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## Highly flexible and transparent InSnO/Ag nanowire/InSnO hybrid electrodes

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## ABSTRACT

We have investigated the electrical, optical, structural, and mechanical properties of InSnO (ITO)/Ag nano wire (NW)/ITO hybrid films on a colorless polyimide substrate as a function of the number of repeated brushpainting cycles for the Ag NWs. By direct brush-painting of the Ag NWs, we effectively embedded the Ag NWs between sputtered ITO films. Due to the high conductivity of a brush painted Ag NWs, the optimized ITO/Ag NW/ITO hybrid films had a low sheet resistance of 12.28  $\Omega$ /square and an optical transmittance of 82.71%. Furthermore, the high strain failure of Ag NWs led to superior flexibility of the ITO/Ag NW/ITO hybrid films compared to single ITO films. The smaller inner and outer bending radius of the ITO/Ag NW/ITO hybrid film compared to single ITO film indicates that embedment of Ag NWs between thin ITO films is a promising solution for improving the flexibility of brittle ITO films.

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### 1. Introduction

Transparent conducting electrodes (TCEs) with low resistivity and high transmittance have been considered as a key component in flat panel displays, photovoltaics, and touch panel screens [1–5]. The rapid advancement of flexible optoelectronics requires properties of TCEs including superior flexibility and room temperature processing as well as low resistivity and high transmittance. Although In<sub>2</sub>O<sub>3</sub>-, ZnO-, and SnO<sub>2</sub>-based transparent conducting oxide films have been employed as main TCE materials, their poor flexibility has been considered a critical problem [6-12]. To solve the problems associated with conventional oxide TCEs, several flexible TCEs such as conducting polymer [7], carbon nanotube (CNT) [8], graphene [9], Ag nanowire (Ag NW) [10,11], and metal grid [12] have been suggested as replacements for the conventional oxide based TCEs. However, each transparent electrode still has a critical problem prohibiting its substitution for conventional InSnO (ITO) electrodes. Recently, hybrid TCEs have been extensively investigated because they combine the merits of the several transparent materials. For example, the oxide-Ag-oxide hybrid multilayer exhibited a very low resistivity of  $2.02 \times 10^{-5} \Omega$ -cm and a high transmittance of 86.54% due to the combined advantages of the Ag metal layer and the top and bottom oxide layer [13]. In our previous work, we reported on the fabrication of a PEDOT:PSS (poly (3,4-ethylene dioxylenethiophene):poly (styrene sulfonic acid))/Ag nanowire (NW)/PEDOT:PSS hybrid electrode that exploited the high conductivity of the Ag NWs and the flexibility of the PEDOT:PSS layer [14]. Choi et al. also reported that Ag nanowire embedded ITO films were applied in flexible organic solar cells as a transparent hybrid electrode [15]. Although the importance of the

0040-6090/\$ - see front matter © 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.tsf.2013.10.028 hybrid transparent electrode is well recognized, the effect of Ag NW density on the electrical, optical, mechanical, and morphological properties of the ITO/Ag NW/ITO hybrid electrode has not been investigated in detail. In addition, to solve the critical problems of Ag NW network electrode, such as poor adhesion, high surface morphology, and low process temperature, the development of the Ag NW and ITO film hybrid electrodes is imperative.

In this work, we report on highly flexible and transparent ITO/Ag NW/ITO hybrid electrodes fabricated by brush painting and DC magnetron sputtering processes. The effect of brush cycles on the electrical, optical, mechanical, and morphological properties of the ITO/Ag NW/ITO hybrid electrodes prepared on a colorless polyimide (CPI) substrate was investigated in detail. In addition, we compared the flexibility of the ITO/Ag NW/ITO hybrid electrode with that of conventional ITO films to determine the potential of the ITO/Ag NW/ ITO hybrid electrode as a promising next generation TCE.

## 2. Experimental details

The Ag NW-embedded ITO films (ITO/Ag NW/ITO) were prepared on a colorless polyimide (CPI) substrate (Mitsubishi) by combining DC sputtering and simple brush painting processes. Fig. 1 is a schematic of the steps taken to fabricate the flexible ITO/Ag NW/ITO hybrid electrode on the CPI substrate. The thin bottom ITO layer (25 nm) was sputtered on the CPI substrate using a conventional ITO (SnO<sub>2</sub>: In<sub>2</sub>O<sub>3</sub> = 10: 90 wt.%) ceramic target at a constant DC power of 100 W, Ar/O<sub>2</sub> flow ratio of 20/0.3 sccm, and working pressure of  $2.026 \times 10^2$  Pa. After deposition of the 25 nm thick bottom ITO layer, Ag NWs were coated on the bottom ITO layer by a simple brush painting process using a general paint brush. The Ag NW inks were prepared by a typical polyol synthesis method. To improve the wetting properties of the Ag NW inks, the surface of the CPI substrate was treated using atmospheric plasma

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Fig. 1. Fabrication process of flexible and transparent ITO/Ag NW/ITO hybrid electrode combining simple brush painting and DC magnetron sputtering.

before the Ag NW brush painting. The brushing speed was kept constant at 2 cm/s. After brushing of the Ag NWs, the samples were dried on a hot plate at 150 °C for 5 min to remove the solvent. The details of the brush painting process and Ag NW ink were described in our previous works [11,14]. By brushing the Ag NWs multiple times, we achieved  $2 \text{ cm} \times 2 \text{ cm}$  Ag NW network coated ITO films at room temperature and atmospheric pressure. To control and optimize the optical and electrical properties of the ITO/Ag NW/ITO hybrid electrodes, the Ag NW network was formed as a function of the number of brushing cycles ranging from one cycle to four cycles. After brush painting of the Ag NWs, the top ITO layer was sputtered on the Ag NW-covered bottom ITO films under the same sputtering conditions. The electrical and optical properties of the ITO/Ag NW/ITO hybrid electrodes were examined using a Hall measurement and a UV/visible spectrometer as a function of Ag NW brush cycles. The microstructural properties of the optimized ITO/Ag NW/ITO hybrid electrodes were analyzed by high resolution transmission electron microscope (HRTEM: JEM-2100F, 200 keV). In addition, the surface morphology of the ITO/Ag NW/ITO hybrid electrodes was investigated using a field effect scanning electron microscope (FESEM: LEO SUPRA 55). The root-mean-square (RMS) roughness and topography of the ITO/Ag NW/ITO hybrid electrodes were analyzed by atomic force microscopy (AFM: PUCO Station STD). We analyzed the surface properties of ITO/Ag NW/ITO using non-contact mode. The mechanical integrity of the ITO/Ag NW/ ITO hybrid electrodes was evaluated by inner and outer bending tests as a function of bending radius. The bending test system was able to automatically measure the change in resistance of the flexible ITO/Ag NW/ITO hybrid electrodes during the outer or inner bending tests. By repeated linear motion of the Cu plates, we were able to measure the change in resistance ( $\Delta R$ ). The bending radius and frequency were approximately 5 mm and 1 Hz, respectively.

## 3. Results and discussion

Fig. 2(a) shows the sheet resistance and resistivity of ITO/Ag NW/ITO hybrid electrodes as a function of brush painting cycles with constant bottom and top ITO layers. It is evident that the combination of conductive Ag NWs with very thin ITO film (25 nm) significantly decreased the total sheet resistance and resistivity of the ITO/Ag NW/ITO hybrid electrode as in a conventional oxide-metal-oxide layer electrode [15]. Because the well-connected Ag NW network can act as a main current path in the ITO/Ag NW/ITO hybrid electrodes, the insertion of the Ag NWs effectively reduces the total sheet resistance

and resistivity. The reduced sheet resistance and resistivity of the ITO/ Ag NW/ITO electrode could be attributed to the increased carrier concentration with increasing brush cycles as shown in Fig. 2(b). Because the embedded Ag NWs could supply carriers into the ITO films, the ITO/Ag NW/ITO hybrid electrodes exhibited higher carrier concentration with increasing brush cycles.



Fig. 2. (a) Sheet resistance, resistivity, (b) mobility, and carrier concentration of ITO/Ag NW/ITO hybrid electrodes prepared with an increasing number of brush cycles.

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