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Flexible powder electroluminescent device on silver nanowire electrode

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ABSTRACT

We have demonstrated the flexible AC powder electroluminescent device based on Ag nanowire electrode. The Ag nanowire electrode showed the nanowire morphology of 20 nm in diameter and 15 μm in length, the transmittance of 87%, and the sheet resistance of 50 Ω/sq, and the higher flexibility than the conventional ITO substrate. The electroluminescence spectra of the Ag nanowire-based device in all frequency and voltage ranges were almost similar with the ITO-based device. In comparison with the ITO-based device, the luminous efficiency of the Ag nanowire-based device was almost same as 1.53 lm/W.

1. Introduction

Alternating-current powder electroluminescent devices (ACPELDs) have a high potential for commercial application, because of uniform light emission, low power consumption, screen printing process with low-cost production, light weight, and flexible form. ACPELDs can be used in portable electronic devices, wallpaper-like solid state lighting, and display screens together with the possibility of a thin architecture (60–100 μm) [1]. An efficient electroluminescence (EL) phosphor (doped zinc sulfide) is excited by charges injected between the front transparent and the rear electrodes in a strong electric field ($\sim 10^6$ V/cm). Dopants on ZnS host allow color tuning over the whole visible range: blue for Cu–Cl, green Cu–Al, and orange for Mn [2]. The conventional indium–tin oxide (ITO) transparent conducting oxides for ACPELDs have been prepared by cost-intensive sputtering techniques. However, low-temperature deposition techniques for flexible polymer substrates such as polyethylene terephthalate (PET) result in higher sheet resistance and surface roughness [3]. Moreover, continuous bending of ITO electrodes leads to cracking and delamination, which are limiting their flexibility [4]. Carbon nanotube (CNT) electrode has been tried as an alternative for ITO electrodes. However, its high sheet resistance ($\sim$kΩ/sq) has been a major challenge for performance of thin-film devices on it [5], even though it was adopted to thin-film device such as organic light-emitting diode (OLED) and thick-film device such as ACPELD [3]. Also, silver nanowire (Ag NW) electrodes are a promising alternative to transparent conductive oxides. The Ag NWs have attracted attention to a large number of applications in the area of optoelectronics and macroelectronics because of their low sheet resistance ($\sim$10 Ω/sq) and high flexibility [6,7]. The Ag NWs can be synthesized by batch chemical reactions, and coated from solution at ambient temperatures [6]. High surface roughness of the Ag NWs film ($>$10 nm) has still given some problems for the performance of thin-film devices, even though it was reported to apply to OLED [7].

In this paper, the transparent Ag NW electrode, for the first time, was applied to flexible AC powder electroluminescent device (ACPELD). The Ag NW electrode with the transparency of about 87% and sheet resistance of 50 Ω/sq was deposited through a bar-coating process, and phosphor and dielectric layers were made through a screen-printing process. The EL spectrum and charge density for Ag NW-based ACPELD were compared with that for ITO-based ACPELD.

2. Experimental details

The ACPELDs were prepared by a screen printing method. The Ag NW-based device consists of a PET substrate/Ag NW electrode/phosphor layer/insulating layer/and rear metallic electrode. At first, Ag NWs (20 nm in diameter, 15 μm in length; AIDEN in Korea)

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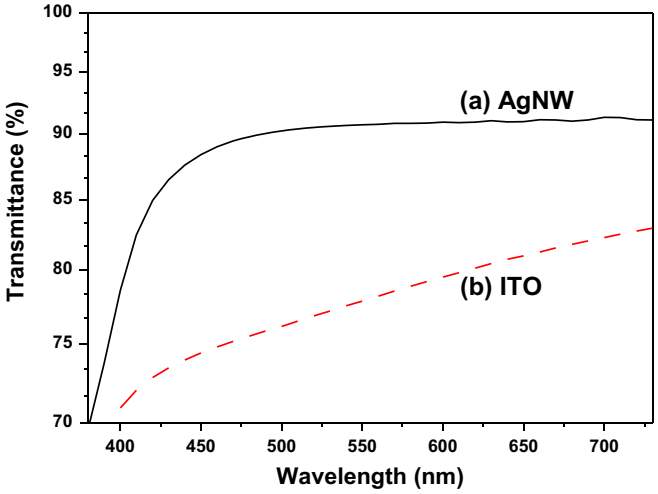

Fig. 1. Transmission spectra of (a) Ag nanowires and (b) ITO PET substrates.

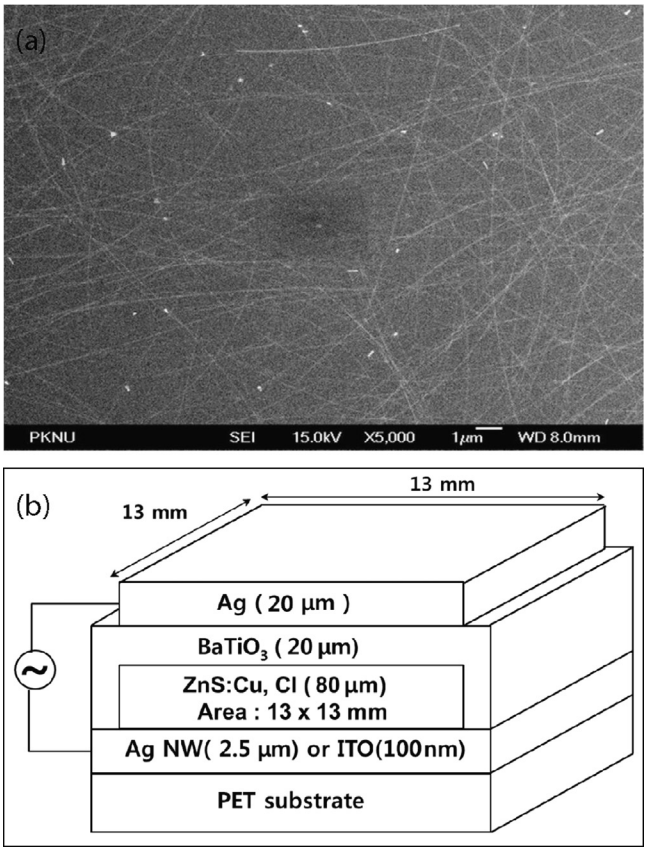

Fig. 2. (a) Top-view SEM image of Ag nanowires and (b) schematic of EL device.

together with hydroxypropyl methylcellulose (HPMC) with mixture ratio of 1.5% and 87.5% in weight were dispersed in deionized water, respectively. The homogeneous dispersion was mixed with a small amount of adhesive, and then was bar-coated on PET substrate heated at 100 °C. The coated Ag NW electrode was baked at 120 °C for 60 s. The resulting coating was about 2.5 μm thick with a sheet resistance of 50 Ω/sq and a transparency of about 90% as seen in Figs. 1 and 2(a). Subsequently ZnS:Cu, Cl powder phosphor (Osram) were dispersed in dielectric cyanoethylcellulose binder with a weight ratio of 6:4 and deposited on the Ag NW PET substrate by a screen printing method. The coated phosphor was then dried for 30 min at 80 °C. The phosphor layer is about 80 μm thick. The high dielectric $BaTiO_3$ powder was mixed with the organic binder with weight ratio of 2:1, and then printed upon the phosphor layer to protect against the electric breakdown. The thickness of dielectric layer is 20 μm. The Ag paste as a rear electrode was screen-printed upon the dielectric layer. As a reference, the ACPELD on a conventional ITO PET substrate (100 nm thick with a sheet resistance of 50 Ω/sq and a transparency of about 75%) was fabricated through the same process as seen in Fig. 2(b). The luminescent areas of all EL devices under comparison were kept at 1.3 cm × 1.3 cm. The total thickness reproducibility of EL device using the screen printing procedure was estimated to be about 5 μm (~5% of total phosphor and dielectric thickness).

The low-voltage capacitances of Ag NW and ITO-based EL devices were 0.43 nF ± 0.02 nF and 0.49 nF ± 0.03 nF at 400 Hz and 1.0 V, respectively. All EL cells were tested in not-aged fresh condition. Applied voltage (V) and charge density (Q) under the sinusoidal voltage were measured using a conventional Sawyer–Tower circuit with a sense capacitor of 0.46 μF at frequency of 400 Hz [8]. The electroluminescence (EL) spectra were measured by using a spectrometer (Ocean Optics, USB4000). The transmittance spectra of Ag NW and ITO PET substrates were measured using a UV–vis spectrophotometer (Shimadzu, UV-3150). A field emission scanning electron microscope (FE-SEM, JEOL, JSM-6700F) was used to obtain surface image of the Ag NW arrays on PET substrate. Time chart measurement was performed to obtain variations in EL intensity and applied voltage as a function of time by using oscilloscope (Tektronix, TDS 2012B) in which EL intensity was recorded on channel 1 by using a silicon photodiode (ORIEL, 71608) and applied voltage was recorded on channel 2.

3. Results and discussion

Fig. 1 shows the transmittance spectra of Ag NW PET substrate, and commercially purchased ITO PET substrate as a reference. The Ag NW substrate is more transparent than ITO substrate in all near-ultraviolet and visible regions. At the 505 nm wavelength of EL peak, the transmittance of the Ag NW substrate (87%) is greater than that of ITO substrate (75%). The transmittance of the Ag NW electrode decreases at shorter wavelengths, but is still greater than 70% at 400 nm.

Fig. 2(a) shows FE-SEM surface image of Ag NW PET substrate. The Ag NW is 20 nm in diameter and 15 μm in length. The coverage of Ag NW in the entire surface area is less than 10%, such that the Ag NW substrate is transparent. Sheet resistance (50 Ω/sq) was obtained through the formation of Ag NW networks.

Fig. 3 shows sheet resistances of Ag NW and ITO PET substrates with increasing numbers of bending–recovery cycles. The substrates were bent to concave and convex curvatures with 5.0 mm radius. After 1000 cycles of bending and recovery, the resistances of Ag NW substrate show a slight change within a standard deviation of about 8% compared to the fresh device. On the other hand, the gradual degradation from start was observed at ITO PET substrate, and after 1000 cycles the resistances of ITO PET substrate show a significant increase by about 50% compared to the fresh device. The degradation mechanism can be understood in terms of cracking of ITO electrode during flexibility test. In addition, we did flexibility tests of both Ag NW and ITO-based ACPELDs under the same condition; however, we could not obtain any reproducible results (not included in this paper) even before 100 cycles. It is suggested that the interfaces among the thick layers of phosphor, dielectric, or rear electrode experience more bending curvatures and more strains than that between the thin Ag NW and the PET film layer, and thus the former interfaces are earlier delaminated than the latter so as to not sustain the normal EL performance.

The EL spectra of Ag NW and ITO-based ACPELDs varying from 20 to 500 V for a fixed frequency of 400 Hz are shown in Fig. 4(a).