric. If successful, our proposed process is expected to be advantageous over other treatment techniques since a simple solvent treatment approach is employed without the requirement for a complicated coating process. Ultimately, this could lead to the generation of functional CCTs for use in next-generation electronic devices.

### EXPERIMENTAL

#### 1. Materials

Methyl hydrogen silicone fluid (wave 803B,  $M_n=2,500$  g/mol, Wave Co., Ltd., South Korea) containing 2 wt% platinum catalyst and an additive-type silicone polymer (Sylgard 184,  $M_n=6,650$  g/mol, Sigma Aldrich Co. Ltd, St. Louis, USA) containing 55 wt% highly conductive filler were employed. The conductive fillers, which are composed of dendritic type copper particles with an aspect ratio of 0.22-0.26 and a particle size of 25-30  $\mu\text{m}$ , were purchased from GGP Metalpowder AG (Eckart Plaza, Germany). The thickness of the textile used for manufacturing the CCT was 0.24 mm, and it was composed of 22% spandex and 78% nylon. Toluene (99.5%)

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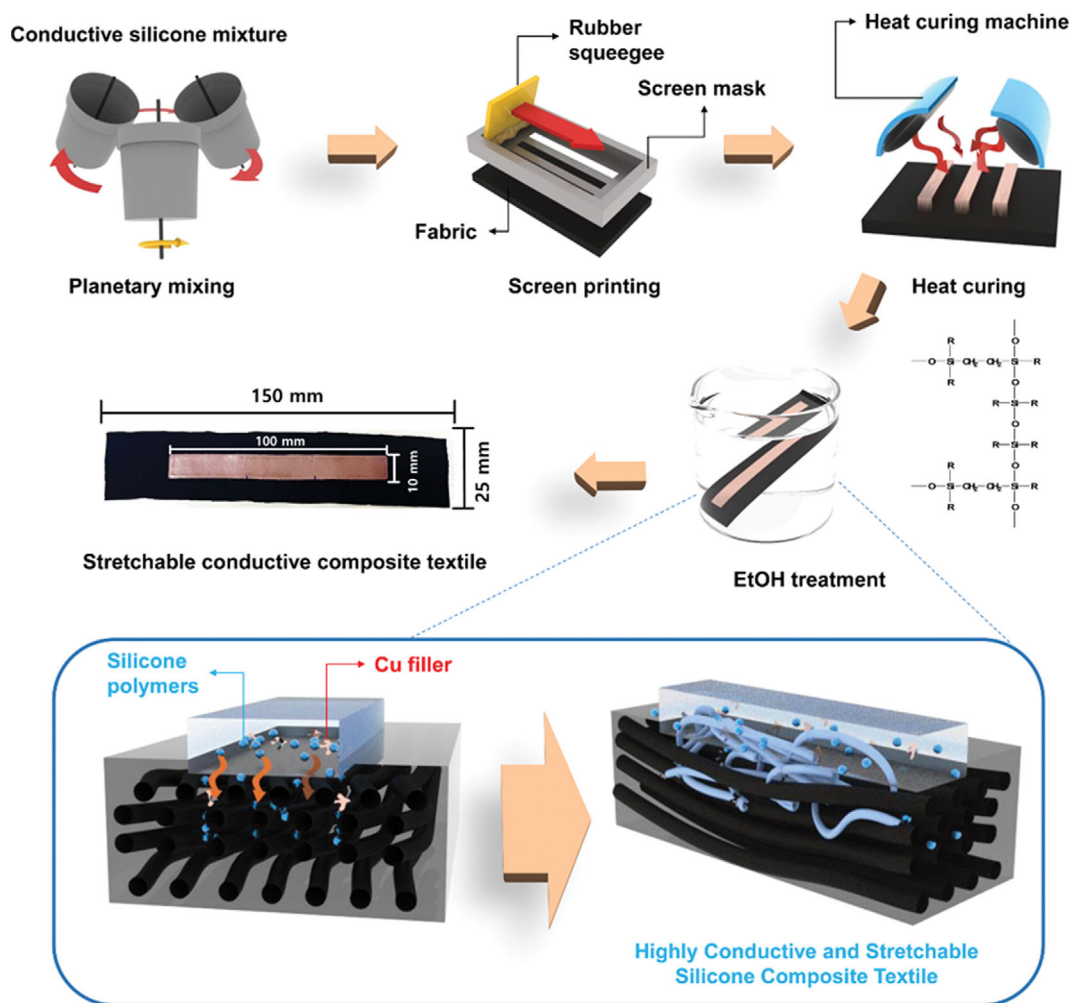


Fig. 1. Schematic illustration of the overall fabrication process including planetary mixing, screen printing, heat curing, and solvent treatment for the EtOH-treated silicone CCT.

and ethanol (99.8%) were obtained from Sigma-Aldrich Co., Ltd. (St. Louis, MO, USA).

## 2. Preparation of the Conductive Silicone Polymer

The stretchable textile was coated with a mixture of toluene (3 g) and the conductive silicone polymer (46 g). The toluene was added to reduce the viscosity of the conductive silicone mixture and thereby match the mask pattern. The above mixture was subjected to planetary centrifugal mixing (AR-100, Thinky, Japan) for 5 min at a revolution/rotation speed of 800/2,000 rpm. After mixing the silicone curing agent and low-viscosity conductive silicone polymer in a weight ratio of 1 : 14 for 5 min to harden the silicone polymer, degassing was carried out for 15 min to eliminate any air bubbles formed inside the conductive silicone mixture during the homogenization process.

## 3. Preparation of the Stretchable Conductive Silicone Composite Textile

The above conductive silicone polymer was coated on a stretchable fabric in the wale direction using the screen-printing approach with a metal mask. The textile was screen-printed with conductive silicone polymer using a rubber squeegee by applying a constant force and angle, and was then thermally cured in a convection oven

(OF-O1E, JEIO Tech, South Korea) at 170 °C for 3 min. Subsequently, the cured silicone composite textile was subjected to EtOH treatment for 6 h. Finally, the obtained CCT specimen was cut into samples measuring 150×25 mm to conduct the repeated strain recovery test.

## 4. Characterization

To confirm the strain recovery characteristics of the EtOH-treated CCT material, the treated specimen was compared with the untreated CCT using a universal testing machine (UTM, LD-M, LLOYD, UK). The strain recovery test was performed ten times at a crosshead speed of 5 mm/min. After five measurements, a break of 5 min was implemented prior to carrying out the subsequent five measurements. The CCT with a strain of 10% (10 mm) was employed in the circulation recovery test, and as the test progressed, permanent deformation of the CCT took place. The strain recovery rate (%) was then calculated using the following Eq. (1).

$$\text{Strain recovery rate (\%)} = \frac{R_{\text{unload}}}{R_{\text{load}}} \times 100 \quad (1)$$

where  $R_{\text{unload}}$  represents the length of the recovered CCT (i.e., when the CCT returns to its initial state), and  $R_{\text{load}}$  is equal to 10

mm, which is the length to which 10% strain was applied. To observe the degree of damage or morphological change to the surface of the stretchable CCT, the deformed CCT was analyzed after repeated strain recovery tests using optical microscopy (OM, BX53, Olympus, Japan). In addition, field-emission scanning electron microscopy (FE-SEM, Gemini SEM300, Zeiss, Germany) was used to investigate the fracture plane of the CCT and then confirm that the mechanical failure between silicone polymer and the textile can be prevented after EtOH treatment. Energy-dispersive X-ray spectroscopy (EDS, XFlash 6I100, Bruker, USA) was used to confirm that the silicone polymer and Cu filler penetrate the fabric after thermal curing and EtOH treatment. The electrical resistance was measured using an LCR meter (72-10465, Tenma, Japan) at a frequency of 100 Hz for 3 min both in the initial state and after repeated strain recovery tests (5 and 10 cycles). The sample size and thickness for measurement of electrical resistance were 100×10 mm and 0.24 mm, respectively.

## RESULTS AND DISCUSSION

### 1. Electrical Conductivity of the EtOH-treated CCTs

In a CCT, the fibers are deformed due to movement of the body and, as a result, the electrical conductivity may decrease owing to the formation of cracks in the silicone and fiber layers. To address this issue, the silicone-embedded CCT was treated with EtOH solvent for 6 h to modify both the surface and the interior of the CCT. For error minimization, the change in the electrical resistance during the repeated deformation of an untreated CCT and the EtOH-treated CCT was measured between two endpoints of the specimen. Thus, Fig. 2 shows the change in the electrical resistance of the CCT depending on the cycle number during the cyclic ten-

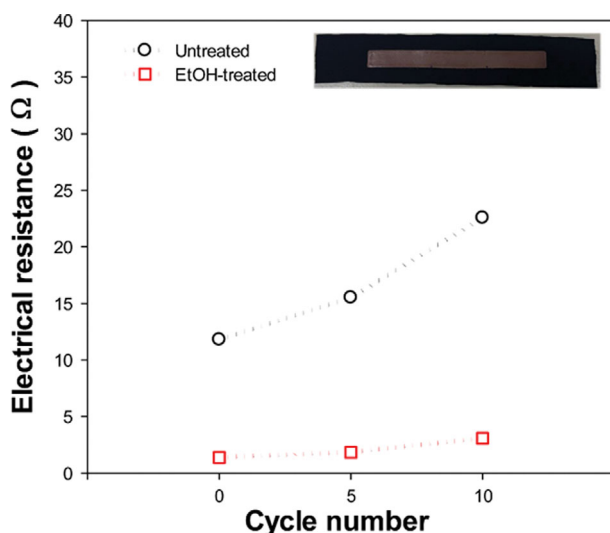


Fig. 2. Average electrical resistance of an untreated CCT (black circle) and the EtOH-treated CCT (red square) depending on the cycle number during the cyclic recovery test. The change in the electrical resistance during the repeated deformation was measured between two endpoints of the specimen with five measurements. The inserted image is the photographic image of EtOH-treated CCT specimen.

sile test. More specifically, the electrical resistance values of the untreated and EtOH-treated specimens before carrying out the repeated tensile test were 11.8 and 1.4 Ω, respectively, while after ten test cycles, the resistance values were 22.5 and 3.1 Ω, respectively. Notably, the EtOH-treated CCT showed a remarkably low electrical resistance, and its resistance value was maintained below a very low level of  $\leq 5 \Omega$  even after repeating the tensile test ten times. In addition, after repeated tensile testing of the untreated sample, the resistance value increased by 10.7 Ω compared to its initial level, while the resistance value of the EtOH-treated sample increased by only 1.7 Ω. This difference between the treated and untreated samples can be attributed to removal of the uncured silicon polymer on the surface and interior of CCT during the long-term ethanol treatment and, as a result, the electrical conductivity was enhanced due to an increased Cu density on the surface and formation of a mechanically interlocking structure between silicone polymer and fabric inside the CCT. Thus, EtOH treatment clearly resulted in a higher electrical conductivity for the silicone-embedded CCT.

### 2. Cyclic Recovery Tests

To render a CCT suitable for application in wearable textiles, it must return to its original state following stretching or wrinkling. Thus, the elasticity characteristics of the manufactured silicone CCT after the recovery cycle tests were analyzed using a UTM. Figs. 3(a) and 3(b) show the elastic recovery performance of the untreated and EtOH-treated CCTs, respectively, wherein it can be seen that the initial load values of these CCT samples were 0.8 and 1.0 N, thereby confirming that the load value increased following EtOH treatment. In addition, in the first cycle after the 5 min rest period (i.e., cycle number 6), the recovery rates of the untreated and EtOH-treated specimens were 76.1 and 79.3%, respectively. For comparison, from Fig. 3(a) the recovery rate of the untreated sample was 77.1% prior to testing (i.e., higher than that of the EtOH-treated sample, 72.9%), while after ten cycles, the recovery rate was 68.1%, representing a reduction of 11.7% from the initial recovery rate. In contrast, after ten cycles, the recovery rate of the EtOH-treated sample was 70.4%, which is an only 3.4% decrease compared to the original recovery rate. These reductions after cyclic recovery tests can be attributed to rearrangement of the silicone polymer network within the textiles. Note that the initial recovery rate of the untreated sample was higher than that of the EtOH-treated sample because EtOH treatment increased the initial tensile load value owing to an increase in the brittle properties caused by loss of the uncured silicon matrix and an increase in the surface Cu ratio. However, EtOH treatment removed the deformation factor, and so after ten cycles, the recovery rate was maintained to a greater degree. These results confirmed that EtOH treatment resulted in superior elastic recovery properties for the silicone-embedded CCT compared to the untreated sample.

### 3. Surface Morphology of the CCTs

To determine how EtOH treatment affects the conductive silicon composite surface, the top surfaces of the untreated and EtOH-treated CCTs were observed using optical microscopy (OM) before and after repeated tensile tests, as shown in Fig. 4(a) and 4(b), respectively. Fig. 4(a) shows that no cracks were observed in the case of the untreated CCT before cyclic recovery testing, but the sizes of

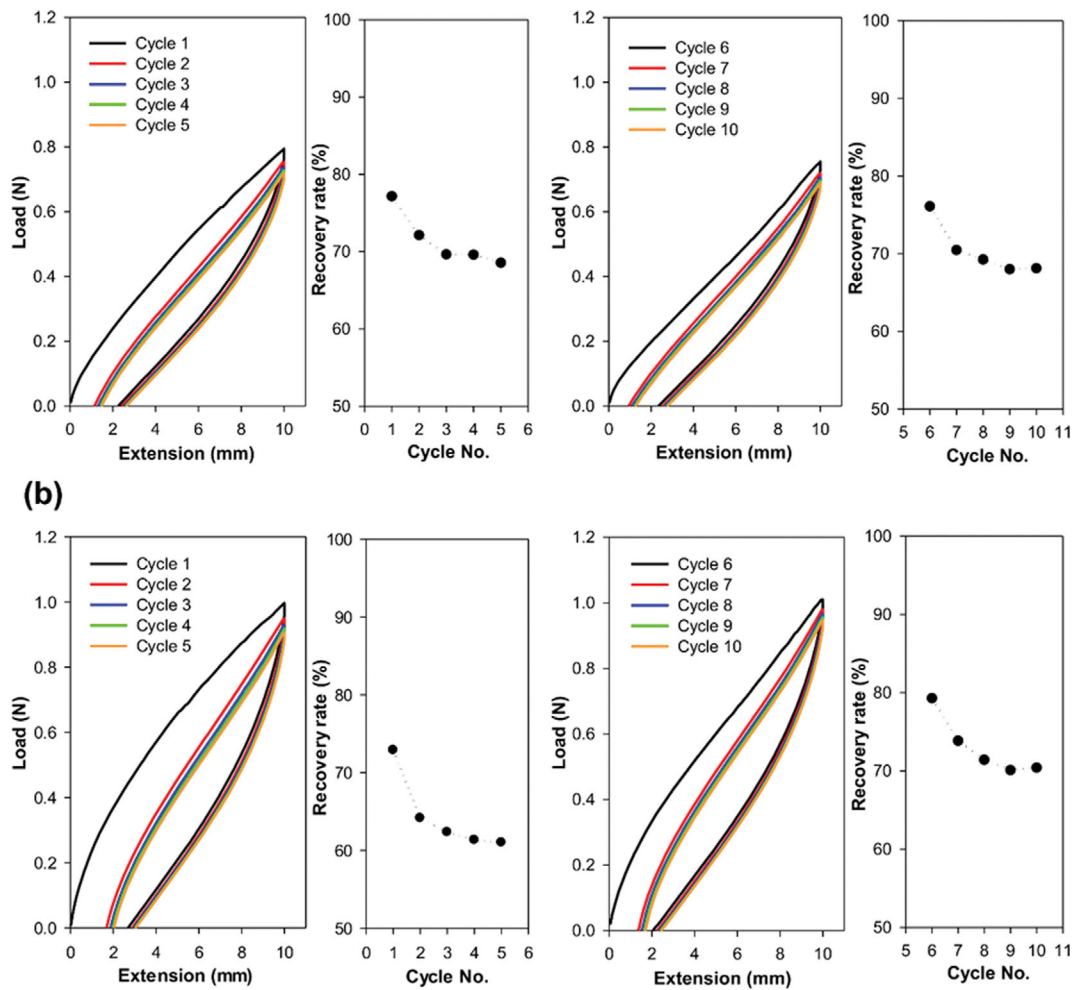


Fig. 3. Cyclic strain recovery behavior of (a) an untreated CCT and (b) the EtOH-treated CCT, depending on the cycle number during the cyclic recovery test. The loop line graph and dashed line graph represent strain recovery curve and strain recovery rate, respectively.

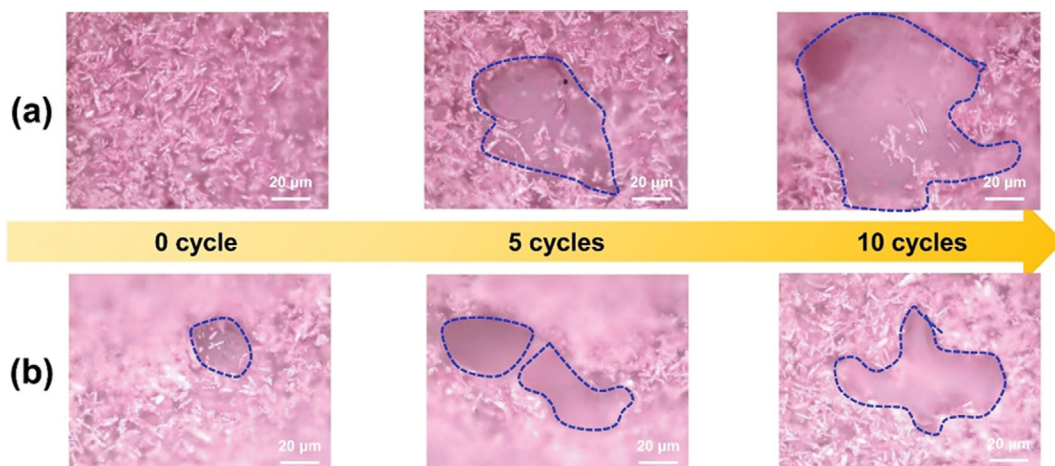


Fig. 4. OM surface images of (a) an untreated CCT and (b) the EtOH-treated CCT depending on the cycle number during the cyclic recovery test. The dashed line area denotes surface cracks on the fabricated CCTs.

the cracks increased significantly as cycling progressed. In addition, following EtOH treatment, some small cracks were present on the silicon CCT surface prior to cyclic test. However, compared

to the untreated CCT, the crack size during cycling increased to a lesser extent. It was considered that the untreated CCT showed no surface cracks in the OM image, whereas the EtOH-treated CCT

exhibited small surface cracks caused by removal of the uncured silicone components, indicating the presence of relatively high Cu content in the surface of the EtOH-treated CCT. In addition, after five and ten cycles of repeated tensile tests for the EtOH-treated CCT specimen, the electrical resistance may have increased due to the presence of cracks on the silicon CCT surface. However, disappearance of the uncured silicone components under EtOH treatment can lead to an increased Cu content on the CCT surface, which in turn results in a higher electrical conductivity of the CCT.

Overall, the increases in electrical resistance stemming from surface cracks and electrical conductivity owing to the increased surface Cu ratio were offset against one another, and so the electrical resistance was maintained.

**4. Cross-sectional Morphology of the CCTs**

The cross-sections of the fabricated CCT were observed using FE-SEM and EDS to determine the effect of EtOH treatment in terms of the degree of mechanical failure or cracking between the silicone matrix and the fibrous layer. Figs. 5 and 6 show the cross-

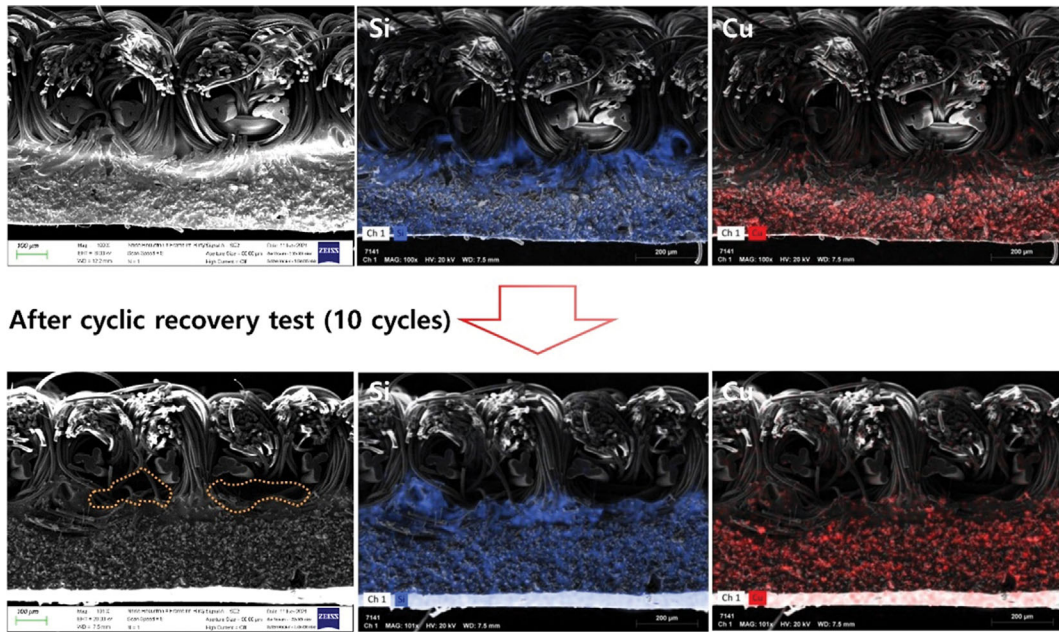


Fig. 5. Cross-sectional FE-SEM and EDS images of the untreated CCT before and after ten cycles of cyclic recovery test. The blue and red areas represent the penetrated Si and Cu elements, respectively.

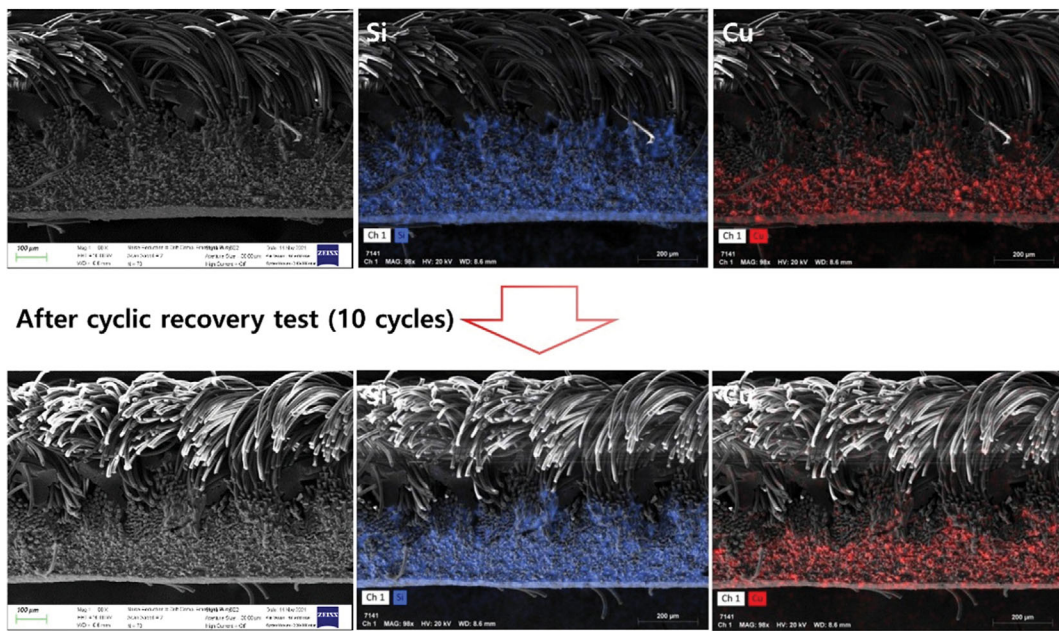


Fig. 6. Cross-sectional FE-SEM and EDS images of the EtOH-treated CCT before and after ten cycles of cyclic recovery test. The blue and red areas represent the penetrated Si and Cu elements, respectively.

sectional FE-SEM and EDS images of the untreated and EtOH-treated CCTs, respectively, before and after ten cycles of cyclic recovery test. It is evident that the silicone polymer and Cu filler penetrated deep into the fabric area after thermal curing and EtOH treatment judging from EDS images. The deep infiltration of conductive silicone polymer can induce the formation of a stable cross-linked network between silicone polymer and host fabric. However, compared to the EtOH-treated CCT, the untreated CCT showed a morphological change after cyclic recovery test. More specifically, upon examination of the SEM cross-section of the untreated CCT after ten repeated tensile tests (Fig. 5), it was possible to observe the region where mechanical failure between the silicone matrix and the fiber layer occurred. This was also confirmed by the decreased strain recovery rate after ten repeated tensile tests, and based on a combination of these results, it appeared that the stable CCT structure had collapsed. Furthermore, the copper area of the untreated CCT increased after the cycle test, suggesting that a number of Cu particles in the uncured silicone polymer were detached from the Cu aggregates and then were newly distributed in a direction perpendicular to the tensile direction [27]. In addition, as OM observations of the conductive composite (Fig. 4(a)) showed, few surface cracks were present prior to testing, while large-area cracks were observed after ten repeated tensile tests. However, the surface OM result of the EtOH-treated CCT specimen (Fig. 4(b)) confirmed that small cracks were present prior to testing, and that these cracks were slightly enlarged after ten test cycles, as indicated above. Similarly, as shown in Fig. 6, no mechanical failure or cracking could be seen in the cross-sectional image even after ten repeated tensile tests, thereby suggesting that a stable mechanically interlocked structure was maintained without mechanical failure of the silicone and fiber layers. The FE-SEM and EDS results therefore confirmed that both the uncured silicon matrix on the CCT surface and the uncured silicon polymer penetrating the textile were effectively removed and, as a result, a stable and highly mechanically interlocking structure was formed in the composite upon treatment with EtOH.

## CONCLUSIONS

We herein reported the preparation of an ethanol (EtOH)-treated conductive composite textile (CCT), where our aim was to employ EtOH treatment to form a stable and highly conductive CCT. Repeated tensile tests confirmed that the prepared EtOH-treated CCT exhibited high electrical conductivity, even after repeated deformation. More specifically, after ten tensile test cycles of the EtOH-treated CCT, the strain recovery rate was 70.4%, which was 2.3% higher than that obtained for the untreated CCT after testing. This result indicates that EtOH treatment removed the strain factor, which led to superior elastic recovery behavior in the treated specimen. Furthermore, after repeated tensile testing, scanning electron microscopy imaging combined with energy-dispersive X-ray spectroscopy showed that a region of mechanical failure between the silicone matrix and the fiber layer was present in the untreated CCT sample. However, following EtOH treatment, a stable mechanically interlocking structure was formed, which was maintained during cycling without mechanical failure. Moreover, optical microscopy observations confirmed that cracks occurred on the surface

of the untreated and EtOH-treated fibers, which were responsible for decreased conductivity. Overall, our results demonstrate that it is possible to effectively manufacture a highly conductive silicone CCT with a stable internal structure through this EtOH-treatment process.

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