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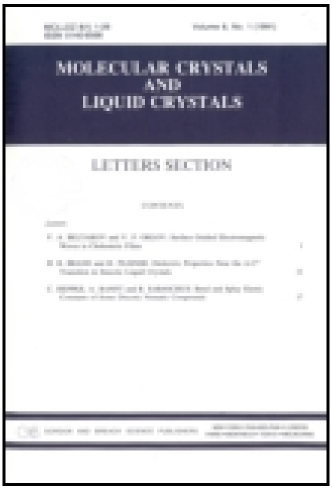

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Flexible Organic Light-Emitting Diodes with Amorphous ITO Anodes on Transparent Cellulose Substrates

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Flexible Organic Light-Emitting Diodes with Amorphous ITO Anodes on Transparent Cellulose Substrates

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We describe flexible organic light-emitting diodes (OLEDs) with indium tin oxide (ITO) anodes fabricated at low substrate temperatures using facing target sputter (FTS) on a transparent-cellulose substrates. The device structure was cellulose paper substrate/SiO_2/ITO/NPB/Alq_3:C545T/BCP/LiF/Al. The ITO thin films were deposited at substrate temperatures below 70°C. X-ray diffraction analysis revealed that the ITO thin films were amorphous. As the substrate temperature during deposition increased from 27 to 70°C, the sheet resistivity decreased from 1,400 to 120 Ω/□. The OLEDs with ITO thin film anode with sheet resistivity of 120 Ω/□ exhibited a maximum luminance of 14,543 cd/m^2 at a driving voltage of 14.75 V.

Keywords Organic light-emitting diode; flexible; cellulose paper; indium tin oxide; facing target sputter; amorphous

Introduction

Organic light-emitting diodes (OLEDs) have received considerable and growing attention over the past two decades due to their fast response, wide viewing angle, thin displays, and applications in two-dimensional lighting [1,2]. To date, most OLEDs have been fabricated on rigid glass substrates; however, glass substrates are fragile, relatively thick, and heavy. Polymer substrates are more flexible and lighter than glass substrates, and are therefore desirable for substrate of OLEDs. Flexible polymer substrates used in the fabrication of OLEDs include polyethylene terephthalate (PET), polyethylene naphthalate (PEN), polymethylmethacrylate (PMMA), polycarbonate (PC), polyethersulfone (PES), and polyimide (PI) [3–8]. However, these substrates have environmental issues. Recently, OLEDs fabricated on transparent cellulose composite substrates have received much attention because they are biodegradable [9–12].

This paper describes OLEDs fabricated on flexible-transparent substrates formed from the most abundant natural polymer on earth: cellulose. ITO thin films are commonly employed as transparent conductive anodes for OLEDs. However, the cellulose paper substrate

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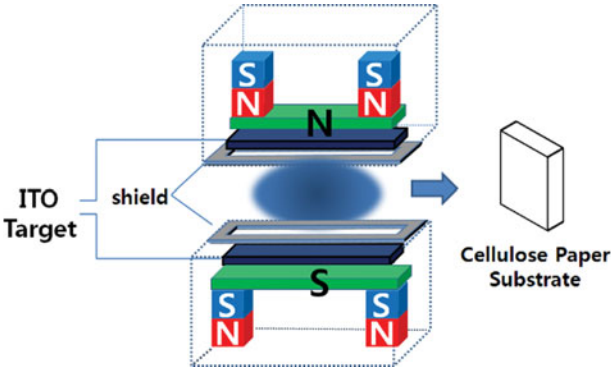

Figure 1. Schematic diagram showing the facing target sputtering system.

is heat-sensitive and loses integrity if heated to temperatures in the range 200–300°C, which are typically used in magnetron sputtering deposition of polycrystalline ITO. We deposited ITO thin films at temperatures below 70°C using facing target sputter (FTS). Since the substrate was placed away from the confined plasma, neutrally charged particles with low energy were incident on the substrate, resulting in amorphous deposition of ITO thin films over a wide range of processing conditions. Since amorphous ITO films do not exhibit the energy barriers that exist at the grain boundaries of polycrystalline materials, electrons in the amorphous ITO thin films can easily move across the thin film, resulting in low resistivity that can be exploited for an OLED substrate.

Experimental

The cellulose paper substrate was fabricated by dissolving cellulose acetate in acetone. A 100-nm-thick SiO_2 film was deposited on the cellulose paper substrate at substrate temperature of 27°C using radio frequency (RF) magnetron sputtering, and then the ITO anode was deposited using FTS through a shadow mask using two ITO targets (ITO 99.99% and In_2O_3–SnO 90:10 wt.%), with an O_2 flow rate of 0.1 sccm, an Ar flow rate of 30 sccm,

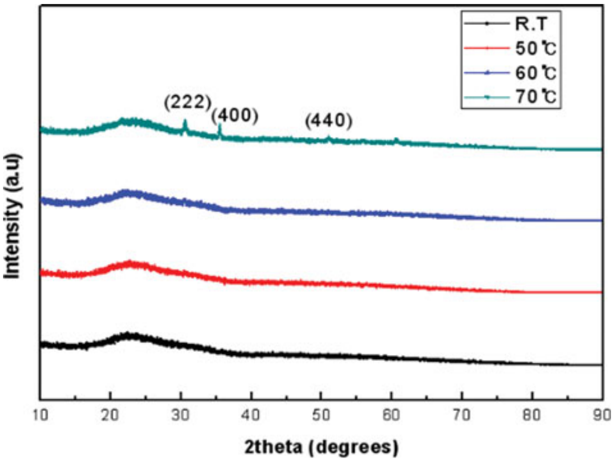

Figure 2. XRD patterns of the ITO thin films deposited at various substrate temperatures.

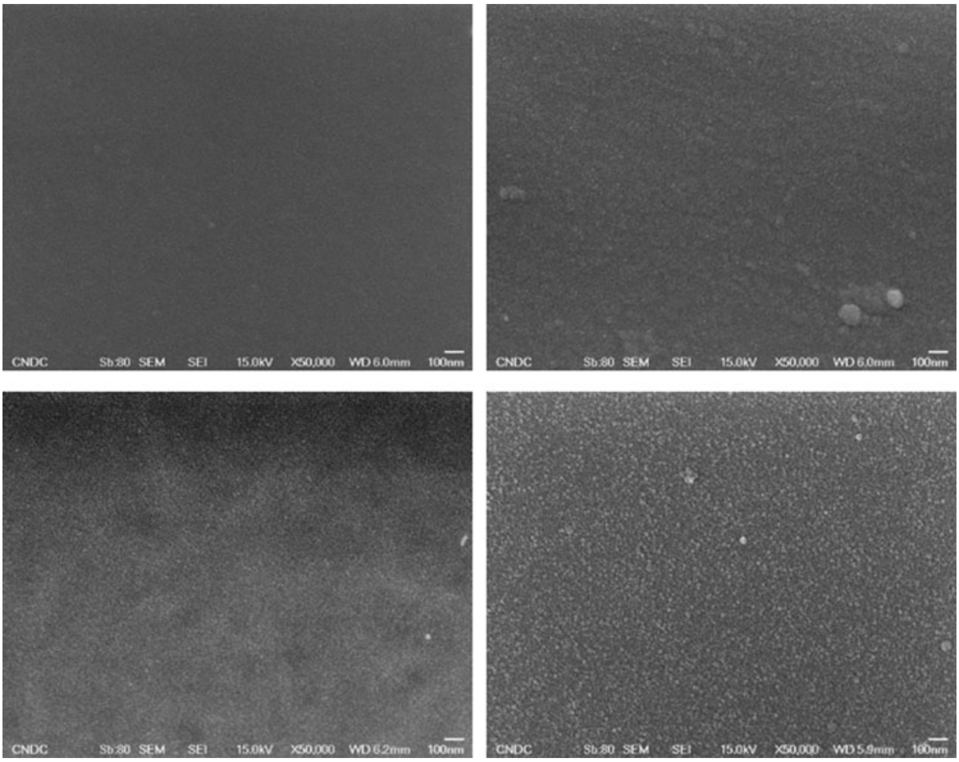

Figure 3. SEM images of the ITO thin films deposited on the SiO_2/cellulose paper substrate at substrate temperatures of (a) 27°C, (b) 50°C, (c) 60°C, and (d) 70°C.

and a base pressure of 10 mTorr. The two targets used in the FTS were located so that they faced each other, and a high-density plasma was formed between the targets, as shown in Fig. 1. The SiO_2/cellulose paper substrate was placed away from the confined plasma, so that neutrally charged sputtered particles, which were not confined in the plasma, were incident on the SiO_2/cellulose paper substrate. ITO thin films were deposited at substrate temperatures in the range of 27–70°C.

The ITO-coated substrate was cleaned using an oxygen plasma at 50 W for 5 min prior to loading into a thermal evaporation chamber with a base vacuum of 7×10^{-7} Torr. A 60-nm-thick N,N′-bis(naphthalene-1-yl)-N,N′-bis(phentl)-benzidine (NPB) layer was deposited on the amorphous ITO/SiO_2/cellulose paper substrate, followed by a 35-nm-thick tris-(8-hydroxyquinoline) aluminum (Alq_3) layer, which was co-deposited with 1 wt.% 10-(2-benzothiazolyl)-2,3,6,7-tetradudro-1,1,7,7,-ramethyl1–1H,5H,11H-[1] benzopyrano [6,7,8-ij]quinolone-11-one (C545T). Following this, a 20-, 30-, or 40-nm-thick 9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) layer was evaporated to form a range of samples with different BCP layer thicknesses. Finally, a 0.5-nm-thick LiF layer followed by a 100-nm-thick Al layer were evaporated onto the organic layers through a shadow mask. The deposition rate of the metal and organic layers was 0.1 nm/s.

The thickness of amorphous ITO thin films was measured by step profiler (Sitek Alpha-step 200). The microstructure of the films was characterized by field emission scanning electron microscope (FE-SEM, JEOL: JSM-7401F) and X-ray diffraction (XRD, BRUKER AXS D8 ADVANCE). Surface morphology of the films was also analyzed by

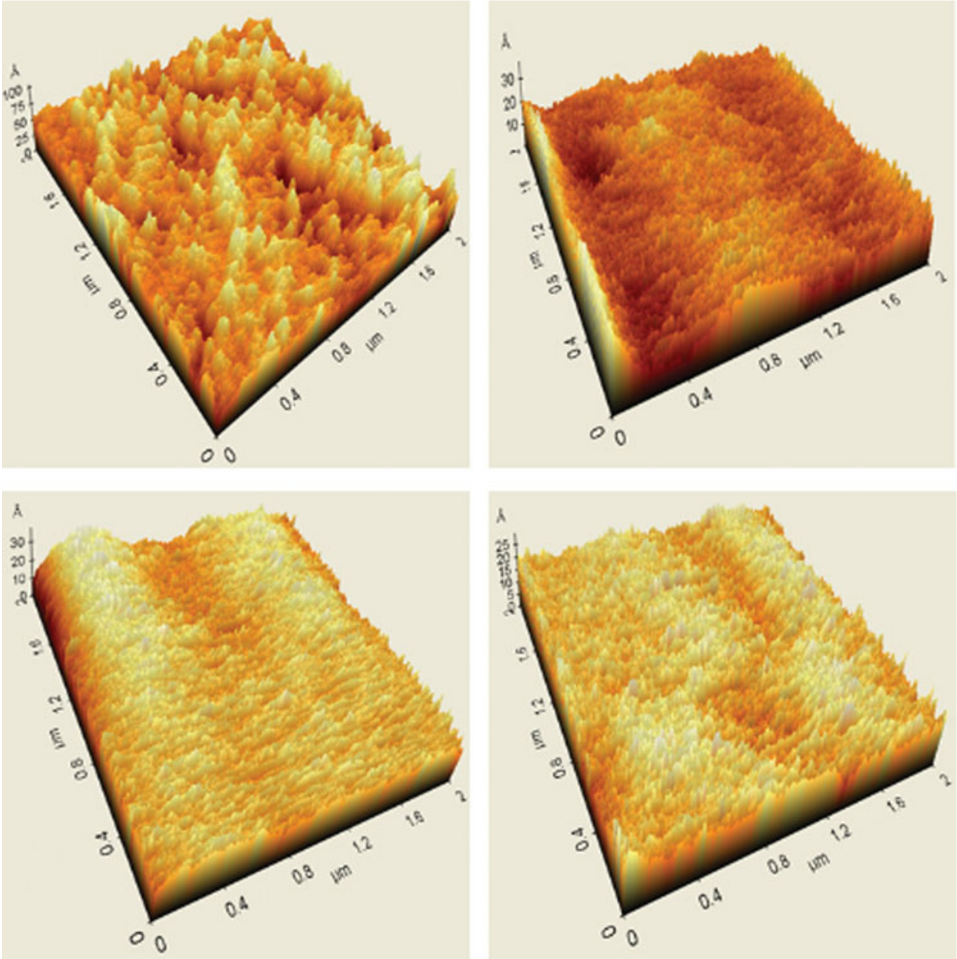

Figure 4. AFM images of the ITO thin films deposited on the SiO_2/cellulose paper substrates at substrate temperatures of (a) 27°C, (b) 50°C, (c) 60°C, and (d) 70°C.

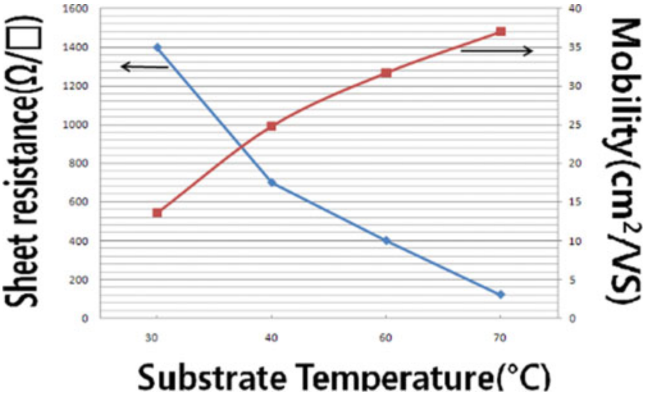

Figure 5. Charge carrier mobility and sheet resistivity of the ITO thin films deposited at various substrate temperatures.

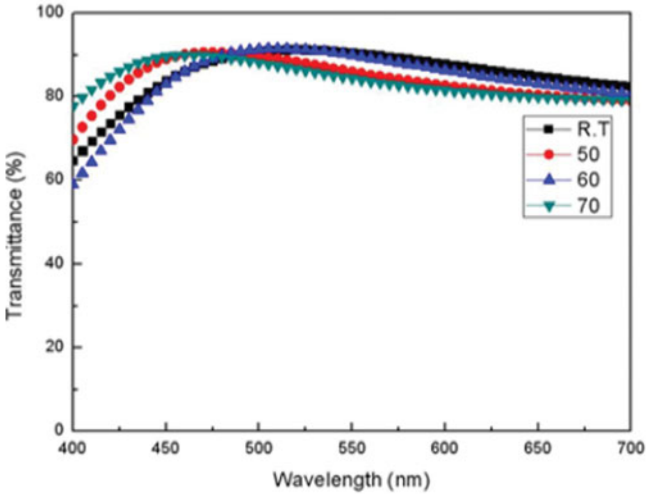

Figure 6. Optical transmittance of the ITO thin films on the SiO_2/cellulose paper substrate deposited at various substrate temperatures.

atomic force microscope (AFM, PSIA XE-100). The optical transmission of the films was examined by UV-Vis spectrophotometer (SHIMADZU UV-1650PC) within visible ranges of 400–700 nm. The electrical characteristics in terms of resistivity, carrier concentration and mobility were investigated by Hall Effect measurement system (ECOPIA HMS-3000) using Van der Pauw test configuration. The current density–voltage and voltage–luminance characteristics of the devices were measured using a computer-controlled Keithley 236 source measure unit and a calibrated fast silicon photodiode (PR650). The devices were measured in air, at room temperature, and without encapsulation.

Results and Discussion

Figure 2 shows X-ray diffraction (XRD) patterns of ITO thin films deposited at substrate temperatures of 27, 50, 60, and 70°C; no diffraction peaks were observed at temperatures below 70°C, which indicates that the ITO layers were amorphous. This is because the particles deposited on the SiO_2/cellulose paper substrate were not charged and therefore had low energies. The ITO thin film deposited at a substrate temperature of 70°C exhibited some artifacts of polycrystalline structure, with weak (222), (400), and (440) peaks; however, as these peaks were weak, the ITO thin films were likely to have been largely amorphous. We can therefore consider 70°C to be the transition temperature between amorphous and polycrystalline deposition using this process.

Figure 3 shows scanning electron microscopy (SEM) images of the surface morphology of the ITO thin films deposited at the different substrate temperatures. The surface morphologies of the ITO thin film depended on the substrate temperature. From Fig. 3(a), the ITO thin films deposited at a substrate temperature of 27°C had a pure amorphous structure. As the substrate temperature increased, small crystallites were formed, and the grain size and density increased with the temperature. Figure 3(d) shows the ITO thin films grown at a substrate temperature of 70°C; grain boundaries were formed between the crystallites, which is consistent with the XRD data. Figure 4 shows atomic force microscopy (AFM) images of the ITO thin films as a function of the substrate temperature. The root

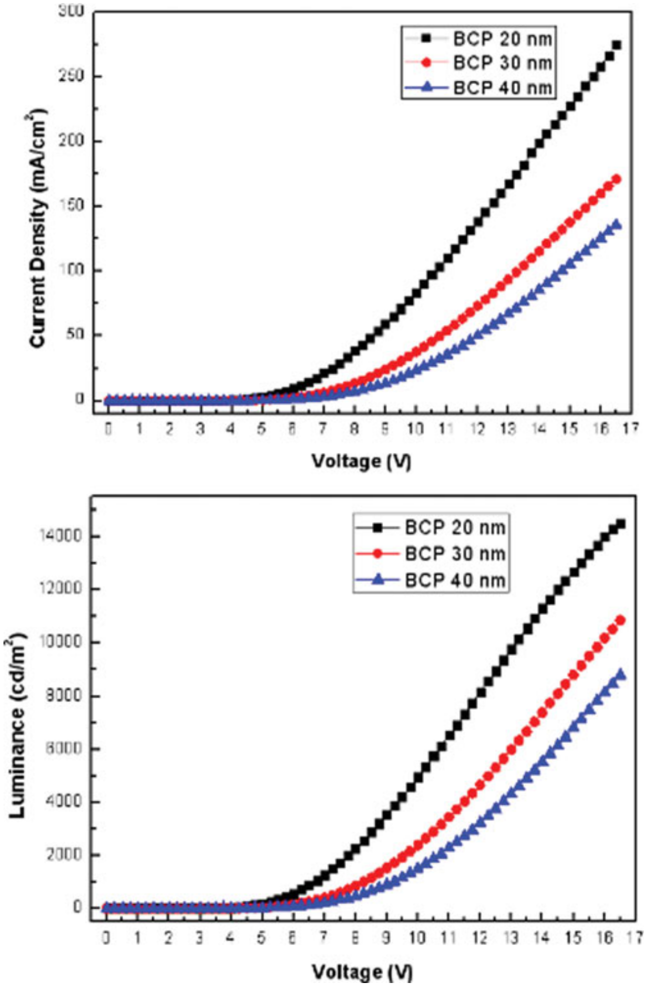

Figure 7. (a) Current density–voltage and (b) luminance–voltage curves of the flexible OLEDs with different BCP layer thicknesses.

mean square (RMS) surface roughness of the thin films fabricated at substrate temperatures of 27, 50, 60, and 70°C were 23.3, 27.6, 23.3, and 27 nm, respectively.

It is well known that the sheet resistance of ITO thin films depends on the substrate temperature during the deposition process, as shown in Fig. 5. As the substrate temperature increased from 27 to 70°C, the sheet resistivity decreased from 1400 to 120 Ω/□, and the charge carrier mobility increased from 13.637 to 37 cm^2/Vs. This may be explained by considering the increase in the size and density of the crystallites with increasing substrate temperature.

The optical transmittances of the ITO thin films were also measured in the wavelength range of 400–700 nm using an ultraviolet (UV)/visible spectrometer. Figure 6 shows the transmittance as a function of wavelength for the substrate temperatures used during deposition. The optical transmittance was in the range of 60–90%. Figure 7 shows the current density–voltage and luminance–voltage curves of the OLEDs deposited at substrate

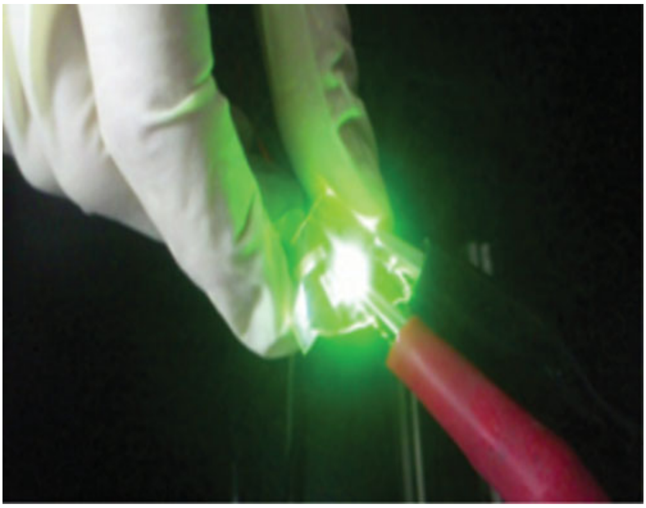

Figure 8. Photograph of an operational OLED with an amorphous ITO anode deposited at a substrate temperature of 70°C.

temperature of 70°C. As the thickness of the BCP layer increased, the operating voltage also increased. At a given applied voltage, the current density and luminance of the OLEDs increased as the thickness decreased. The maximum luminance of the OLEDs was 14,543, 10,863, and 8,759 cd/m^2 for BCP layer thicknesses of 20, 30, and 40 nm, respectively. Figure 8 shows an bright image obtained from the bended OLED deposited at substrate temperature of 70°C operating at an applied bias of 14.75 V.

Conclusions

We fabricated flexible OLEDs on transparent cellulose paper substrates with ITO thin-film anodes deposited at temperatures up to 70°C using FTS. The ITO thin films were amorphous when deposited at substrate temperatures below 70°C. The sheet resistivity of the ITO thin film deposited at 70°C was 120 Ω/□. The current density–voltage curves of the devices exhibited typical diode behavior. The OLEDs with amorphous ITO deposited at substrate temperature of 70°C and a 20-nm-thick BCP layer exhibited a maximum luminance of 14,543 cd/m^2 at an applied voltage of 14.75 V.

Funding

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