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Superior performance of organic light-emitting diodes with microcavity effect

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ABSTRACT

The electro-optics characteristics of organic light-emitting diodes (OLEDs) with microcavity effect were investigated. The transmittance of the substrate with Ag anode was lower than that of the indium-tin-oxide (ITO) anode due to the opaque characteristics of Ag. The current density of the device with Ag anode was high due to the low sheet resistance of Ag anode. The current efficiency of the device with Ag anode was high due to microcavity structure between Ag anode and Al cathode. Especially, the devices of Ag anode show over 43% higher current efficiency than that of conventional ITO anode. The low transmittance of Ag anode using microcavity effect realized as one kind of Fabry–Perot filters was also verified.

1. Introduction

OLEDs present electroluminescence emissions through the recombination of electron–hole pairs. OLEDs have several advantages, such as light-weight, thinness, low driving voltage, and high contrast ratio. Since OLEDs can be used as a flexible display like plastic, it will be an important technology to replace existing displays in future.

The first observation of electroluminescence in organic materials was in a single crystal of anthracene with electrodes and an external kV-order bias in 1965. However, there were no practical applications of the organic electroluminescence owing to extremely high operation voltage (~100 V) [1]. Tang et al. reported a device with organic light-emitting material. This device consists of function-separated organic layers between two electrodes: charge-transport and emission layers. OLED was achievable at a driving voltage below 10 V [2]. ITO substrates play a very important role in anode electrodes of the field of optoelectronic devices such as flat panel display devices. ITO substrates have low electrical resistance and high transmittance in the visible range of the optical spectrum [3]. ITO anode is brittle and the short supply of indium. Therefore, an alternative anode for ITO needs. Van Slyke et al. reported that the performance of OLED can be enhanced by copper phthalocyanine (CuPc) as an inserted carrier-injection layer between an anode and a charge-transport layer [4]. They demonstrated a highly stable OLED. Operating with an initial luminance of 510 cd/m^2, the half-lifetime was about 4000 h and was achieved by using a multilayer device structure with a CuPc-stabilized carrier-injection contact. Material such as Molybdenum oxide (MoOx) was also used as representative carrier-injection layer or charge-transport layer [5–7]. MoOx was an effective material due to its high valence band which is suitable for hole-injection [8]. Hole-injection was enhanced by MoOx and driving voltage of green fluorescent device could be lowered by 1.3 V at 1000 cd/m^2 by using a thin MoOx layer. Kröger et al. reported the effect of MoOx as a charge-transport layer [9]. When MoOx is co-evaporated with 4,4′-Bis(N-carbazolyl)-1,1′-biphenyl (CBP), a significant increase in conductivity is observed, compared to intrinsic CBP thin films. This increase in conductivity is due to electron transfer from the highest occupied molecular orbital of the host molecules to very low lying unfilled states of embedded MoOx clusters. Microlens and microcavity are used to fabricate a high efficiency of OLEDs [10–12]. Zhang et al. investigated high efficiency OLEDs based on a microcavity structure comprised of a dielectric mirror and a metal mirror [12]. The maximum brightness arrived at 37,000 cd/m^2 at a current density of 460.0 mA/cm^2 and the current efficiency at a current density of 0.23 mA/cm^2. The microcavity effect with the proper combination of device structure including the metal electrode can achieve high efficiency. The device of Ag anode with a low sheet resistance as a substitute electrode for ITO can achieve a low driving voltage. Charge carrier-injection and transport layer by co-evaporation between MoOx and N,N′-bis(1-naphthyl)-N,N′-diphenyl-1,1′-diphenyl-1,4′-diamine (α-NPD) can also achieve a low

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driving voltage [7]. We investigated the electro-optics characteristics of the device with Ag anode.

2. Experiment

An ITO substrate for OLEDs was purchased from Geomatic Co. Ultrasonic cleaning processes of ITO substrates are as follows: acetone and semico-clean was used for eliminating organic materials. After the use of semico-clean, pure water and 2-propanol were used to remove semico-clean and moisture, respectively. The glass substrates were also cleaned from washing in pure water, acetone, and 2-propanol with ultrasonication. Then, organic and residual organic solvents were removed by UV ozone treatments using an UV ozone cleaner NL-UV253, Japan Laser Electronics (Present: Filgen (Co.)). After placing the ITO substrates into the chamber, the chamber was purged by O_2 at 0.2 MPa for 45 s. The irradiation of UV light (10 min) removed the organic materials on the substrate surface. After the UV ozone treatment, the chamber was purged by 0.2 MPa N_2 for 1 min to eliminate the remaining ozone. After substrate washing, the substrate was placed into the vacuum deposition chamber immediately in order to prevent contamination of the substrate surface. α-NPD was purchased from Nippon Steel Chemical Co. The measurements of the current–voltage–luminance characteristics were carried out by GUI programming language (LabVIEW, National Instruments) under a pressure of 1×10^{-1} Pa at room temperature. The current was measured with lamp voltage (rising rate: 0.5 V/s) by a source measurement unit (2400 Source Meter, Keithley Instrument, Inc.). The currents and voltages were recorded with a personal computer through a GP–IB interface.

3. Results and discussion

The Ag anode is attempted to replace an ITO anode owing to the insufficient supply and shivery characteristics of indium. Therefore, we used the Ag anode and compared the results with those with an ITO anode. The co-evaporation thin films are deposited with the ratio of α-NPD:MoOx=1:1 simultaneously. The work functions of Ag (20 nm), ITO, Ag (20 nm)/MoOx (10 nm), ITO/MoOx (10 nm), Ag (20 nm)/MoOx:α-NPD (1:1, 10 nm) and ITO/MoOx:α-NPD (1:1, 10 nm) are estimated to be 4.7, 5.0, 5.73, 5.70, 5.34 and 5.36 eV, respectively. The work function of α-NPD was also estimated to be 5.4 eV [10]. The work function of the MoOx layer is higher than those of ITO and the Ag anode by more than 0.7 eV. Although there is no barrier height between MoOx and α-NPD, the injection efficiency is thought to be poor because work function is too deep. The work function of a co-evaporation thin film is higher than those of an ITO and Ag anode by more than 0.34 eV. There is almost no barrier height between the hole-injection material-modified anode and α-NPD. Therefore, the carrier-injection from hole-injection material is higher than that from the anode. It is suggested that the energy matching between MoOx and α-NPD can be improved by the co-evaporation. The sheet resistances of Ag (20 nm) and ITO are estimated to be 2.5 and 6.3 Ω/sq, respectively. Therefore, the current can flow easily owing to the low sheet resistance of the Ag anode.

Table 1 shows the characteristics of the substrate with a Ag or ITO anode. The transmittance of the 150 nm-thick ITO anode in the visible light range is more than 75%. The thin film anode (20 nm) in order to extract light was fabricated due to opaque characteristics of Ag film. Therefore, the 20 nm-thick Ag shows lower transmittance than that of the ITO substrate. Fig. 1 shows Atomic Force Microscope (AFM) images of the hole-injection layer (HIL) on the anode. As shown in Table 1, correlations among the roughness of HILs on the anode do not find. We investigate the current density–voltage characteristics of the devices with Ag anodes or ITO anodes. The device consists of [ITO or Ag (20 nm)]/[MoOx:α-NPD (1:1, 10 nm) or MoOx (10 nm)]/α-NPD (50 nm)/Alq3 (50 nm)/LiF (0.6 nm)/Al (100 nm). Hereafter, the specimen is called by the set of anode and hole-injection layer, as the other parts are common, e.g. Ag/MoOx:α-NPD, ITO/MoOx. The driving voltages at a current density of about 0.1 A/cm^2 for devices Ag/MoOx:α-NPD (1:1, 10 nm), Ag/MoOx (10 nm), ITO/MoOx:α-NPD (1:1, 10 nm), and ITO/MoOx (10 nm) were 6.7, 7.4, 7.7, and 9.4 V, respectively. The current densities of the Ag anode are much higher than those of the ITO anode owing to the low sheet resistance of the Ag anode. The current densities of MoOx:α-NPD are much higher than those of MoOx owing to the MoOx doping concentration between MoOx and α-NPD [9]. The difference between the Fermi level of a MoOx:α-NPD and the HOMO level of a MoOx:α-NPD is reduced by increasing hole densities due to the p-type doping of α-NPD [10]. Therefore, the current density of Ag anode with a MoOx:α-NPD is highest among the used devices owing to the low sheet resistance and p-type doping effect. Fig. 2 shows the luminance–voltage characteristics of the devices with Ag anodes or ITO anodes. The luminance–voltage characteristics show same trends such as current density–voltage characteristics. Fig. 3 shows the luminance–current density characteristics of the device with Ag anodes or ITO anodes. The luminance of the device of the Ag anode with MoOx:α-NPD shows the highest among the used devices owing to the superior front luminance property with the Ag/MoOx:α-NPD using the lowest transmittance. A microcavity is a structure formed by reflecting faces on the two sides of a spacer layer or optical medium, so there is very low this effect between ITO (transparent) and Al (reflector). The microcavity effect shows the spectral narrowing of the emission band and enhanced output [7]. The current efficiency of the device of the Ag anode can be improved due to the microcavity effect of the interference phenomena induced by internal reflection of light between the semitransparent Ag anode and the Al cathode. The device of the Ag/MoOx:α-NPD shows about 43% higher current efficiency than that of the ITO/MoOx. It is considered that the device can abstract strong lights due to the microcavity effect between the Al and Ag/MoOx with the low transmittance by the 20 nm-thick Ag using an opaque characteristic. The device with the Ag/MoOx:α-NPD shows about 73% higher current efficiency than that with the ITO/MoOx:α-NPD. Tao et al. reported a large amount of internal reflection at the interface owing to the large refractive indices mismatches [13]. The current efficiency of the ITO/MoOx:α-NPD is slightly higher than that of the ITO/MoOx owing to the optical characteristics between the Al and ITO/MoOx:α-NPD with the high mismatch possibility of refractive indices at the interface between air and the ITO/MoOx:α-NPD of low transmittance

Table 1
Characteristics of the substrate with a Ag or ITO anode.

Substrate	Transmittance (%) at peak wavelength	FWHM (nm)	Peak wavelength (nm)	Rp (nm)	Rv (nm)	Ra (nm)
ITO/MoOx	87	115	532.5	17.5	−11.8	1.81
ITO/MoOx: α-NPD	83	106	520	15.4	−12.6	2.18
Ag/MoOx	49	80	547	18.1	−10.8	2.57
Ag/MoOx: α-NPD	41	85	566	15.0	−11.0	2.27

Rp—maximum peak height, Rv—maximum valley depth, and Ra—average roughness.