Copper micromesh-based lightweight transparent conductor with short response time for wearable heaters

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Abstract
Thickness-controlled transparent conducting films (TCFs) were fabricated by transfer printing a 100 nm thick Cu micromesh structure onto poly(vinyl alcohol) (PVA) substrates of different thicknesses (~ 50, ~ 80, and ~ 120 μm) to develop a lightweight transparent wearable heater with short response time. The Cu mesh-based TCF fabricated on a ~ 50 μm thick PVA substrate exhibited excellent optical and electrical properties with a light transmittance of 86.7% at 550 nm, sheet resistance of ~ 10.8 Ω/sq, and figure-of-merit of approximately 236, which are comparable to commercial indium tin oxide film-based transparent conductors. The remarkable flexibility of the Cu mesh-based TCF was demonstrated through cyclic mechanical bending tests. In addition, the Cu mesh-based TCF with ~ 50 μm thick PVA substrate demonstrated a fast Joule heating performance with a thermal response time of ~ 18.0 s and a ramping rate of ~ 3.0 °C/s under a driving voltage of 2.5 V. Lastly, the reliable response and recovery characteristics of the Cu mesh/PVA film-based transparent heater were confirmed through the cyclic power test. We believe that the results of this study is useful in the development of flexible transparent heaters, including lightweight deicing/defogging films, wearable sensors/actuators, and medical thermotherapy pads.

Keywords: Wearable transparent heater, Transparent conductor, Metal mesh, Thermal response time, Transfer printing

Introduction
Transparent heaters (THs) have been studied for multiple potential applications, including smart windows [1, 2], defrosters [3, 4], thermochromic displays [5, 6], physical/chemical sensors [7, 8], and other advanced heat-generating systems [9–11]. High optical transparency and excellent electrical conductivity are two important factors for high-performance THs. Moreover, this technology is mainly developed based on the research results of existing transparent conductors with transparent conductive oxides (TCOs), such as indium tin oxide (ITO) and fluorine-doped tin oxide (FTO) films [12]. However, owing to their brittle nature, it is extremely difficult to adapt TCO films in the development of next-generation transparent conductors for flexible/wearable TH applications [11]. Thus, there has been active research on the replacement of commercial TCO films for flexible/wearable THs.

The potential candidate materials for ITO/FTO-free flexible/wearable THs include conductive polymers [6], carbon-based nanomaterials [5, 13, 14], metal-based nano/micro-materials [3, 4], and other hybrid materials [15, 16]. Conductive polymers, such as poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), have outstanding advantages in terms of mechanical flexibility and solution processibility; however, conductive polymers have low electrical conductivity, and poor stability against heat and moisture, thereby limiting their application in high-performance optoelectronic devices...
Meanwhile, carbon nanomaterials, such as graphene and carbon nanotubes, have excellent thermal conductivity and flexibility; however, the manufacturing costs for high-quality carbon-based nanomaterials through thermal and plasma chemical vapor deposition techniques are usually expensive [11, 19]. Accordingly, owing to their relatively simple manufacturing process (high productivity) and superior physical properties, such as optical/electrical conductance and mechanical flexibility, the development of metal-based flexible THs with metal nanowires (NWs) and mesh structures have recently garnered considerable attention as an alternative to commercial TCO films [3, 4, 9, 20].

In this study, we introduce a new strategy to develop a lightweight flexible TH with short response time for wearable theramotherapy and heating systems. A 100 nm thick conductive Cu mesh structure with optically transparency and electrically conductivity was fabricated onto poly(vinyl alcohol) (PVA) substrates of different thicknesses (~50, ~80, and ~120 μm) through transfer printing method. A lightweight flexible Cu mehs/PVA film-based transparent conducting film (TCF) based on a ~50 μm thick PVA substrate with optical and electrical properties comparable to commercial ITO glass was successfully fabricated. Further, the Cu mesh/PVA film-based TCF demonstrated its fast, stable, and reliable performance for TH applications. Finally, for its actual application, the Cu mesh/PVA film-based TH was attached to a finger and the back of the hand.

**Experimental**

**Materials**

Ultraviolet (UV)-curable polyurethane acrylate (PUA) imprint resin (MINS-311RM) and 99.99% Cu were purchased from Changsung Sheet Co., Ltd. (Korea) and Taewon Scientific Co. Ltd. (Korea), respectively. And, PVA powders (Mw of 85,000–124,000) and trichloro(1H,1H,2H,2H-perfluoroctyl)silane (97%) were purchased from Sigma-Aldrich Inc. (USA).

**Optical, electrical, and structural characterization**

The optical transmittance spectra of the prepared Cu mesh-based TCFs were obtained using a UV–visible near-infrared spectrometer (Cary 5000 UV–vis-NIR, Agilent, USA) with air as the reference. The electrical conductance (sheet resistance) of the Cu mesh-based TCF was measured using a four-point probe method with a sheet resistivity meter (FPP-2000, DASOL ENG, Korea). The TH structure was analyzed using a field-emission scanning electron microscope (FE-SEM, Sirion, FEI, USA) and optical microscope (BX51, OLYMPUS, Japan). The weight of the fabricated flexible TCFs were measured using a precision electronic balance (HM-202, A&D Co. Ltd, Japan). The sheet resistance, optical transmittance, and weight of the TCF were measured at least five times, and then the average value was taken.

**Evaluation of the electrothermal performance of the Cu mesh-based TCFs**

To investigate the Joule heating performance of the Cu mesh-based TCF, direct current (DC) voltages of 0.5, 1.0, 1.5, 2.0, and 2.5 V was applied to the TCF through contact with the Ag side. The resulting temperature change was recorded by a direct measurement using a thermocouple (ST-50, RKC Instrument Inc., Japan) mounted on the PVA substrate part without a Cu mesh. The temperature on the surface of the TH was measured three times and the average value was used. The obtained temperature profile was confirmed using an infrared (IR) camera (Seek Compact, Seek Thermal Inc., USA).

**Results and discussion**

In this work, TCFs based on a uniform Cu mesh structure were fabricated by a transfer printing method without a lift-off, etching or electroplating process [24]. First, a PUA mold with an engraved micromesh pattern was replicated from a silicon master through a replicate molding method (see Additional file 1: Figure S1). Here, the silicon master used in this work could be prepared by photolithography and dry etching techniques. The surface of the PUA mold was treated with a layer of 1H,1H,2H,2H-perfluoroctyl-trichlorosilane to form an anti-sticking layer covering. A thin Cu layer with a thickness of 100 nm was deposited on the surface of the PUA mold using an electron-beam evaporator, as shown in Fig. 1a. For this step, Cu has an evaporation rate of approximately 0.1 nm/s. Here, the Cu layer deposited on the PUA mold with the anti-sticking layer was designed to be easily peeled off. A transparent PVA substrate was used as the receiver for the selective transfer of the Cu mesh layer from the PUA
mold to fabricate the TH substrate. A 20 wt.% aqueous PVA solution with viscosity of ~20,000 cP was bar-coated onto a cleaned glass plate and cured in a drying oven at 50 °C for 24 h, thereby a PVA film with constant thickness was obtained by controlling the gap between the glass plate and blade. After thermal curing, the PVA film was carefully separated from the glass plate. The Cu thin film-deposited PUA mold was then placed in contact with the surface of the PVA film. The PUA mold/PVA film was assembled through hot-pressing by applying ~1.5 bar static pressure at ~100 °C for 3 min. Subsequently, the PUA mold/PVA film was naturally cooled to room temperature (RT). Finally, a uniform Cu mesh structure on the PVA film was obtained after peeling the PUA mold. Figure 1b shows a photograph and surface FE-SEM image of the Cu mesh directly printed on the PVA film. Additional file 1: Figure S2 shows an optical microscope image of the Cu mesh/PVA film surface observed at a low magnification. A Cu mesh structure with a line width of 1.5 µm and a line spacing of 150 µm was successfully formed on the PVA film. Further, the optical transparency of the Cu mesh/PVA film is confirmed.

Figure 2 shows the performance of the Cu mesh/PVA film as a transparent conductor. As shown in Fig. 2a, a red light-emitting diode (LED) turned on using the ~50 µm thick Cu mesh/PVA film-based TCF. The as-prepared Cu mesh/PVA film-based TCF exhibited an optical transmittance of 86.7% at 550 nm, average transmittance of ~85.0% over the entire visible range (see Additional file 1: Figure S3), and average sheet resistance of ~10.0 Ω/sq (see Additional file 1: Figure S4). The optical transmittance at 550 nm, sheet resistance, and figure-of-merit (FOM) of the commercial ITO glass and Cu mesh/PVA film-based TCFs with various thicknesses are summarized in Table 1. The FOM value is calculated as the ratio of the electrical conductance ($\sigma_{dc}$) to the optical conductance ($\sigma_{opt}$), following the equation: [24]
where $R_S$ and $T$ are the measured sheet resistance and the optical transmittance at 550 nm, respectively, and $Z_0$ is the impedance at free space (377 $\Omega$). As shown in the Table 1, the Cu mesh/PVA film with ~50 $\mu$m thick PVA substrate has an FOM value of 236.1, which is similar to that of commercial ITO glass (235.6). The weights of the Cu mesh-based TCFs fabricated on ~50, ~80, and ~120 $\mu$m thick PVA films were measured to be 27.8, 42.7, and 65.9 mg, respectively. The weight per unit area of the Cu mesh-based TCF with ~50 $\mu$m PVA substrate was approximately 0.0695 mg/mm$^2$. As seen in Fig. 2b, the optical and electrical conductance of the Cu mesh/PVA film-based TCF with ~50 $\mu$m thick PVA substrate was comparable to that of other metal-based transparent conductors [9, 20, 22, 24–36].

To apply the fabricated Cu mesh/PVA film-based TCF to flexible/wearable electronic devices, the change in the current–voltage relationship under bending should be investigated. Thus, the current of the Cu mesh/PVA film-based TCF with ~50 $\mu$m thick PVA substrate was measured with different input voltages with its flat and bent state. As shown in Fig. 3a, the current steadily increased up to 288 mA at an applied voltage of 3.0 V in the flat state. In contrast, when the Cu mesh/PVA film was bent with an outside bending radius of 5 mm, the current slightly decreased to 277 mA at 3.0 V. The slight decrease in the current at a relatively high input voltage range (2.0–3.0 V) is mainly attributed to the decrease in the local connectivity between the evaporated Cu particles induced by the bending stress.

Figure 3b shows the changes in the sheet resistance of the Cu mesh/PVA film and ITO/PVA film-based TCFs fabricated with the same thickness (~50 $\mu$m) as the bending radius decreases. The sheet resistance of the ITO/PVA film-based TCF with an initial sheet resistance of ~30 $\Omega$/sq significantly increased when the bending radii were less than 10 mm due to the increased brittleness of ITO thin film [29]. The cracks formed on the surface of the ITO film could be confirmed after the mechanical bending test, as shown in the optical microscope image inserted in Fig. 3b. In contrast to the ITO/PVA film-based TCF, there were no changes observed in the initial sheet resistance (~11 $\Omega$/sq) of the Cu mesh/PVA film-based TCF even at a bending radius as small as 4 mm. Figure 3c shows the changes in the sheet resistance...
with the increase of the bending cycles for the outward and inward bending fatigue tests of the Cu mesh/PVA film-based TCF with ~50 μm thick PVA substrate at a fixed bending radius of 4 mm. After 1,000 cycles, the sheet resistance of the Cu mesh/PVA film-based TCF marginally increased to ~20% for the inner bending tests, whereas it increased by ~80% for the outer bending tests. These experimental results indicate that the uniform Cu mesh structure is more vulnerable to failure under external bending stress than internal bending stress.

The excellent flexibility of the Cu mesh/PVA film-based TCF was further demonstrated by lighting a red LED using the fabricated Cu mesh/PVA film-based TCF as the flexible transparent conductor, as shown in Fig. 4. Particularly, there are no noticeable changes in the LED brightness before and after bending the TCF.

Figure 5a shows the time-dependent temperature of the Cu mesh/PVA film as a TH with a sheet resistance of ~11.0 Ω/sq. By applying different bias voltages (0.5–2.5 V), the temperature of the Cu mesh/PVA film-based TH rapidly increased from RT to saturated values until the thermal equilibrium was achieved. The temperature of the TH rapidly increased with respect to the driving voltage. At a driving voltage of 2.5 V, the temperature of the TH reached ~85.0 °C. The heat generation of the Cu mesh/PVA film-based TH first occurred through Joule heating. The generated heat subsequently dissipated through conduction in the substrate, air convection, and
radiation [9]. When the applied voltage was cut off, the temperature of the TH decreased to RT in ~60 s. Figure 5b shows the time–temperature curves of the TH with different PVA film thicknesses at a constant input voltage of 2.5 V. The response time, which is defined as the time required to reach 90% of the steady-state temperature during operation, of the Cu mesh/PVA film-based THs with a PVA film thickness of ~50, ~80, and ~120 µm were 18.0, 21.5, and 23.5 s, respectively. Here, the average response time of the THs with various PVA film thickness was reproducible with a relative standard deviation of only 2.1%. And, the response time of the TH based on Cu mesh/PVA film with a thickness of ~50 µm is much faster than that of the flexible TH reported so far (see Additional file 1: Table S1). The thinner Cu mesh/PVA film TH exhibited shorter thermal response time and faster ramp rate (see Table 2). The decrease in the heater response time due to the decrease in the thickness of the heater substrate (PVA film) could be explained as follows. For a Cu mesh/PVA film-based TH at a given driving voltage, the heating (ramping) rate and saturated temperature depend on the thermal properties of the heating element (Cu mesh structure) and the heater substrate (PVA film). Generally, response time is related to the time constant (τ) of the transient thermal response depicted as [16]

\[ \tau = \frac{C}{hA} \]  

(2)

where C refers to the heat capacity, h is the heat transfer coefficient, and A is the surface area. In this study, the heating element (Cu mesh structure) thickness of ~100 nm can be deemed negligible compared to the thickness of the heater substrate (PVA film), which is in the range of 50–120 µm. Therefore, the heat capacity (C) of the Cu mesh structure could be neglected and that of the PVA film dominates the transient process of the electrothermal response. Thus, τ can be rewritten as Eq. (3).

\[ \tau = \frac{C}{hA} = \frac{cm}{hA} = \frac{\rho cd}{h} \]  

(3)

where c, m, ρ, and d are the specific heat capacity, mass, density, and thickness of the PVA film, respectively. Therefore, the response time of the Cu mesh/PVA film-based TH could be reduced by reducing the thickness of the heater substrate (PVA film).

The repeatability of the heat generation of the TH was investigated by a cyclic test of 10 on/off cycles. As shown in Fig. 5c and Table 3, there were no significant changes in the thermal response/recovery characteristics of the Cu mesh/PVA film-based TH during the cyclic test. Joule heating demonstrated a self-annealing effect on the Cu mesh structure. As shown in Fig. 6, the sheet resistance of the Cu mesh/PVA film-based TH exponentially dropped from ~10.9 Ω/sq to ~8.9 Ω/sq (18.3% decrease) after self-annealing at ~85 °C for 3 h. The decrease in the sheet resistance is attributed to the increased electrical connectivity between the Cu particles after Joule heating (self-annealing) [35]. Hence, the sheet resistance of the Cu mesh/PVA film-based TH can be effectively reduced through the self-annealing process, thereby reducing the power consumption and improving the heat generation performance of the TH.

Figure 7, Additional file 1: Figures S5, and S6 show the operation of the Cu mesh/PVA film-based THs with various shapes and sizes. When the TH was attached to curved surfaces, such as a round-bottom flask and finger (see the Additional file 1: Figure S5 and S6, respectively), the TH retained its normal functions. In addition, the TH fixed on the back of the hand operated normally when the fist was clenched and opened, as shown in the Fig. 7. These results indicate the applicability of the fabricated Cu mesh/PVA film-based TCF as a flexible/wearable TH.
Taken together, the proposed method has the advantage that there are no additional processing techniques such as lift-off, etching, or electroplating, compared with the previous studies on the fabrication of high-performance flexible/wearable THs [23, 30, 32, 35]. In addition, it was found that the thermal response time of

Table 2  Comparison of the performances of the Cu mesh/PVA film heaters fabricated with various PVA film thicknesses

| Thickness (µm) | Rs (ohm/sq) | Max. Temp. at 2.5 V (℃) | Response time (s) | Ramping rate (℃/s) |
|---------------|-------------|-------------------------|-------------------|-------------------|
| 50            | ~11.0       | 84.7                    | 18.0              | 2.94              |
| 80            | 84.5        | 21.5                    | 2.46              |
| 120           | 84.4        | 23.5                    | 2.25              |

Table 3  Changes in the performance of the Cu mesh/PVA film-based TH during the cyclic test

| Cycle No | Rs (ohm/sq) | Max. Temp. (℃) | Response time (s) | Min. Temp. (℃) |
|----------|-------------|----------------|-------------------|----------------|
| 0        | 11.0        | –              | –                 | –              |
| 1        | 10.8        | 84.6           | 18.0              | 26.9           |
| 2        | 10.8        | 84.7           | 18.0              | 26.7           |
| 3        | 10.9        | 84.5           | 17.5              | 26.8           |
| 4        | 10.8        | 84.7           | 18.0              | 26.7           |
| 5        | 10.8        | 84.8           | 18.5              | 26.8           |
| 6        | 10.8        | 84.9           | 18.5              | 26.8           |
| 7        | 10.9        | 84.8           | 18.0              | 26.7           |
| 8        | 11.0        | 84.9           | 18.5              | 26.8           |
| 9        | 10.8        | 84.9           | 18.5              | 26.9           |
| 10       | 10.9        | 84.6           | 18.0              | 26.8           |

Fig. 5  a Heat generation performance of the Cu mesh/PVA film heater (20 mm × 20 mm) as a function of time at an applied voltage of 0.5, 1.0, 1.5, 2.0, and 2.5 V. b Time–temperature curves of the Cu mesh/PVA film TH of different PVA substrate thicknesses. The time provided in the graphs indicate the response time of the THs. c Results of the cyclic test of the heat generation of the Cu mesh/PVA film heater with an applied voltage of 2.5 V

Fig. 6  Change in the sheet resistance of the Cu mesh/PVA film with respect to the thermal treatment time by Joule heating
the heater was shortened in proportion to the decrease in the thickness of the heater substrate.

**Conclusion**

In summary, we developed a lightweight flexible TCF by transfer printing a 100 nm thick Cu mesh structure onto PVA films of various thicknesses for application in wearable heaters. A Cu mesh-based lightweight TCF fabricated on a ~50 μm thick PVA substrate demonstrated optical and electrical properties comparable to those of commercial ITO glass. In addition, the Cu mesh/PVA film-based TCF exhibited superior mechanical flexibility compared to the ITO thin film sputtered on the same substrate. The TCF demonstrated its rapid Joule heating behavior with a response time of ~18.0 s and a ramping rate of ~3.0 °C/s at a driving voltage of 2.5 V. The reliable response and recovery characteristics of the Cu mesh/PVA film-based TH were confirmed through the cyclic tests. Lastly, the lightweight TH attached to a human hand or round bottom flask successfully retained its normal functions. Based on the above research results, we expect that the Cu mesh-based lightweight TCF could be used as an ITO/FTO-free electrode/heater for advanced wearable electronic devices.

**Abbreviations**

TCF: Transparent conducting film; PVA: Poly(vinyl alcohol); TH: Transparent heater; TCO: Transparent conductive oxide; ITO: Indium tin oxide; FTO: Fluorine-doped tin oxide; PEDOT:PSS: Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate); NW: Nanowire; UV: Ultraviolet; PUA: Polyurethane acrylate; DC: Direct current; IR: Infrared; RT: Room temperature; LED: Light-emitting diode; FOM: Figure-of-merit; \( \sigma_{dc} \): Electrical conductance; \( \sigma_{opt} \): Optical conductance; \( R_S \): Sheet resistance; \( T \): Optical transmittance at 550 nm; \( Z_0 \): Impedance at free space; \( \tau \): Time constant; \( C \): Heat capacity; \( h \): Heat transfer coefficient; \( A \): Surface area; \( c \): Specific heat capacity; \( m \): Mass; \( \rho \): Density; \( d \): Thickness film.
**Supplementary Information**

The online version contains supplementary material available at https://doi.org/10.1186/s40486-021-00132-5.

**Additional file 1.** Fabrication, optical and electrical properties, and heater performances of the Cu mesh/PVA film-based TCF.

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**Authors’ contributions**

HK conducted the main experiment. YK supervised the project. All authors read and approved the final manuscript.

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**Availability of data and materials**

All data generated or analyzed during in this study are included in this published article and the supplementary information.

**Declarations**

**Ethics approval and consent to participate**

Not applicable.

**Consent for publication**

Not applicable.

**Competing interests**

The authors declare no competing interest (both financial and non-financial).

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