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Soft fabric-based flexible organic light-emitting diodes

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ABSTRACT

We reported the first organic light-emitting diodes (OLEDs) on actual soft fabrics that can be used for a wearable display. Polyurethane (PU) and poly(vinyl alcohol) (PVA) layers, which only degrade slightly the flex stiffness of bare fabrics due to their ductile characteristics, were used as planarization layers via a simple fabrication process involving lamination and spin-coating. Therefore, many of the mechanical characteristics of the bare fabric substrates were retained in the planarized fabric substrates. Non-inverted top-emitting OLEDs, designed by considering the optical microcavity effects, were fabricated on a planarized surface by thermal evaporation. The fabricated OLEDs on soft fabrics showed a high current efficiency of around 8 cd/A, reliability during a 1000 cycle bending test with a bending radius of 5 mm, and clear green emission up to an emission angle of 70°. Consequently, we developed high-performance OLEDs on very similar to real fabric via a simple universalized fabrication method.

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1. Introduction

Wearable electronics have been attracting much attention as next-generation electronics. Textile substrates have advantages such as a high flexibility and a lightweight. Also, textiles are intimate connecting material between humans and their electronic devices, and people obviously wear cloth anywhere and at any time. Thus, integrating electronic devices into the textiles has been studied for human convenience [1]. Novel applications such as sensors [2,3], thin film transistors [4], energy storage devices [5] and energy generating devices [6,7] based on textiles or fibers have been reported recently. In particular, there has been great interest in "wearable displays" as these items have the potential to open a new industry - not only of pertaining to the information display itself but also an area that converged with other electronic devices such as medical sensors and energy devices. Therefore, researchers have been amassing ways to develop wearable display

1566-1199/\$ - see front matter @ 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.orgel.2013.09.001 devices with textiles and fibers. Some prototype display devices based on textiles or fibers have been demonstrated with novel fabrication methods and materials. Examples include, organic electroluminescent devices [8], inorganic electroluminescent devices [9], ionic transition metalcomplex-based nanofibers [10], and photonic bandgap fibers [11].

The application of actual clothes fabric to textile-based electronics is a vital topic for human-friendly wearable electronics. However, there is a large practical gap between previously reported substrates for wearable electronics and actual fabrics for clothes regarding the use of a single-fiber material without a weaving process or porous woven textiles weaved with thick fibers over large meshes. Also, the low luminous efficiency and high operating voltages reported in previous works can be a barrier to humanfriendly wearable electronics. Thus, display devices with high luminous efficiency and a low operating voltage on genuine soft fabric substrates are clearly required before wearable displays can be realized.

In many previously reported papers, the organic electronic devices fabricated on flexible substrates such as



Letter





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polymers and woven meshes consisted of polymer-based solar cells [12,13], as these devices are much more stable than OLEDs due to their low current density and low operating voltages, which typically are $0-20 \text{ mA/cm}^2$ under 1 V. On the other hand, OLEDs are operated higher current density and voltage levels of 0–500 mA/cm² under 20 V. Thus, they are very unstable when fabricated on uneven surfaces and when mechanical stress is applied. As a result, there are no reports of the fabrication of OLEDs as electronic devices. In our work, we reported the first organic light-emitting diodes (OLEDs) on a commercial soft fabric that can be used for a wearable display. OLEDs are suitable for wearable displays because they offer the possibility of creating a flexible display panel and due to their high luminous efficiency, low driving voltage and light weight [14]. However, there is a crucial problem for OLEDs on fabric substrate in that OLEDs are easily broken down when fabricated on uneven substrates, as the total thickness of the organic layers between two metal electrodes is approximately 100 nm. Fabrics surface roughness of tens of micrometers originating from the weaving process, and a drastic change in the surface morphology induces short circuits or defects by cracks in OLEDs. Thus, planarization is a critical issue when fabricating OLEDs on fabrics. On the other hand, issues related to the surface roughness have to be resolved most of all, as the reliability of both the fabrication methods and the electronic devices are not guaranteed on uneven substrates. To solve the roughness problem, we developed a simple planarization process using ductile materials for the planarization of fabric substrates. Also, we endeavored to focus on examining and improving the mechanical characteristics of the fabric itself, because it is important to retain the flex stiffness of fabric substrates.

Consequently, we developed our planarization method using polyurethane (PU) and poly(vinyl alcohol) (PVA) films and designed non-inverted top-emitting OLEDs considering the optical cavity effects for high efficiency. This is the first report of the fabrication of flexible OLEDs based on commercial soft fabrics.

2. Experimental

2.1. Fabrication and characterization of substrates

Plain woven fabric made of polyester fibers was supplied by Kolon Glotec Inc. Sheet-type PU films (Kolon Glotec Inc.) were laminated under pressure at room temperature with a thickness in a range of 20–50 µm. The PU sheets have two parts: 20 µm-thick PU with low viscosity on the bottom side to ensure that the woven structure of bare fabric is smooth, and the 10 µm-thick PU with high viscosity on the upper side to create a flat surface. 99 + % hydrolyzed PVA (molecular weight 89,000-98,000 g/mol, Sigma Aldrich) was diluted in de-ionized water (DI water) for 5 wt.%. The DI-water was heated to 90 °C on a hot plate and a small dose of PVA powder was then inserted repeatedly. This was then cooled to room temperature. We activated the surface of the PU with an air assisted plasma treatment for 10 min to promote the wetting of the aqueous PVA suspension. The PVA solution was spin-coated at

3000 rpm for 40 s and then annealed on a hotplate in ambient air at 120 °C for 20 min. For multilayer films, the same process was repeated. The PU was laminated in an ambient environment, and all other processes, i.e., the plasma treatment, the spin-coating process, and thermal evaporation process, were conducted in a cleanroom environment. In addition, blowing using nitrogen was used to remove particles from the surface at every step of the fabrication process. The surface morphologies were measured on a field-emission scanning electron microscope (Silion, FEI) and by an atomic force measurement system (XE-100, Park System). Cross-section images were taken by a focused ion beam-type SEM (Quanta 3D FEG, FEI). The flex stiffness, as evaluated by a cantilever test, was measured by the Korea Textile Development Institute according to standards set by the International Organization for Standardization (ISO) [15].

2.2. Fabrication and characterization of OLEDs

Non-inverted top-emitting OLEDs were fabricated by a successive thermal evaporation process. A 50 nm-thick Ag layer was deposited at a deposition rate of 2 Å/s to form an anode. Next, a 5 nm-thick hole injection layer of tungsten oxide (WO3), a 50 nm-thick hole transport layer of N,N'-Bis(naphtanlen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB), a 40 nm-thick emission layer of tris(8-hydroxy-guinolinato)aluminum (Alg3) and a 2 nm-thick electron injection layer of 8-hydroxyguinolatolithium (Lig) were evaporated sequentially at deposition rates of 0.3 Å/s, 1 Å/s, 1 Å/s and 0.2 Å/s, respectively. Subsequently, a 1 nm-thick Al layer, a 20 nm-thick Ag layer and a 50 nm-thick NPB layer were deposited at corresponding deposition rates of 0.1 Å/s, 2 Å/s and 1.5 Å/s to form a semitransparent cathode and capping layer. The current density and the voltage were measured by a Keithley 2400 sourcemeter, and the luminance and the emission spectrum were measured by a spectroradiometer (CS-2000, Konica Minolta). To calculate the radiant intensity profile and changes of the color coordinates, the measurements were conducted at emission angles of 0°, 10°, 20°, 30°, 40°, 50°, 60° and 70° at a fixed azimuthal angle of 0°. The typical device area of one cell was 9 mm². Optical microscope images of emitting cells were taken under magnification of 60 times by a digital microscope (GE-5, View Solution Inc.). Cyclic bending tests were performed on a customized bending station (Fig. S1b, Supporting Information). The fabricated fabric-based OLEDs were attached onto a 100 µm-thick PET plastic film and then bent 1000 times with a bending radius of 5 mm. Samples were exposed to the atmosphere for about 60 min during the bending test.

3. Results and discussion

Fig. 1 depicts schematic diagram, photographs and SEM images of the fabricated fabric-based OLEDs. The overall structure of the planarized fabric substrates and the designed non-inverted top-emitting OLEDs are described with the thickness information (Fig. 1a). The purpose of

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