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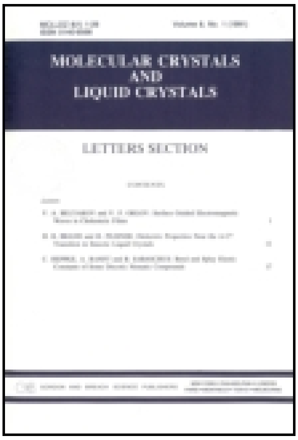

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Flexible Top Emission Organic Light Emitting Diodes with Ni and Au Anodes Deposited on a Cellulose Paper Substrate

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Flexible Top Emission Organic Light Emitting Diodes with Ni and Au Anodes Deposited on a Cellulose Paper Substrate

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Cellulose paper was prepared and utilized as a substrate for flexible top emission organic light-emitting diodes (TE-OLEDs). We successfully fabricated flexible TE-OLEDs with Ni and Au anodes on SiO_2/cellulose paper substrate. The SiO_2 barrier layer coated on the cellulose paper substrate enhanced the surface roughness of the cellulose paper substrate. The surfaces of 100 nm thick Ni and Au films on SiO_2/cellulose paper substrate become continuous on the SiO_2/cellulose paper substrate, resulting in low sheet resistance of the Ni and Au films. While the maximum luminance of the flexible TE-OLED with Au anode was about 3516 cd/m^2 at a driving voltage of 12 V, that of the flexible TE-OLEDs with Ni anode was 1280 cd/m^2 at a driving voltage of 10 V.

Keywords Au anode; cellulose paper; flexible; Ni anode; TE-OLEDs

Introduction

Organic light-emitting diodes (OLEDs) have attracted much attention in the past two decades because they have a fast response, wide viewing angle, thin display and two-dimensional lighting applications [1,2]. On the other hand, top-emission organic light-emitting diodes (TE-OLEDs) have attracted much attention in recent years because of their high aspect ratio active matrix organic light-emitting diodes (AMOLEDs). The AMOLEDs have many advantages, such as high resolution, low power consumption, and large display area. Most of the OLEDs are fabricated on a glass substrate and are then encapsulated with metal or glass lids. The glass substrates are fragile, relatively thick, and heavy. Furthermore, it is difficult to form the OLEDs into arbitrary shapes for applications when they are fabricated using a glass substrate. Recently, flexible OLEDs have received much attention because of their superior advantages such as their thinness, rigidity, and flexibility [3]. Replacing the glass substrate with a flexible substrate can extend the applicability of OLEDs. An OLED display using a flexible substrate, such as plastic or paper, is thinner

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and lighter, compared to one using a glass substrate. Therefore, flexible substrates have a distinct advantage over glass substrates, in many applications [4,5].

Metal oxides such as indium tin oxide (ITO) and indium zinc oxide (IZO) are used as an anode of TE-OLEDs [6,7]. However, metal oxides such as ITO and IZO can be deposited at higher temperatures than 200°C [8]. Therefore, metal anodes such as Ni and Au can be used in TE-OLEDs since they are deposited at room temperature, have a high work function, and have proper reflectance [9,10].

In this paper, we developed TE-OLEDs using Ni and Au anodes deposited on SiO_2/cellulose paper substrate. The cellulose paper was employed since it is made using the most abundant natural polymer on earth. Cellulose is a very suitable material for the purpose owing to the demand for environmentally friendly and biocompatible products [11,12]. We investigated the electrical and structural characteristics of Ni and Au anodes deposited on SiO_2/cellulose paper substrate. The optical and electrical characteristics of TE-OLEDs, using Ni and Au anodes on SiO_2/cellulose paper substrate, were also investigated.

Experimental

Flexible TE-OLEDs were fabricated on cellulose paper substrate. The cellulose solution was made by dissolving cellulose fibers in an acetone solution with mechanical stirring. The cellulose solution was uniformly coated on a glass substrate using a spin coater, and dried, to fabricate cellulose paper. After drying the cellulose solution on the glass substrate, the cellulose paper was detached on the glass substrate by heating the substrate at 100°C for 24 h. A 100 nm thick SiO_2 layer, used as a barrier layer was deposited on the cellulose paper substrate by the radio frequency (RF) magnetron sputtering method. While Ni anode was sputtered on the SiO_2/cellulose paper substrate, Au anode was deposited on the SiO_2/cellulose paper substrate by e-beam evaporator. The thicknesses of the Ni and Au films were 20, 50, and 100 nm. The patterns of the Ni and Au anodes were defined by depositing the Ni and Au film through a shadow mask, onto the SiO_2/cellulose paper substrate. After cleaning the substrate, the anodes namely the Ni and Au films were treated by oxygen plasma at 50 W for 5 min before thermal evaporations of the organic and cathode metal layers were carried out. A 40 nm thick N,N’-bis(naphthalene-1-yl)-N,N′-bis(phentl)-benzidine (NPB) layer was deposited on the Au or Ni coated SiO_2/cellulose paper substrate, followed by the deposition of a 35 nm thick tris-(8-hydroxyquinoline) aluminum (Alq_3) layer that 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). After that, a 30 nm thick 4,7-diphenyl-l, 10-phenanthroline (BPhen) layer was evaporated. Finally, a 15 nm thick Ca and a 10 nm thick Ag metal layers were sequentially evaporated, on the organic layer through a shadow mask. The completed device structures are cellulose paper/SiO_2 (100 nm)/Au (100 nm)/NPB (40 nm)/Alq3:C545T (35 nm,1wt%)/BPhen (30 nm)/Ca (15 nm)/Ag (10 nm), and cellulose paper/SiO_2 (100 nm)/Ni (100 nm)/NPB (40 nm)/Alq3:C545T (35 nm,1wt%)/BPhen (30 nm)/Ca (15 nm)/Ag (10 nm). The deposition rates of metal and organic layers were 0.1 nm/s.

The surface roughness was investigated by atomic force microscope (AFM, PARK SYSTEMS XE-100). The structural characteristics of the SiO_2/cellulose paper with Ni and Au anodes were also investigated by scanning electron microscope (SEM, JEOL:JSM-7401F), and X-ray diffractometer (BRUKER AXE D8 ADVANCE). The sheet resistance of the Ni and Au films were measured by 4-point probe method (Modusystems SR-4-6L).

The transmittance and reflectance were measured using a spectrophotometer (SHIMADZU UV-1650PC), with a normal incidence of monochromatic light at the sample surface side.

The current density-voltage-luminance (J-V-L) characteristics of the devices were measured using computer-controlled Keithley 236 source-measure units and a calibrated fast silicon photodiode (PR650). The devices were measured in air at room temperature without encapsulation.

Results and Discussion

Before depositing the Ni or Au film used as an anode, a 100 nm thick SiO_2 layer was coated on the cellulose paper substrate to enhance the surface roughness. Figure 1 shows atomic force microscopic (AFM) images of the bare cellulose paper substrate, a 100 nm thick SiO_2 layer on the cellulose paper substrate, and a 100 nm thick Ni or Au film deposited on the SiO_2/cellulose paper substrate. As shown in Fig. 1(a), the surface roughness of the bare cellulose paper without SiO_2 layer was about 31 nm. On the other hand, as seen in Fig. 1(b), the surface roughness of the SiO_2 layer on the cellulose paper reduced to about 18 nm. When 100 nm thick Ni film was coated on the SiO_2/cellulose paper substrate by the sputtering method, the surface roughness of the Ni film on the SiO_2/cellulose paper substrate was about 9 nm as shown in Fig. 1(c). On the other hand, the surface roughness of Au film coated on the SiO_2/cellulose paper substrate by vacuum evaporation method was about 4 nm as shown in Fig. 1(d). This indicates that the SiO_2 layer could improve the surface roughness of the cellulose paper substrate. It can be seen from Fig. 1(c) and

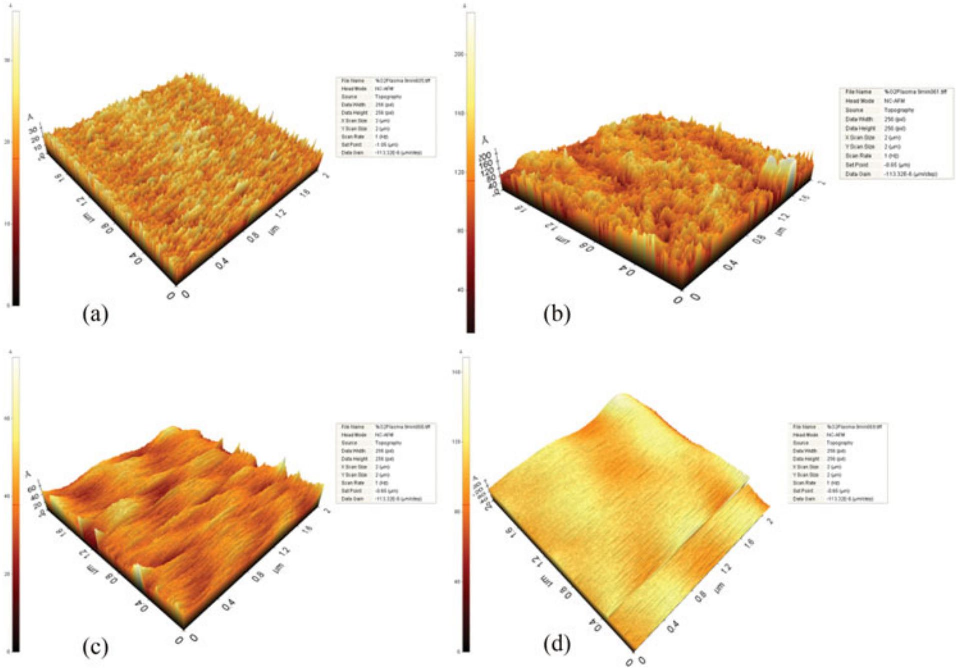

Figure 1. AFM images of Ni or Au films deposited on SiO_2/cellulose paper substrate: (a) paper; (b) paper/SiO_2; (c) paper/SiO_2/Ni; (d) Paper/SiO_2/Au.

Fig. 1(d) that the depositions of anodes such as Ni and Au on a SiO_2/cellulose paper substrate also enhance the surface roughness of the substrate.

Figure 2 shows SEM photographs of the surface morphology of the Ni and Au films with several thicknesses deposited on the SiO_2/cellulose paper substrate. The surface morphologies of the Ni and Au films depend on the thickness of the Ni and Au films, as shown in Fig. 2. In the initial stage of Ni and Au sputtering, the Ni and Au islands are nucleated on the SiO_2/cellulose paper substrate. It can be seen from Fig. 2 that as the thickness of the

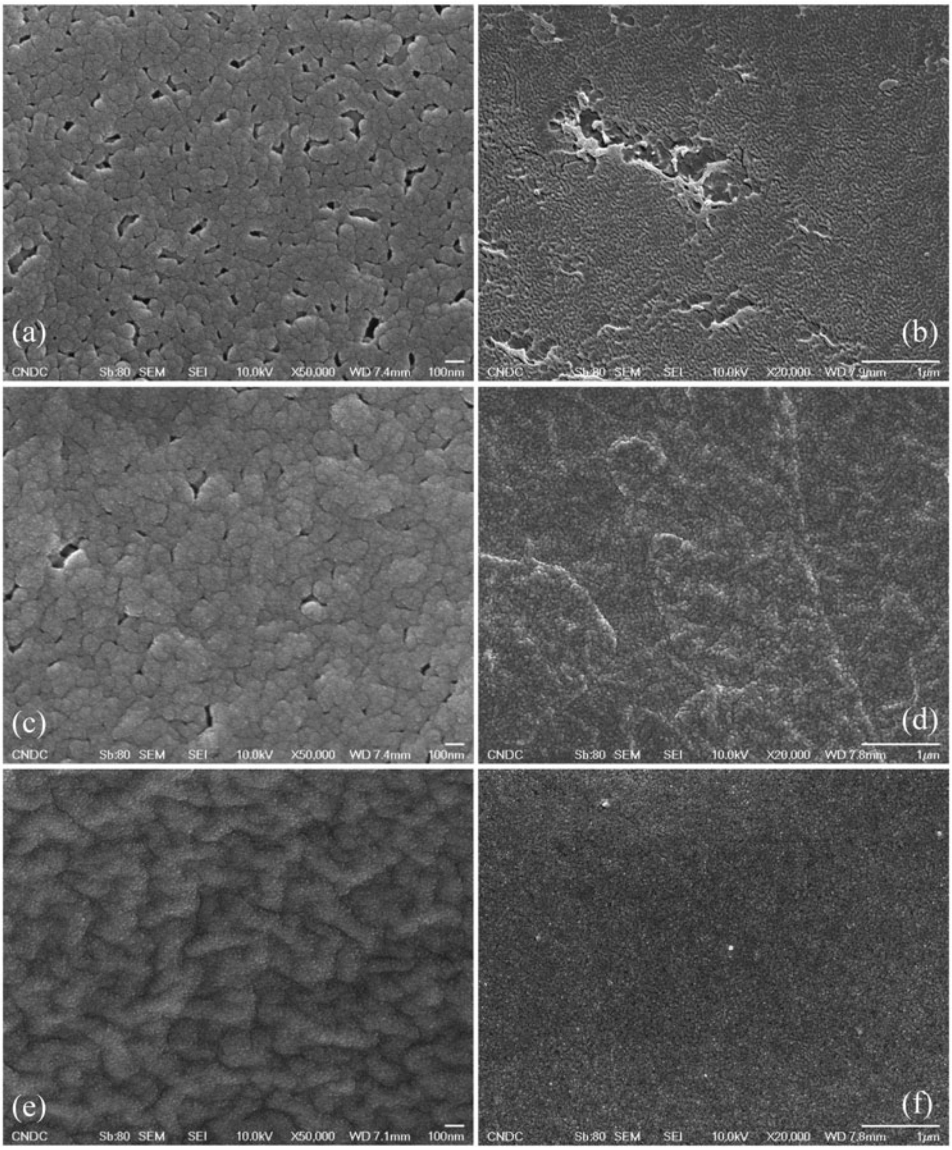

Figure 2. SEM images of Ni or Au films deposited on cellulose paper. (a) 20 nm (b) 50 nm (c) 100 nm (d) 20 nm (e) 50 nm (f) 100 nm.

Table 1. Sheet resistance of Au and Ni films

Thickness of Anode (nm)	20	50	100
Sheet resistance of Ni/SiO_2/cellulose paper (Ω/□)	150∼200	77∼110	27∼30
Sheet resistance of Au/SiO_2/cellulose paper (Ω/□)	20∼30	12∼16	7∼10

anodes increases, islands grow on the SiO_2/cellulose paper substrate, and finally the Ni and Au films become continuous on the SiO_2/cellulose paper substrate.

Figure 3(a) shows X-ray diffraction patterns obtained for the Au film deposited on the SiO_2/cellulose paper substrate. In the case of Au film deposited on the cellulose paper, the X-ray diffraction patterns exhibit crystalline film corresponding to (111) diffraction peak. Fig. 3(b) shows X-ray diffraction patterns obtained for Ni film deposited on the SiO_2/cellulose paper substrate. The X-ray diffraction analysis shows that Ni films with thicknesses of 20, 50, and 100 nm on the cellulose paper exhibit amorphous films.

Table 1 shows sheet resistances of the Ni and Au films with several thicknesses deposited on SiO_2/cellulose paper substrate. As the thicknesses of the Ni and Au films increases, the sheet resistance of the Ni and Au films decreases as expected. This also indicates that the results are consistent with those obtained from Fig. 2. As the Ni and Au films become continuous, the sheet resistance decreases.

The Ca/Ag bilayer was used as a semitransparent cathode for the TE-OLEDs. The light transmittance of the Ca/Ag cathode on the SiO_2/cellulose paper substrate was measured in the wavelength range from 380 to 780 nm by UV/visible spectrometer. Figure 4 shows the light transmittance of the Ca/Ag cathode on the SiO_2/cellulose paper substrate. The light transmittance of the Ca/Ag cathode on the SiO_2/cellulose paper substrate ranged from 47% to 72% in the visible light wavelength range. The sheet resistance of Ca/Ag is about 10 Ω/□ on the glass substrate [13]. However, the sheet resistance of the Ca/Ag bilayer on the SiO_2/cellulose paper substrate was measured to be about 30 Ω/□.

Figure 5 shows the current density-voltage curve of the flexible TE-OLED fabricated on the SiO_2/cellulose paper substrate. The flexible TE-OLED exhibits a typical diode curve. A current density of 1 mA/cm^2 for the flexible TE-OLEDs with Au anode was achieved at a voltage of 5 V. The current density exponentially increases with increasing applied voltage, reaching 321 mA/cm^2 at a voltage of 12.25 V. On the other hand, a current density of 1 mA/cm^2 for the flexible TE-OLED with Ni anode was achieved at a voltage of 4 V. The current density increases exponentially with increasing applied voltage, reaching 91 mA/cm^2 at a voltage of 10V. It is also can be seen from Fig. 5 that the current density does not saturate with increasing voltage. We think that this could be explained by an oxidation of Ni. An insulator formed at the surface by the oxidation of Ni results in higher voltage for current to flow. This causes a degradation of device which destroys the device.

Figure 6 shows the luminance-voltage curve for the flexible TE-OLEDs fabricated on the SiO_2/cellulose paper substrate. The thicknesses of the Ni and Au anodes used in these experiments were 100 nm. While the maximum luminance of the flexible TE-OLED with Au anode was about 3516 cd/m^2, that of the flexible TE-OLEDs with Ni anode was 1280 cd/m^2. The luminance of the flexible TE-OLEDs with Ni anode was lower than that of the flexible TE-OLEDs with Au anode.

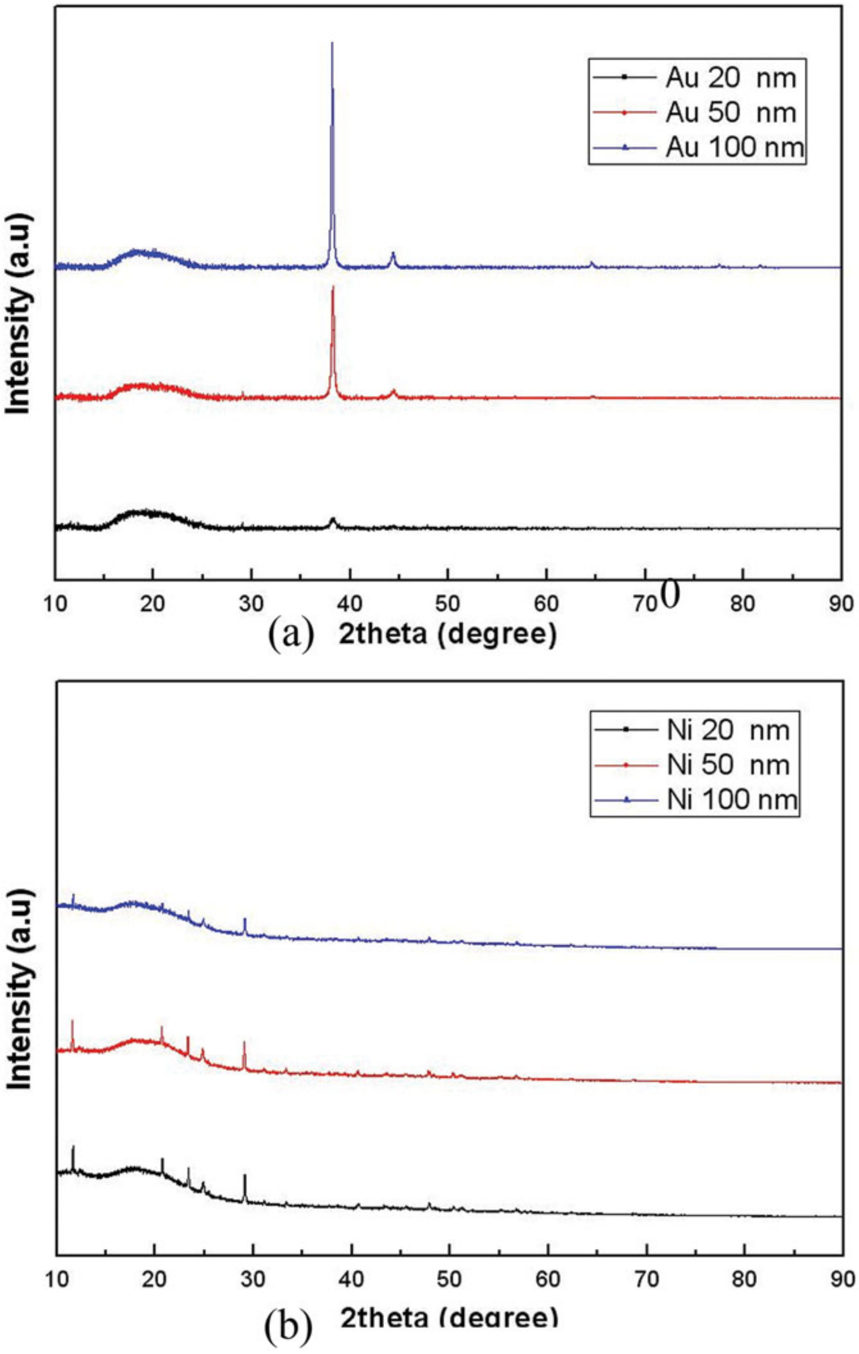

Figure 3. X-ray diffraction patterns of Ni or Au films deposited on cellulose paper. (a) Paper/SiO_2/Ni 20 nm (b) Paper/SiO_2/Au 20 nm (c) Paper/SiO_2/Ni 50 nm (d) Paper/SiO_2/Au 50 nm (e) Paper/SiO_2/Ni 100 nm (f) Paper/SiO_2/Au 100 nm.

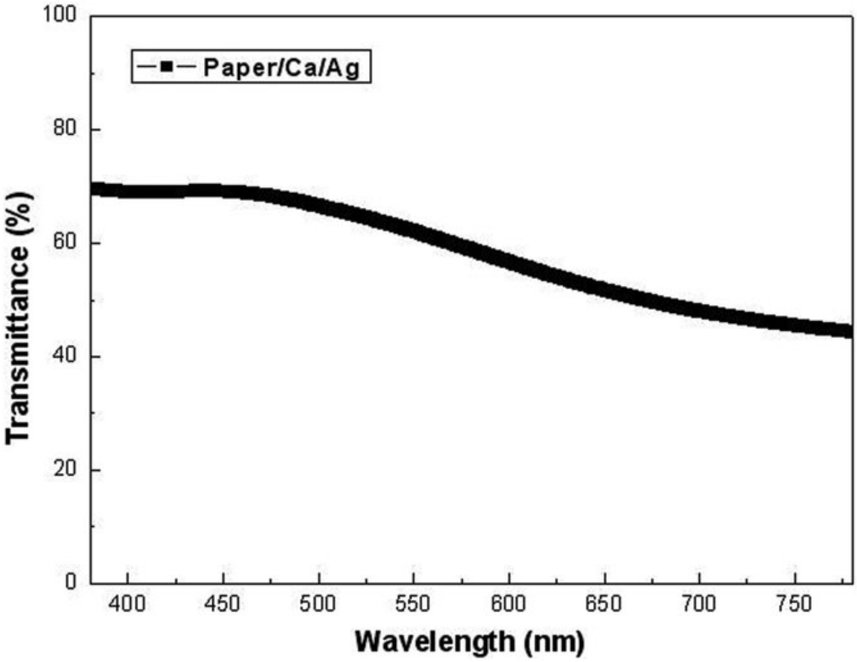

Figure 4. Transmittance of Ag/Ca/cellulose paper substrate in the visible spectrum range.

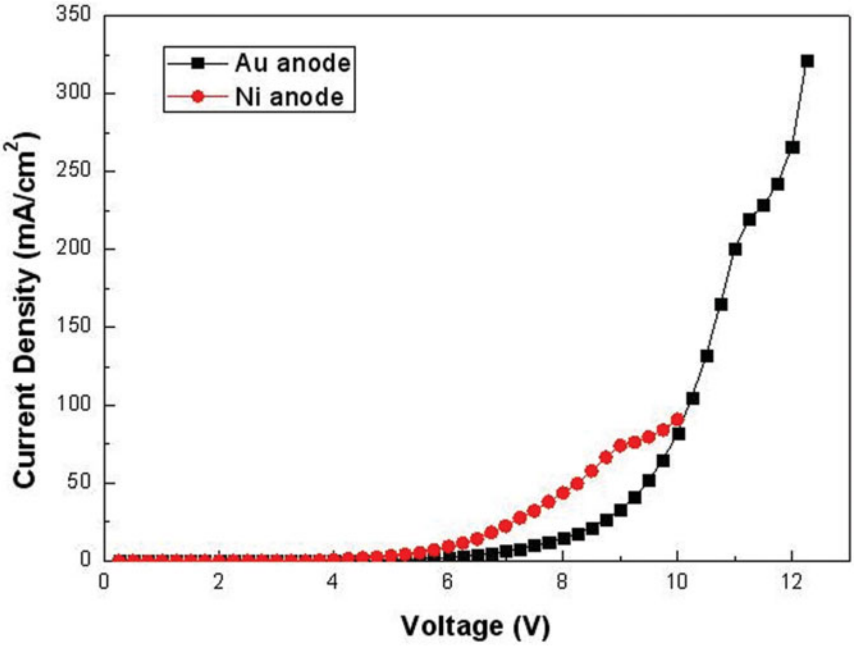

Figure 5. Current density-voltage curve of flexible TE-OLED fabricated on cellulose paper substrate.

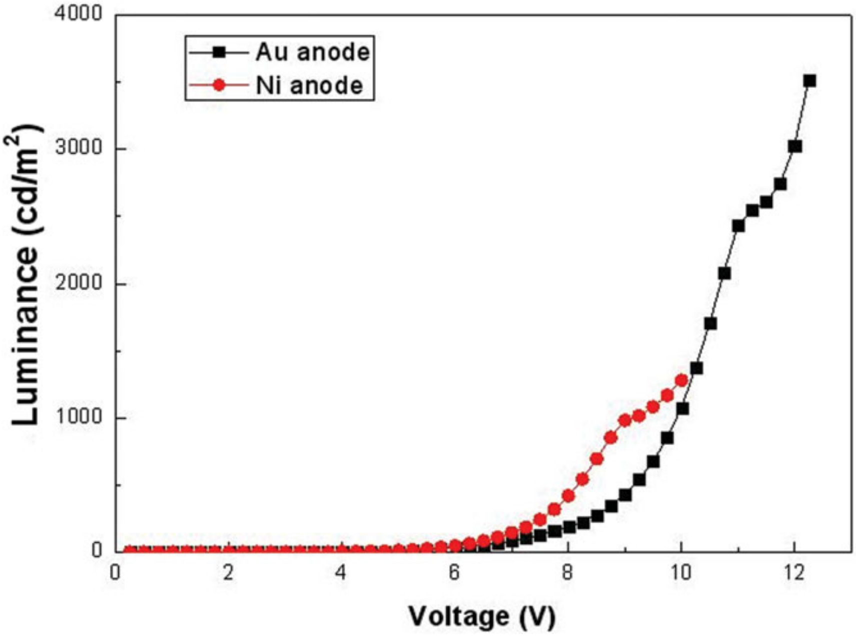

Figure 6. Luminance-voltage curves of flexible TE-OLEDs fabricated on cellulose paper substrate.

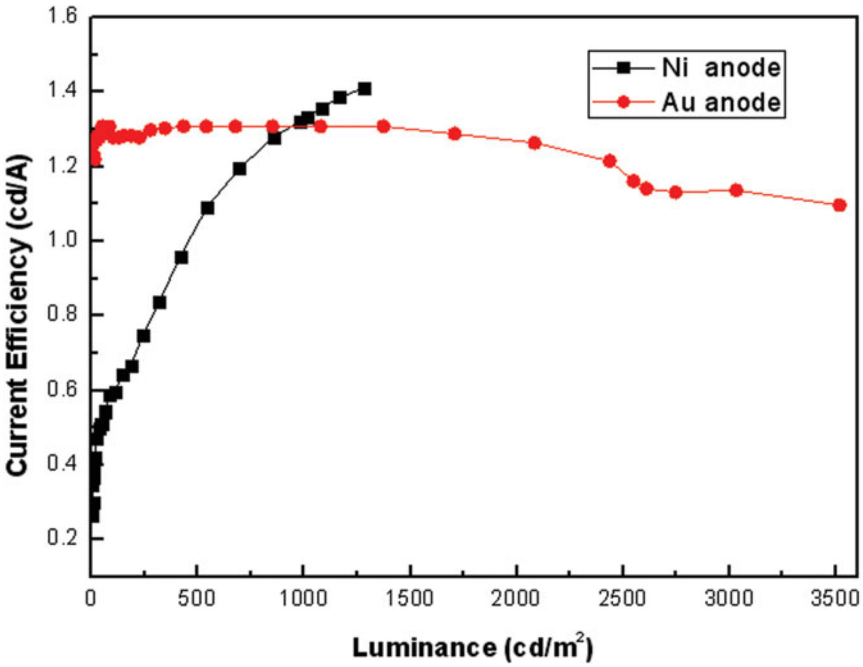

Figure 7. Current efficiency-Luminance curves of flexible TE-OLEDs fabricated on cellulose paper substrate.

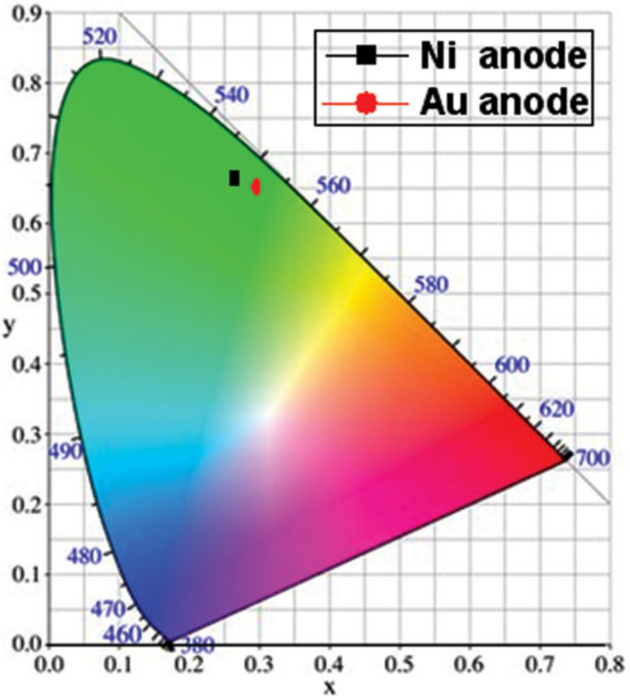

Figure 8. CIE coordinates of flexible TE-OLEDs fabricated on cellulose paper substrate.

Figure 7 shows current efficiency-luminance curves for the flexible TE-OLEDs. The current efficiency of the flexible TE-OLEDs with Au anode reached maximum at 1.3083 cd/A at 1372 cd/m^2 as shown in Fig. 7. The current efficiency of 1.3083 cd/A at 1372 cd/m^2 obtained from the flexible TE-OLEDs with Au anode is much higher than that of 0.085 cd/A at 200 cd/m^2 investigated on bacterial cellulose nanocomposite film similar to this cellulose paper substrate [14]. However, the flexible TE-OLEDs with Ni anode exhibits different characteristic. The current efficiency does not saturate increasing luminance. This can also be explained by the oxidation of Ni anode. The external quantum efficiency obtained from the current efficiency was *** at ** cd/m^2.

Figure 8 shows CIE coordinates of the flexible TE-OLEDs. While the CIE coordinates measured at 10 V for the flexible TE-OLEDs with Ni anode were (0.266,0.656), that measured at 10 V for the flexible TE-OLEDs with Au anode were (0.291,0.655). We observed that the CIE coordinates of the flexible TE-OLEDs with Ni anode were greenshifted compared with those with Au anode.

Conclusions

We successfully prepared flexible TE-OLEDs with Ni and Au anodes on SiO_2/cellulose paper substrate. We investigated the structural characteristics of the SiO_2/cellulose paper substrate. The cellulose paper employed in this study was a very suitable material as a substrate for TE-OLEDs. The surface roughness of the cellulose paper substrate was 31 nm. The SiO_2 layer coating on the cellulose paper substrate enhanced the surface roughness of the cellulose paper substrate. The surfaces of 100 nm thick Ni and Au films on

SiO_2/cellulose paper substrate become continuous on the SiO_2/cellulose paper substrate, resulting in low sheet resistance of the Ni and Au films. The X-ray diffraction analysis showed that while the Ni film on the cellulose paper exhibits amorphous films, the Au film shows crystalline structure. The current density-voltage curve of the devices exhibited a general diode curve. While the maximum luminance of the flexible TE-OLED with Au anode was about 3516 cd/m^2, that of the flexible TE-OLEDs with Ni anode was 1280 cd/m^2. This study shows that cellulose paper could be an environmentally promising substrate for flexible TE-OLEDs.

Acknowledgments

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