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Improved Performance of Top-Emission Organic Light Emitting Device With a Mixed Single Layer

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Top-emission organic light emitting devices (TEOLED) with a mixed single layer by mixing of electron transport materials, hole transport material and dope material were investigated. With an optimized mixing ratio of PyPySPyPy:Alq_3:α-NPD:Rubrene [25:50:25:1], maximum power efficiency, 3.45 lm/W, and maximum luminance, 7,000 cd/m^2 was obtained. Compared to the conventional heterostructure device, the performance was improved with a factor of more than two. The dependence of organic layer thickness on device performance was also investigated and the result showed that 100 nm was an appropriate thickness for good performance of mixed single layer TEOLED. In addition, the maximum luminance and power efficiency were obtained at 20,000 cd/m^2 and 7.5 lm/W, respectively, when TPD layer of 45 nm was capped onto top metal electrode.

Keywords Capping layer; mixed single layer; top-emitting organic light emitting device (TEOLED)

1. Introduction

Advances in technologies of organic light-emitting devices (OLEDs) in recent years have imposed substantial demands in efficient top-emitting OLEDs (TEOLED) [1–3]. TEOLEDs render feasible fabrication of OLED displays on opaque substrates, such as, silicon wafers and metallic foils. Another aspect of TEOLEDs compared to conventional bottom-emitting OLEDs (BEOLEDs) is their higher light outcoupling efficiency because of light confinement due to substrate waveguide mode is eliminated.

However, TEOLEDs show quite strong microcavity effects because of the reflective bottom electrode and the partially reflective top electrode used, as well as, the overall device thickness being in the order of the emission wavelength. Consequently, the performance of a TEOLED depends critically on the optical properties of the electrodes, a composition of organic materials, the thickness of the organic layers, and position of the emission zone. For display and lighting applications, the lifetime of the devices is another important issue, which depends not only on the stability of

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the composition of organic materials [4], but also on the combination at the metal/organic and organic/organic interfaces [5]. Moreover, an understanding of mechanism in the OLED degradation under operation is still issue. Especially for the TEOLED, the operational stability has remained largely unexplored and faced large challenge for practical application.

In our previous work, OLEDs using a mixed single layer was proposed and identical characteristics compared to the stacked heterojunction device could be obtained by optimizing the weight ratios of hole and electron transport materials [6]. By using the mixed single layer, a discrete heterojunction interface could be eliminated. Compared with the multilayer structure, the lifetime of the device operation was improved and the suppression of dark spots was confirmed by evaluating the dark spots growth before and after measurement of half lifetime [7]. In this study, we have investigated TEOLEDs with a mixed single layer by mixing of electron transport materials tris-(8-hydroxy-quinoline) aluminum (Alq_3) and 2,5-bis(6′-(2′,2″-bipyridyl))-1,1-dimethyl-3,4-diphenylsilole (PyPySPyPy), hole transport material 4,4′-bis[N-(1-napthyl)-N-phenyl-amion] biphenyl (α-NPD) and dopant material 5, 6,11,12-tetraphenylnaphthacene (rubrene). The mixing ratio of all organic materials is optimized for obtaining the best characteristics. Capping layer on top electrode is used for improving light outcoupling.

2. Experimental

Aluminum-neodymium alloy (AlNd, Al: Nd = 98: 2, Kobelco Research Institute) is selected as cathode in order to flatten the electrode surface and avoid the shor-circuiting [8]. Gold is used as a semitransparent anode. As shown in Figure 1, mixed single-layer device has a structure of AlNd/PyPySPyPy + Alq_3 + α-NPD + rubrene (100 nm)/MoO_3 (50 nm)/Au (20 nm). Device characteristics are compared to the heterostructure device of AlNd/PyPySPyPy (20 nm)/Alq_3 + rubrene (30 nm)/α-NPD (50 nm)/MoO_3 (50 nm)/Au (20 nm). Glass substrate used is alkaline earth boro-aluminosilicate glass (Corning 1737). Mixed organic materials are evaporated at a pressure of about 4×10^{-6} Torr at a rate of $1 \sim 3$ Å/s. The AlNd cathode and gold anode are evaporated using a metal mask. The device area is $2 \times 2\,mm^2$.

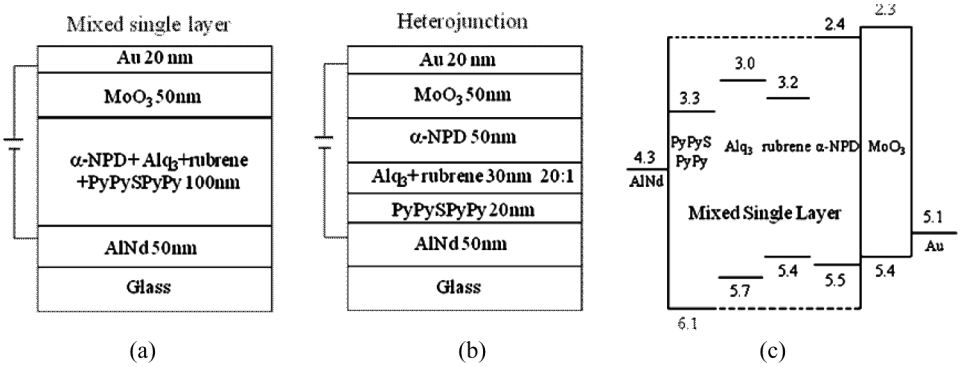

Figure 1. Device structure of inverted (a) mixed single layer TEOLED and (b) heterojunction TEOLED. (c) Energy diagram of the mixed single layer TEOLED.

Devices are encapsulated with a counter glass substrate filled with molecular sieve in a dry nitrogen glove box. Device characteristics are measured using semiconductor parameter analyzer (HP 4155B) and luminance meter (Topcon BM-3).

3. Results and Discussion

For mixed single layer OLED, the device characteristics were strongly depended on the mixing ratio of organic materials [9–11]. Especially in TEOLEDs, electron and hole injection electrodes were placed at bottom and top, respectively, which was reverse position to the bottom-emitting OLEDs. Charge transport, carrier balance and carrier recombination conditions for the mixed single layer TEOLED will be different from those for bottom-emitting OLED and the mixing ratios of organic materials should be optimized.

With optimized mixing ratio of PyPySPyPy: Alq_3: α-NPD: rubrene = 25: 50: 25: 1, radiative recombination probabilities of holes and electrons were increased and the maximum luminance and the maximum efficiency were obtained at 7,000 cd/m^2 and 3.45 lm/W, respectively.

3.1. *Comparison With Heterojunction Device*

Figures 2 (a)-(c) shows current density vs voltage (*J-V*), luminance vs current density (*L-J*) and power efficiency vs current density voltage (η-*V*) characteristics for mixed single layer TEOLED and heterostructure TEOLED, respectively. From Figure 2 (a), we can see that the operational voltage of the mixed single layer device at higher current densities was higher than that of, i.e., 17.0 V for mixed single layer device and 12.8 V for heterojunction device at 100 mA/cm^2. One of possible reason of increment of operational voltage in the mixed single-layer device was larger hopping distance for hole and electron transport with respect to heterojunction device [12]. On the other hand, as seen from Figure 2 (b) and (c), the mixed single layer device shows better performance than that of heterojunction device. For example, by comparing with heterojunction device with a maximum power efficiency of 0.92 lm/W and maximum luminance of 2,970 cd/m^2, the mixed single layer device showed superior device characteristics by a factor of more than two.

It was noticed that the power efficiency in TEOLEDs was decreased markedly with increased current density, i.e., from 3.45 lm/W at 0.03 mA/cm^2 to 0.20 lm/W at 460 mA/cm^2. This may be related to the quenching of singlet-excited state of the rubrene dopant. For verifying the effects resulted by rubrene, we also fabricated a mixed single layer TEOLED with same structure just didn't dope rubrene in the mixed single layer. The characteristics were shown in Figure 3. As we can see, although the power efficiency of device without rubrene dopant is lower than that of device with 1.0 wt% rubrene dopant when current density is less than 20 mA/cm^2, the power efficiency shows a slowly decreasing trend in this range of current density, from 0.78 lm/W to 0.50 lm/W. With further increase of current density, the decrease of power efficiency is accelerated. This may be attributed to exciton dissociation resulted by Joule-heat generated at larger current density. In fact, although the use of fluorescent dopants in the emission layer generally produces a significant increase in electroluminescence efficiency, the magnitude of this increase is often attenuated at high brightness, i.e., when the OLED is driven at a high current density [13,14].

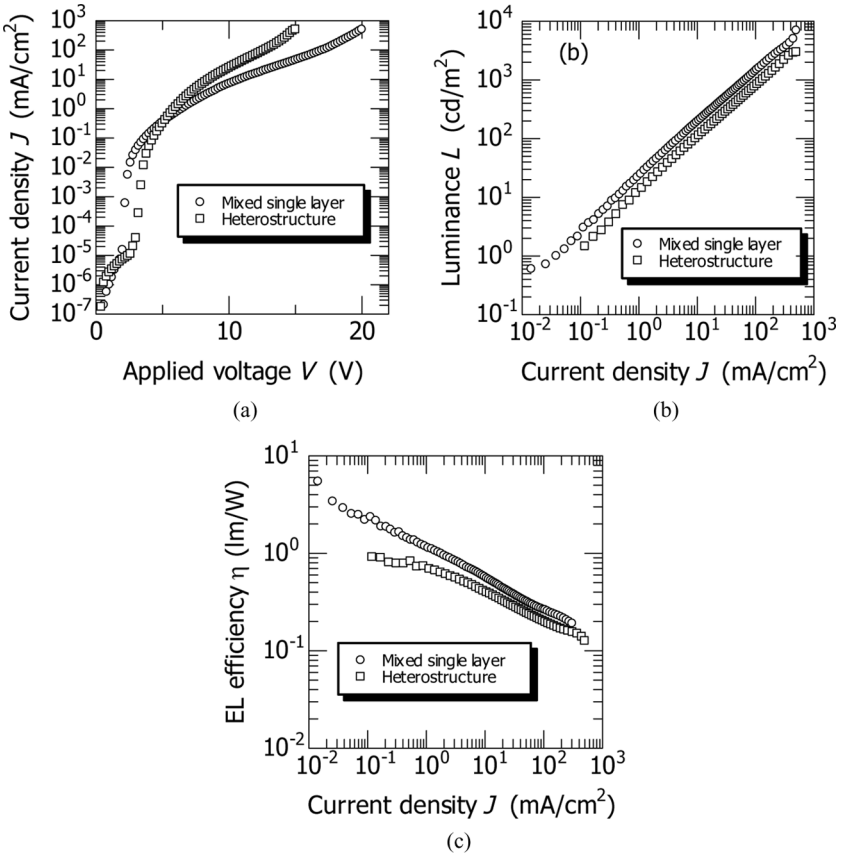

Figure 2. (a) Current density vs voltage (*J-V*), (b) Luminance vs current density (*L-J*) and (c) power efficiency vs current density (η-*J*) characteristics of mixed single layer TEOLED and heterostructure TEOLED.

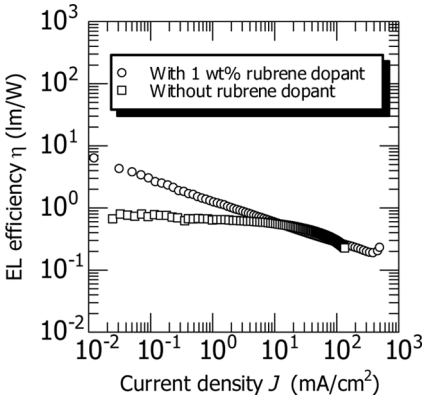

Figure 3. Power efficiency vs current density (η-*J*) characteristics of mixed single layer TEOLED with and without rubrene dopant.

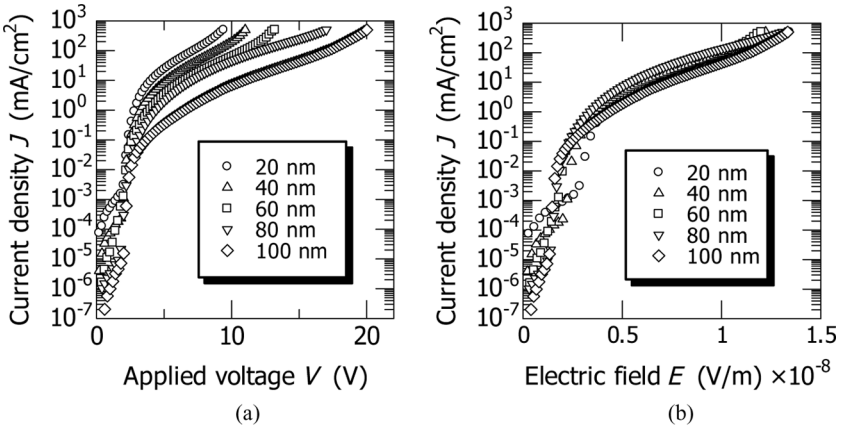

Figure 4. The effects of organic layer thickness on (a) current density vs driving voltage (*J-V*) and (b) current density vs electric field (*J-E*).

3.2. *Thickness Dependence of Organic Layer*

The mixed single layer structure makes it possible to lower the driving voltage of device by decreasing thickness of mixed single organic layer. Figure 4 (a) and (b) shows the effects of organic layer thickness on current density vs driving voltage (*J-V*) and current density vs electric field (*J-E*), respectively. The driving voltage of mixed single layer TEOLED at 100 mA/cm^2 decreases with the thickness of the mixed single layer, ranging from 17.0 V for 100 nm to 7.0 V for 20 nm. Reducing thickness lowers the device driving voltage. However, from Figure 4 (b), we can see that *J-E* curves upon different thickness of organic layer are all identical. It clearly demonstrates that the *I-V* characteristics depend, not on the voltage, but instead on the electric field strength. This may be implied a tunneling mode for carrier injection through a barrier at the interface of electrode/mixed single organic layer.

Although the driving voltage can be lowered by reducing thickness of organic layer, the electroluminescence (EL) intensity and power efficiency of device was deteriorated due to microcavity effects. Table 1 lists the comparison of device characteristics in different organic layer thickness. In addition, there is a huge effect of organic layer thickness on the emission spectrum. Figure 5 shows the different emission spectra for the corresponding organic layer thickness. At thickness of

Table 1. The comparison of device characteristics of mixed single layer TEOLEDs for the corresponding organic layer thickness

Sample thickness	Voltage (V) @ 100 mA/cm^2	L (cd/m^2) @ 100 mA/cm^2	L (cd/m^2) Max	η (lm/W) @ 100 mA/cm^2	η (lm/W) Max
20 nm	7.0	250	1,400	0.12	0.65
40 nm	8.9	640	2,480	0.22	1.62
60 nm	11.1	920	4,070	0.25	2.10
80 nm	12.2	1,180	5,060	0.31	2.32
100 nm	17.0	1,500	7,000	0.27	3.45

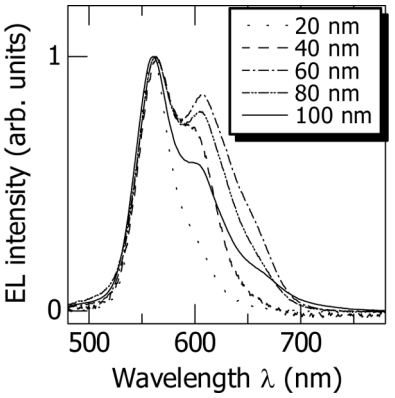

Figure 5. The EL spectra of mixed single layer TEOLEDs for the corresponding organic layer thickness.

20 nm, the EL peak position is at about 560 nm. With increasing of thickness, another peak located about 610 nm is also appeared and it reaches maximum at the thickness of about 60 nm. Further increase the thickness of organic layer results in the disappearance of the dual peaks into the original peak.

3.3. *TPD Capping Layer*

In the TEOLEDs, the light outcoupling was limited by the poor transparence of the metal film while it can be enhanced by depositing a light outcoupling layer onto the top metal electrode [15–17]. Here, we investigated the effects of TPD capping layer (45 nm, refractive index n = 1.85) on performance of mixed single layer TEOLEDs with an optimized mixing ratio PyPySPyPy: Alq_3: α-NPD: rubrene = 25: 50: 25: 1. The thicknesses of capping layer were optimized and referred from Ref. 18.

Figure 6 shows the *η-J* characteristics of mixed single layer TEOLED capped with and without TPD thin films. Table 2 lists the comparison of TEOLEDs characteristics with and without capping layer. Obviously, the introduction of capping

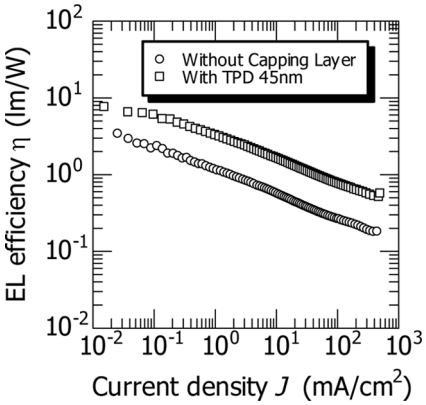

Figure 6. Power efficiency vs current density (*η-J*) of mixed single layer TEOLED with and without TPD capping layer.

Table 2. Electrical and optical characteristics of mixed single layer TEOLEDs with and without TPD capping layer

Sample	L (cd/m^2) @ 100 mA/cm^2	L (cd/m^2) Max	η (lm/W) @ 100mA/cm^2	η (lm/W) Max
Without capping layer	1,500	7,000	0.27	3.45
With TPD (45 nm)	3,700	20,000	0.78	7.52

layer improved luminance and efficiency of the device. The maximum luminance and power efficiency reached 20,000 cd/m^2 and 7.52 lm/W, respectively, when a 45 nm TPD capping layer was capped onto Au anode. Compared to TEOLED without capping layer, the light outcoupling efficiency was enhanced with a factor of more than two. Upon optimized thickness of capping layer, the transmittivity of top electrode can be improved, resulting in enhancement of light outcouping. And the reflectivity of top electrode also can be lowered, in a certain degree, weakens the microcavity effect of TEOLEDs. To better understand the performance enhancement, further study of angular dependence of emission intensity and spectral characteristics on the capping layer thickness is necessary.

4. Conclusions

In summary, we have investigated TEOLEDs with a mixed single layer by mixing of electron transport materials, hole transport material and dopant material. The mixed single layer TEOED has a better performance compared to heterostructure TEOLEDs. The dependence of organic layer thickness is also investigated and the result shows that 100 nm is an appropriate thickness for good performance of mixed single layer TEOLED. In addition, the luminance and power efficiency of mixed single layer TEOLED were enhanced markedly when a 45 nm TPD layer was capped onto top metal electrode.

Acknowledgments

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