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Fabrication of substrate-free double-side emitting flexible device based on silver nanowire-polymer composite electrode

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ABSTRACT

Flexible light emitting diodes are a promising component for future electronic devices, but require a simple structure and fast fabrication method. Organic light emitting diodes are a viable option as they are lightweight, thin, and flexible. However, they currently have costly fabrication procedures, complicated structures, and are sensitive to water and oxygen, which hinder widespread application. Here, we present a novel approach to fabricate flexible light emitting devices by employing Ag nanowire/polymer composite electrodes and ZnS phosphor particles. The composite electrode was fabricated using inverted layer processing, and used as both a bottom electrode and a dielectric layer. The high mechanical stability of the composite allowed the device to be free standing and mechanically flexible, eliminating the need for any additional support. Using Ag nanowires in both the top and bottom electrodes made a double-sided light emitting device that could be applied to wearable lightings or flexible digital signages.

1. Introduction

Organic light emitting diodes (OLEDs) have been extensively researched in the last two decades because of their impressive properties; they are lightweight, thin, have intrinsic flexibility, low-voltage operation, and a high luminous efficiency [1–8]. The device properties depend on the structural configuration, which can be optimized to attain some unique characteristics. For instance, using a polymer film thinner than 10 μm as a substrate can result in extremely flexible devices with a bend radii less than 1 mm [9]. However, important processing and structural challenges need to be addressed in order to achieve flexible OLEDs. For example, the surface roughness of the transparent electrode should be sufficiently smooth to prevent vertical shorts or leakages [10]. Considering that some candidate materials for replacing the brittle indium tin oxide (ITO) conducting layer, e.g., carbon nanotubes, reduced graphene oxides, and silver nanowires (AgNWs), function based on their low density percolation, roughness is one of the most important challenges to resolve. Other functionalities, such as transparency or double-sided luminance [11,12] can be attained by tuning the OLED structures, but only if the diode materials (consisting of at least four or five layers of organic compounds) and fabrication scheme are fully optimized. The design/synthesis of materials and their stacking for device fabrication are rather difficult tasks, because there are many optimization parameters to consider, e.g., the work function or energy level, color, transmittance, life time, carrier transporting or blocking efficiency, and stability of each materials and their interfaces. In addition, current highly efficient OLEDs are based on small molecule materials that are generally fabricated by fully dry-processed apparatuses, which means that manufacturing costs are very high and fabrication is limited to a few large companies. Sensitivity to water and oxygen has been another inherent drawback of widespread OLED application [13,14].

For practical application of flexible displays, the device fabrication scheme should be simple and include solution processing. This would significantly lower the fabrication costs and facilitate practical flexible display applications with smaller markets, such as decorative, architectural, bio-medical, and military devices. Some previous studies reported that an inorganic LED can be a possible solution to this issue because of its rather moderate sensitivity to oxygen and surface roughness of the electrode [15,16]. However, a complicated procedure to achieve reliable LEDs on a flexible or stretchable substrate hinders its widespread use in fabrications of

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flexible displays. Another candidate is an electroluminescent phosphor (e.g., doped ZnS dispersed in an organic binder) sandwiched between two electrodes. Such a device can be fabricated in a simple manner and is excited in a strong electric field commonly driven by applying an alternating current [17–20].

Here, we present a novel approach to fabricate a flexible, double-sided light emitting device by employing alternating current electroluminescence (ACEL) with an AgNW/colorless polyimide (cPI) composite electrode. This composite acts both as a single side electrode and dielectric layer, and does not require an additional substrate due to the high modulus of elasticity and high strength of the cPI. Using the same AgNWs for the other electrode produced a device with double-sided emission.

2. Material and methods

A schematic of the device fabrication scheme is shown in Fig. 1a. A glass substrate was first cleaned sequentially with detergent, deionized water, and isopropanol, and then an AgNW dispersion (Nanopyxis Ltd., Korea) with an average wire diameter and length of 30 nm and 20 μm, respectively was spin-coated and then dried by an infrared light emitting diode for 5 min. It was then spin-coated with a varnish of cPI (Kolon Industries Inc., Korea). The sample was annealed at 200 °C for 1 h to form a cPI film of 15 μm in thickness on the glass. ZnS particles (National EL Technology, Korea) dispersed in isopropyl alcohol were spin coated onto the cPI and heated at 120 °C for 30 min to form an electroluminescent layer with a thickness of 45 μm (Fig. 1b). Then the AgNW dispersion was spin-coated once more on the cured ZnS layer to form the top electrode of the device, followed by IR irradiation to remove any remaining solvent (Fig. 1c). Finally, the fabricated device was peeled off the glass, resulting in a flexible light emitting device (an SEM micrograph of the bottom electrode is shown in Fig. 1d). To achieve effective peel-off, the samples were soaked in water to assist the removal of the films from the supporting glass substrates by the hygroscopic swelling of the cPI.

The optical transmittance of the films was measured using a UV–visible spectrophotometer (Jasco V-560, Japan), while the sheet resistance (R_s) was measured with a non-contact measurement system (EC-80P, Napson Corp., Japan). The surface morphology was analyzed using atomic force microscopy (AFM; XE-100TM, Park Systems, USA). A field-emission scanning electron microscope (FESEM; JSM6700F, JEOL Ltd., Japan) was used to investigate the microstructures of the AgNW networks. The mechanical stability of the AgNWs/cPI was evaluated using an automatic bend-testing machine (Jaeil Optical Systems, Korea), where a bending radius of 0.5 mm was used to induce ~1.5% strain. The films were bent at a cycle rate of 0.3 Hz, with their resistance being measured during the outward bending cycles. To further evaluate the mechanical stability of the fabricated device, a smaller radius of 0.3 mm was employed to induce ~10% strain. An adhesive tape (Scotch Magic Tape, 3M, USA) was used to evaluate the adhesion between the AgNWs and cPI. An AC power source (6600 series, Extech Electronics, Taiwan) was used to power the ACEL device, and the luminance was measured by a luminance meter (LS-100, Konica Minolta, Japan).

3. Results and discussion

In order to form a typical ACEL device with inorganic phosphors, simple consecutive stacks of five layers are needed: substrate/bottom electrode/dielectric layer/emissive layer/top electrode [21]. The fundamental light-emitting mechanism of the devices is known to be solid-state cathode luminescence, in which the EL arises from impact excitation of the emitting layer by hot electrons accelerated through an inorganic oxide layer [21]. The dielectric layer between the bottom electrode and emissive layer implies that the light emission is field driven. Interestingly, the existence of the dielectric layer provides a novel possibility of achieving a simpler device architecture; if a flexible, transparent, free-standing polymer can be used to form the layer, a substrate is not required to support the device. Here, we selected cPI as a dielectric material for insulating the ZnS particles from the bottom electrode (a percolating network of AgNWs), because of its high modulus of elasticity, transparency, mechanical stability, and glass transition temperature (>350 °C) [22,23]. The high mechanical stability of the cPI

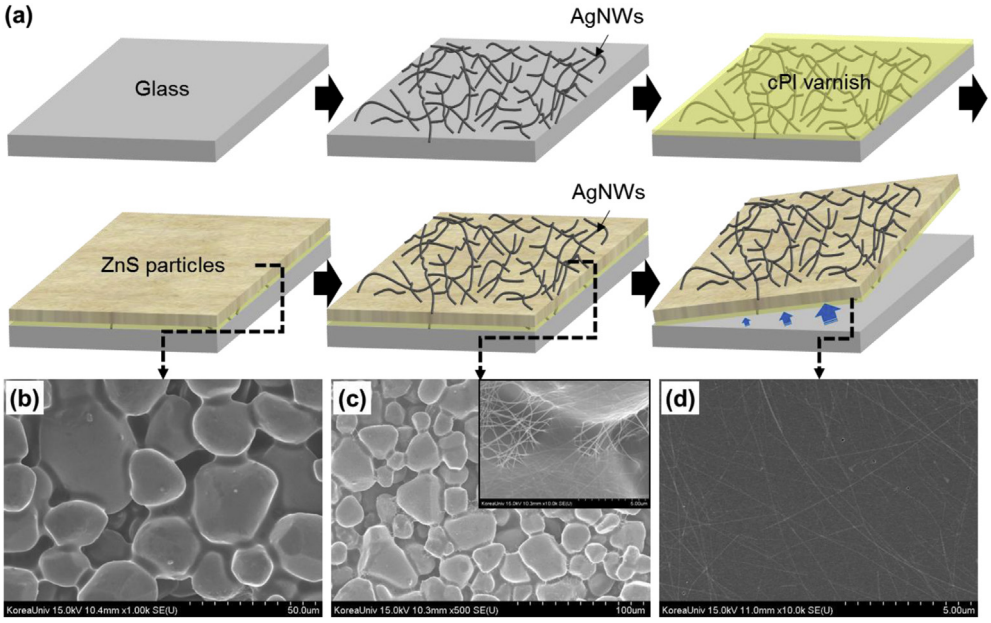

Fig. 1. (a) Schematic of fabrication procedure for flexible ACEL devices, (b) SEM micrograph of ZnS particles, (c) AgNWs deposited on top of the ZnS phosphorescent particles, and (d) fully embedded AgNWs at the cPI surface.