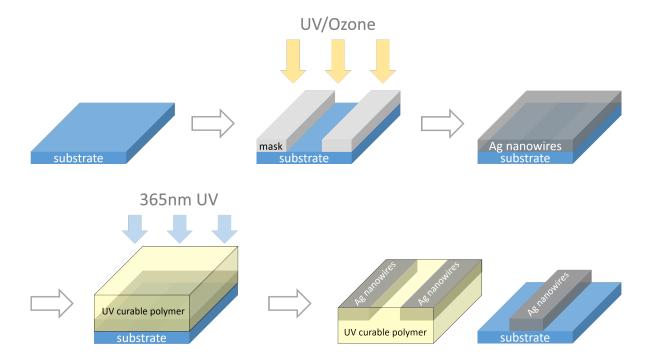
# Supplementary Information

# Critical work of adhesion for economical patterning of silver nanowire-based transparent electrodes

Dongwook Ko,<sup>a</sup> Bongjun Gu,<sup>a</sup> Seok Ju Kang,<sup>b</sup> Sungjin Jo,<sup>c</sup> Dong Choon Hyun,<sup>d</sup> Chang Su Kim,<sup>e</sup> Jongbok Kim<sup>\*a</sup>

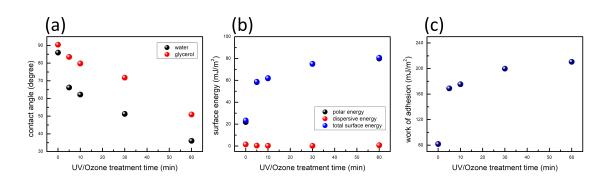
# <u>A new lithographic method for simultaneously patterning two types of AgNW-based</u> <u>flexible transparent electrodes in a single process</u>



**Fig. S1.** Experimental procedure to pattern AgNW-based transparent electrodes by controlling surface adhesion between AgNWs and the substrate.

To simutaneously construct AgNW-embedded and AgNW-exposed fleixble transparent electrodes with a desirable pattern in a single process, we prepared the substrates with low surface energy and weak adhesion property (PMMA-coated, PET or acryl substrates). After selectively masking them, we modified the surface properties by the UV/ozone, low-pressure oxygen plasma or air plasma treatment. Then, we spin-coated AgNWs on the entire substrate. After drying the AgNWs, we spin-coated NOA 63 on the AgNW film and cured it by 365nm

UV. When detaching NOA 63 from the substrate, different surface energies and works of adhesion induced selective embedding of AgNWs to NOA 63, generating a AgNW-embedded electrode with a desirable pattern on the top and a AgNW-exposed electrode with a desirable pattern on the bottom in a single process.



The surface energy and adhesive force of PET substrates

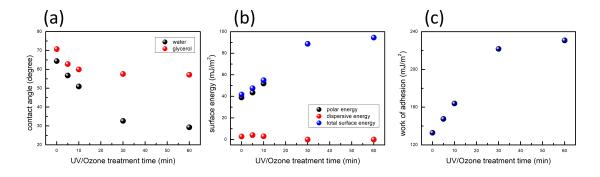
**Fig. S2.** (a) Contact angles of DI water and glycerol on the PET substrates and (b) their polar, dispersive and total surface energies depending on the UV/ozone treatment time. (c) work of adhesion between AgNWs and the PET substrates with UV/ozone treatment.

To examine the surface energy of the PET substrates with UV/ozone treatment time, we measured the contact angles of DI water and glycerol on the PET substrates. Fig. S2a shows the contact angles of DI water and glycerol on the PET substrates depending on the UV/ozone treatment time. Prior to UV/ozone treatment, the contact angles of DI water and glycrol were 86.0° and 90.45°, respectively. The UV/ozone treatment sequentially decreased the contact angles, eventually reaching a DI water contact angle of 36.1° and a glycerol contact angle of 51.0° after the UV/ozone treatment for 60 min. Then, we calculated their polar and dispersive surface energies from the contact angles of DI water and glycerol by the Owens-Wendt model. Fig. S2b represents the polar, dispersive and total surface energies of the PET substrates with UV/ozone treatment time. The polar surface energy of the PET substrates increased with UV/ozone treatment time, while the dispersive surface energy were comparable regardless of UV/ozone treatment time. Thus, total surface energy increased from 23.4 to 80.8 mJ m<sup>-2</sup> after

the UV/ozone treatment for 60 min. The adhesive force between the AgNW film and PET substrates increased with increasing the surface energy of PET substrates. It was 81.8 mJ m<sup>-2</sup> prior to UV/ozone treatment, while it reached 210.6 mJ m<sup>-2</sup> post to the UV/ozone treatment for 60 min.

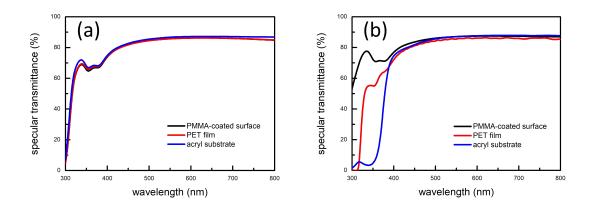
## The surface energy and adhesive force of acryl substrates

Fig. S3a and S3b show the contact angles of DI water and glycerol, and the polar, dispersive, and total surface energies of the acryl substrates depending on the UV/ozone treatment time. As-prepared acryl substrate represented a DI water contact angle of  $64.4^{\circ}$  and a glycerol contact angle of  $70.7^{\circ}$ , indicating a polar energy of  $39.0 \text{ mJ m}^{-2}$ , dispersive energy of 2.8 mJ m<sup>-2</sup>, and total surface energy of  $41.7 \text{ mJ m}^{-2}$ . The UV/ozone treatment decreasd the contact angles of DI water and glycerol, which induced significant increase of the polar surface energies and slight decrease of the dispersive surface energies. Thus, total surface energies increased to  $94.5 \text{ mJ m}^{-2}$  after the UV/ozone treatment for 60 min. The work of adhesion between the AgNWs and acryl substarates continued to increase with UV/ozone treatment. While as-prepared acryl substarate had the adhesive force of  $132.8 \text{ mJ m}^{-2}$ , the acryl substrate represented the adhesive force of  $230.7 \text{ mJ m}^{-2}$  after the UV/ozone treatment for 60 min.



**Fig. S3.** (a) Contact angles of DI water and glycerol on the acryl substrates and (b) their polar, dispersive and total surface energies depending on the UV/ozone treatment time. (c) work of adhesion between AgNWs and the acryl substrates depending on the UV/ozone treatment time.

The transmittance of AgNW-embedded and AgNW-exposed transparent electrodes



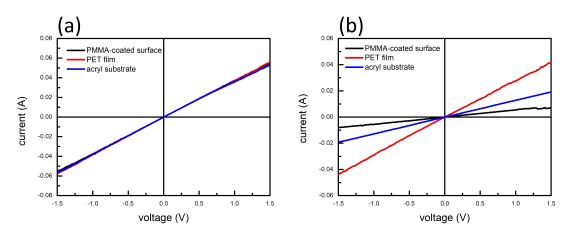
**Fig. S4.** The specular transmittance of (a) AgNW-embedded and (b) AgNW-exposed transparent electrodes. AgNW-embedded electrodes were prepread using three different substrates including the PMMA-coated substrate, PET substrate and acryl substrate. AgNW-exposed electrodes were also constructed by coating the AgNW solution on three different substrates (PMMA-coated surface, PET substrate and acryl substrate).

We prepared three different AgNW-embedded and three different AgNW-exposed transparent electrodes using the PMMA-coated substrate, PET substrate and acryl substrate as a base and characterized their specular transmittance. Specifically, we spin-coated a AgNW solution on three different substrates at 1200 rpm for 1 min. After drying them, we coated the NOA 63 and cured it. Peeling it off from three differnt substrates completed to construct three different AgNW-embedded transparent electrodes. In addition, we constructed three different substrates at 1200 rpm for 1 min and drying them. Then, we measured their transmittance by UV/visible spectroscopy. Fig. S4a shows the specular transmittance of three differet AgNW-embedded transparent electrodes. Since they were made of the same conductive ink and UV-curable polymer, it was reasonable to have comprable transmittance at all wavelength. On the other hand, the transmittance of three differet AgNW-exposed electrodes looked different as shwon in Fig. S4b. While a AgNW-exposed electrode on a PMMA-coated substrate showed the hightest average transmittance, an acryl substrate resulted in the lowest average

transmittance. Becuase incident light should pass through different bases and they have different transmittance, it is reasonable three different AgNW-exposed electrodes had different transmittance. However, the transmittance in the visible range were comparable.

#### The electrical property of AgNW-embedded and AgNW-exposed transparent electrodes

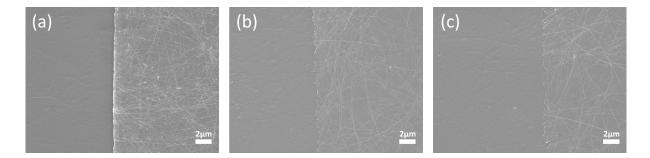
We also meausred current-voltage (I-V) characteristics of three different AgNW-embedded and three different AgNW-exposed transparent electrodes to compare their electrical property (Fig. S5). When comparing the I-V curve of three differnt AgNW-embedded electrodes (Fig. S5a), they looked comparable, indicating they have similar electrical resistance. The electron can flow through the AgNW network or the space without the AgNWs in AgNW-based transparent electrode. Becuase all AgNW-embedded electrodes were composed of the AgNWs and NOA63, they had identical electron pathway with same electrical resistance, resulting in comparable I-V characteristics and electrical resistance. However, AgNWexposed electrodes shows different I-V characteristics. While a AgNW-exposed electrode on a PMMA-coated substrate showed the highest electrical resistance, a AgNW-exposed electrode on a PET film represented the lowest electrical resistance. Although three different electrodes were made of AgNWs, AgNWs spin-coated on different substrates with different electrical resistance. Thus, they resulted in differnt I-V characteristics and electrical resistance.



**Fig. S5.** I-V characteristics of three three different AgNW-embedded and three different AgNW-exposed transparent electrodes

## Various surface treatments to pattern the AgNW-based transparent electrodes

To control surface energies of the substrates in the above-mentioned lithographic process, we introduced three different surface treatments (UV/ozone, low-pressure oxygen plasma and air plasma). Fig. S6 shows SEM images of AgNW-embedded flexible transaprent electrodes that were patterned by different surface treatemtns; (a) UV/ozone, (b) low-pressure oxygen plasma and (c) air plasma. Regardless of surface treatment methods, we observed AgNW-embedded electrodes with a clear pattern, indicating the versatility of new patterning process in flexible electronics.



**Fig. S6.** SEM images of AgNW-embedded transparent electrodes which were patterned by controlling surface adhesion. The surface adhesion was controlled by (a) UV/ozone, (b) low-pressure oxygen plasma and (c) air plasma treatment, respectively.

### Various AgNW films with different surface energy for a new patterning process

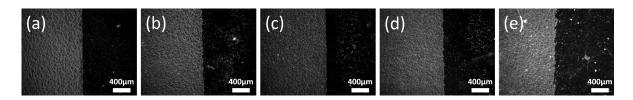


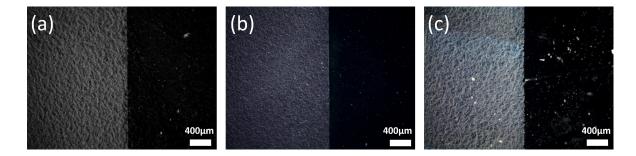
Fig. S7. OM images of AgNW films with different surface energies after performing a new patterning process. The surface energies of AgNW films were (a) 141.6 mJ m<sup>-2</sup>, (b) 163.9 mJ m<sup>-2</sup>, (c) 174.5 mJ m<sup>-2</sup>, (d) 180.2 mJ m<sup>-2</sup> and (e) 193.5 mJ m<sup>-2</sup>, respectively.

To investigate the versatility of new patterning process for various AgNW films, we prepared five different AgNW films with different surface energies by chaning AgNW densities and exmined whether a new patterning process can pattern those AgNW films. Fig.

S7 shows OM images of AgNW-embedded transparent electrodes that were prepared with different AgNW films having different surface energies. It confirmed that the new patterning process is applicable to various AgNW films having different surface energies.

# Various substrates for a new patterning process

We tested three different substrates (PMMA-coated substrate, PET substrate and acryl substrate) as a substrate to pattern the AgNW-based transparent electrodes by a new patterning process. Fig. S8 shows OM images of the AgNW-embedded transparent electrodes that were patterned on different substrates by a new patterning process. Becuase all substrates have low surface energies and weak adhesions without surface treatment, selective surface treatment to increase adhesive force between the substrate and AgNW film succeeded to pattern AgNW films.



**Fig. S8.** AgNW films that were patterned on different substrates. (a) PMMA-coated glass substrate, (b) PET substrate and (c) acryl substrate.