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High-efficiency stacked white organic light-emitting diodes

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We report efficient tandem white organic light-emitting diodes (WOLEDs) by using bathocuproine: Cs_2CO_3/MoO_3 as an effective interconnecting layer. We utilized two primary colors of sky blue and orange fluorescent emitters to obtain efficient white electroluminescence. Although single WOLEDs using two adjacent emitting layers showed a maximum current efficiency of 7.96 cd/A with Commission Internationale d'Eclairage (CIE) coordinates of (0.28, 0.34), the tandem WOLED device made by stacking two single color OLEDs in series demonstrated doubled maximum current efficiency of 17.14 cd/A with CIE coordinates of (0.28, 0.41). The stacking of different single color OLEDs in series instead of double stacking of WOLEDs can be useful to achieve highly efficient WOLEDs because it can reduce the number of layers of the devices. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2837419]

Organic light-emitting diodes (OLEDs) have attracted intensive and increasing attention since they are stong candidates for next generation displays and solid-state lighting sources.[1,2] In particular, white OLEDs (WOLEDs) with high efficiency, low operating voltage, and long lifetime have also attracted a great deal of attention due to their applications to large-area full color displays coupled with color filters, mobile displays with high resolution, and lighting sources for both general illumination and liquid crystal display backlights, which require maskless fabrication processes.

White emission from OLEDs can be achieved by mixing three primary colors (red, green, and blue)[3–6] or two complementary colors (e.g., sky blue and orange)[7–14] from different light-emitting small molecules or polymers. Generally, WOLEDs based on small molecules have been fabricated in a multilayer structure with two or more emitting layers[3–11] or with a single emitting layer of the active host doped with fluorescent or phosphorescent dyes.[12–14]

Since it is desirable to obtain high brightness and high efficiency at low current density in OLED displays and lighting sources, vertically stacked (or call as "tandem") OLEDs composed of multiple electroluminescent (EL) units connected in series have been introduced.[7,8,15–17] One of the major challenges in tandem OLEDs is to prepare the effective charge-generating interconnection unit between EL units. The unit is basically formed by coupling of a *n*-type layer (or electron injecting conductive layer) and a *p*-type layer (or hole injecting conductive layer). The *p*-type layers used so far include *p*-doped organic hole-transporting materials using tetrafluorotetracyano-quinodimethane (F_4-TCNQ) (Ref. 18) or WO_3 (Ref. 19) and inorganic materials such as indium tin oxide (ITO),[20] V_2O_5,[20] WO_3,[8,19] and MoO_3.[7] The *n*-type layer is in general metal-doped organic electron-transporting layers using low work function metals such as Cs,[18,19] Li,[6,7] and Mg (Refs. 8 and 17) or metal carbonates (Ref. 16). The good combination of the *n*-type and *p*-type layers can provide an effective interconnection unit in tandem OLEDs. In turn, the tandem OLED structure is useful to obtain high luminance and efficiency in WOLEDs.

Here, we report a tandem white OLED using Cs_2CO_3-doped bathocuproine (BCP)/MoO_3 as an effective interconnecting layer to connect the individual EL units. Optically transparent MoO_3 and Cs_2CO_3 are more cost effective and easier to handle by thermal evaporation compared with contaminating organic dopant F_4-TCNQ for *p*-type layer and highly reactive metals for *n*-type layers. The white EL in tandem OLEDs reported here was obtained by using two primary colors of sky blue and orange fluorescent emissions, which were respectively generated from each EL unit in series. It is demonstrated that the tandem white OLED device fabricated by stacking of two different single color OLEDs (we call *asymmetric tandem*) in series instead of double stacking of WOLEDs (we call *symmetric tandem*) showed a very high maximum current efficiency of 17.14 cd/A with Commission Internationale d'Eclairage (CIE) coordinates of (0.28, 0.41).

We fabricated blue and white OLED devices, which have the same device structures except for the emitting layers. Figure 1 shows the chemical structures of the materials we have utilized in this work. The 20 nm thick hole injection layer of (*N*,*N*′-[*p*-di(*m*-tolyl)aminophenyl]-*N*,*N*′-diphenyl bendizine) (DNTPD) was formed by spin coating on a precleaned ITO substrate with a sheet resistance of 15 Ω/sq. Then, the 30 nm thick hole transporting layer of *N*,*N*′-bis-(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine was vacuum deposited on the DNTPD layer. As for the single stack white emitting device, the device comprises two emission layers. The first layer emits blue light by doping 5 wt % 4,4′-bis[2-{4-(*N*,*N*-diphenylamino)phenyl}vinyl] biphenyl (DPAVBi) in 2-(*t*-butyl)-9,10-bis (2′-naphthyl)anthracene (TBADN). The second one emits orange light obtained by doping 10 wt % rubrene in TBADN. Then, the emitting layer was capped by an electron transporting layer consisting of a 30 nm thick

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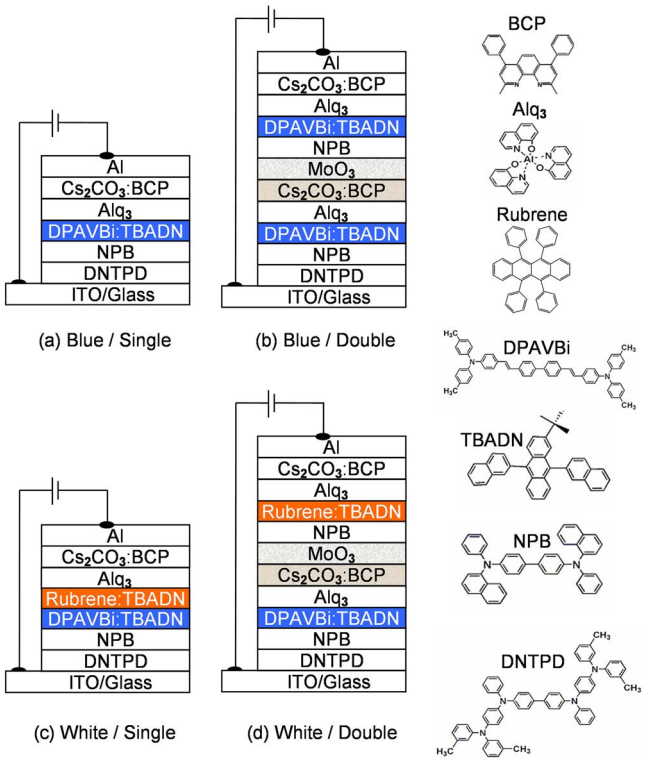

FIG. 1. (Color online) Molecular structures of the organic materials used for fabrication of blue and white organic light-emitting diodes and the device structures of single-unit (standard) and double-unit (stacked) OLED devices for blue and white emissions.

tris(8-hydroxyquinoline) aluminum (Alq_3) layer, followed by a second 20 nm thick BCP layer mixed with Cs_2CO_3 in 1:1 molar ratio. As for the double stack white emitting device, we separate each color EL element by a thermally deposited, 20 nm thick MoO_3 charge-generation layer, as Fig. 1 shows. Finally, a 150 nm thick Al cathode layer was deposited on the Cs_2CO_3: BCP layer using a metal shadow mask to define an emitting area of 4.0 mm^2. After the cathode formation, the devices were encapsulated using UV-epoxy resin and a glass lid with desiccant under dry nitrogen atmosphere (<1.0 ppm H_2O and O_2). Luminance measurement was carried out using a Topcon BM-8 luminance meter and EL spectra were taken on a Hamamatsu Photonics PMA-11 optical multichannel analyzer with an XYZ stage at room temperature.

We tried to confirm that the charge generating interconnection unit of Cs_2CO_3:BCP/MoO_3 has good interconnection capability by fabricating a double stacked single-color (i.e., blue) OLED. Figure 2 shows the current-voltage-luminance characteristics of the single-unit (standard) and double-unit (stacked) blue OLEDs using 5 wt % DPAVBi-doped TBADN as the emitting layer whose device structures are shown in Figs. 1(a) and 1(b). Judging from the current density versus voltage and luminance versus voltage characteristics [Figs. 2(a) and 2(b)], the operating voltages of the stacked device were almost doubled. Figure 2(c) shows that at a current density of 20 mA/cm^2, the double stacked device gives a current efficiency of 16.5 cd/A while the single-unit device gives 9.2 cd/A. This indicates that the present interconnection unit is highly effective in the stacked devices. The single-unit blue OLED showed the maximum current efficiency of 9.3 cd/A with CIE x,y color coordinates of (0.16, 0.24). The maximum current efficiency of the double-unit stacked blue OLED was almost doubled (17.1 cd/A)

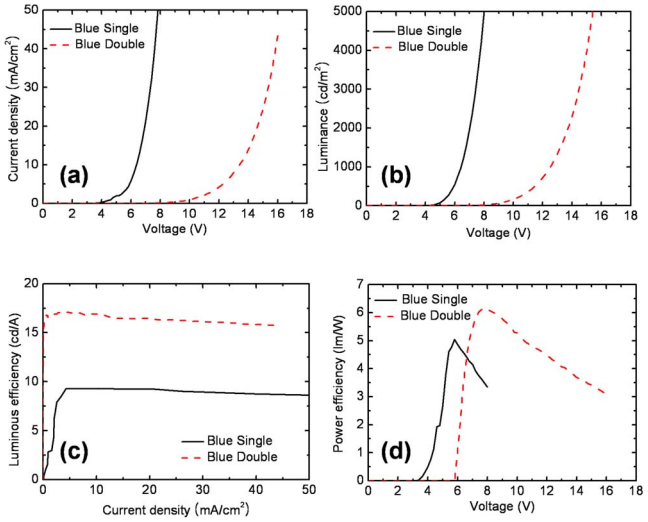

FIG. 2. (Color online) Current-voltage-luminance characteristics of single-unit (standard) and double-unit (stacked) blue OLED devices.

with CIE x,y color coordinates of (0.16, 0.29). The double-unit stacked blue OLED showed better maximum power efficiency (6.15 lm/W at 12 cd/m^2 and 4.64 lm/W at 500 cd/m^2) than the single-unit blue OLED (5.05 lm/W at 402 cd/m^2 and 4.92 lm/W at 500 cd/m^2). From all these data, we concluded that the interconnection unit of Cs_2CO_3:BCP/MoO_3 allows efficient opposite hole and electron injection into two adjacent emitting units to give efficient tandem devices. The effectiveness of present interconnection unit can be ascribed to the electric-field-assisted bipolar charge spouting in Cs_2CO_3:BCP layer, as proposed in the case of Mg:Alq_3/V_2O_5.[21]

We employed the interconnection unit to fabricate a double-unit stacked WOLED while keeping the device structure and the layer thickness the same. First of all, we fabricated a WOLED based on a single-unit standard structure using adjacent two emitting layers composed of 10 nm thick DPAVBi:TBADN and 10 nm thick rubrene:TBADN [the device structure is shown in Fig. 1(c)]. Here, we utilized TBADN as a common host for emitting dopants. Then, we fabricated a tandem WOLED based on a double-unit structure of two different single color EL units, where the EL unit using DPAVBI:TBADN emitting layer is located at the bottom and the other one using rubrene:TBADN emitting layer is located at the top [see Fig. 1(d)]. As Figs. 3(a) and 3(b) show, the operating voltages of the double-unit stacked WOLED were almost doubled, which implies that the interconnection unit also works effectively in the double-unit stacked device. The maximum current efficiency of a single-unit device was 7.96 cd/A with CIE x,y color coordinates of (0.28, 0.34). The double-unit stacked WOLED device showed nearly doubled maximum current efficiency of 17.14 cd/A with CIE x,y color coordinates of (0.28, 0.41). The maximum power efficiencies of single-unit (standard) and double-unit (stacked) WOLED were 5.95 lm/W at 7.3 cd/m^2 (3.04 lm/W at 500 cd/m^2) and 6.49 lm/W at 179 cd/m^2 (4.75 lm/W at 500 cd/m^2), respectively. Our result showed that the stacked device composed of two single color EL units (i.e., each EL element generates only one single color emission) can be effectively used to achieve a highly efficient white emission. This approach is practically useful as it can reduce the number of layers of the tandem WOLED devices. In addition, by using the approach, we can

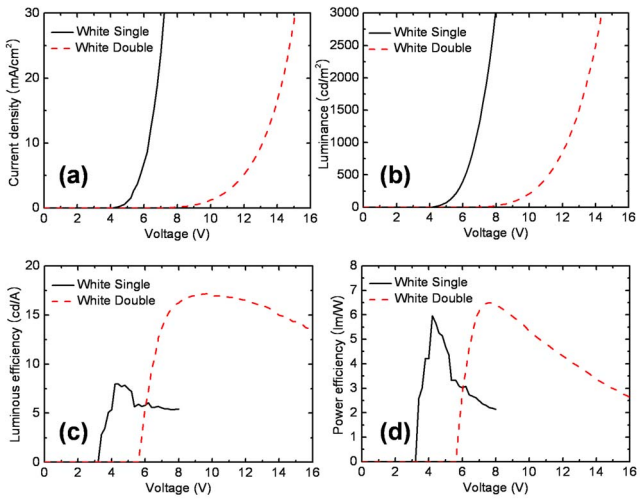

FIG. 3. (Color online) Current-voltage-luminance characteristics of single-unit (standard) and double-unit (stacked) white OLED devices.

control the white emission spectrum by tuning the emission spectrum of each emitting unit separately.

Figure 4 shows the EL spectra of the blue and white OLED devices by comparing the single-unit OLEDs with the double-unit stacked OLEDs. It is interesting that the EL spectrum of the blue tandem device changed very slightly with respect to the single-unit standard device. However, it is observed that the green emission is more generated from the tandem structure, which can be attributed to the optical effect stemming from the different optical paths of the emitted light. The WOLEDs based on the tandem structure also generated more greenish emission, as Fig. 4(b) shows.

In summary, tandem blue and white OLEDs by using BCP:Cs_2CO_3/MoO_3 as an effective interconnecting layer are demonstrated. We found that the BCP:Cs_2CO_3/MoO_3 interconnection unit works very well in blue and white stacked devices, as we observed almost doubled operating voltages and current efficiencies for the tandem devices in comparison with the single-unit devices. We utilized the interconnection unit to obtain highly efficient fluorescent WOLEDs. We utilized two primary colors of sky blue and orange fluorescent emitters to achieve efficient white EL. Although single-unit WOLEDs using two adjacent emitting layers (sky blue and orange) showed a maximum current efficiency of 7.96 cd/A with CIE coordinates of (0.28, 0.34), the tandem WOLED device made by stacking two single color OLEDs in series demonstrated more than doubled maximum current efficiency of 17.14 cd/A with CIE coordinates of (0.28, 0.41). Our results suggest that the stacking of different single color OLEDs in series instead of double stacking of WOLEDs provides highly efficient fluorescent white emission. This kind of asymmetric stacking approach of different single color units in series using an effective interconnection unit to make highly efficient WOLEDs can be very useful as it can reduce the number of layers in the devices and we do not need to control the charge transport/blocking/balance to get a right exciton profile for white emission from the adjacently deposited different-color-emitting layers in a single emitting unit. However, one thing we have to check in the asymmetric tandem devices is whether the two stacks tend to age differently during operation, which can be an important issue for WOLEDs. We are going to study this issue as a next research topic.

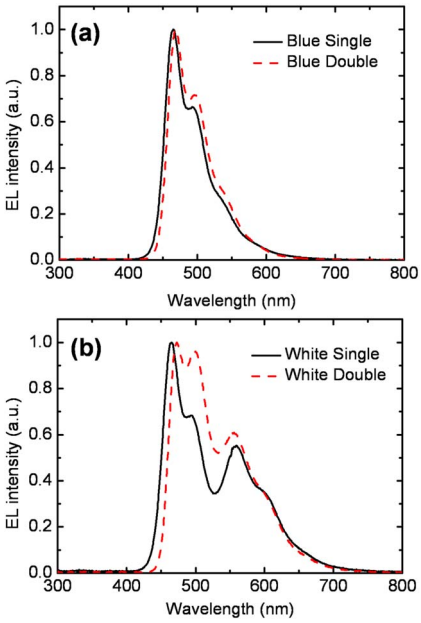

FIG. 4. (Color online) Electroluminescent spectra of single-unit (standard) and double-unit (stacked) blue and white OLED devices, which were measured at the current density of 1 mA/cm^2.

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