

Influence of connecting units' thicknesses on tandem organic devices' performances

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Abstract

The present work investigates the influence of the Alq₃:Mg and MoO₃ thicknesses in the connecting unit on the performance of tandem organic light-emitting devices (OLEDs). By systematically varying the Alq₃:Mg and MoO₃ thicknesses, we obtained a higher current efficiency of 37.3 cd/A for a device with 30 nm Alq₃:Mg and 3 nm MoO₃ layer as connecting units. The optimal device performance is enhanced by at least 14%, compared with those of devices we fabricated in this paper. It suggests that appropriate Alq₃:Mg and MoO₃ thicknesses can enhance the charge generating ability for connecting units. On the other hand, it was found that the charge transporting layer would decrease strongly because of much thicker or thinner MoO₃ thicknesses. The results demonstrate that it is an effective method to improve the performance of OLEDs by using an optimal thickness for Alq₃:Mg and MoO₃ layers.

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1. Introduction

Tandem-type organic light-emitting diodes (OLEDs) are recently reported to be used for providing high luminance and enhancing the current efficiency, for tuning the emission spectra of devices through stacking units emitting different colors [1,2]. Tandem OLEDs generally consist of two or more emissive units, which are electrically connected by connecting units in series. Under an applied electric field, electrons and holes are generated within the connecting unit and injected into adjoining elements. Kido found that the luminance efficiency of the tandem device with N units is usually N times as high as that of the single-unit device. The performance of tandem OLEDs depends critically on how efficient carriers can be injected into the emissive units. The exploration of high-performance connecting units is paid much attention [3,4,7,10–13]. A concept typically consisting of a p–n junction is used

widely. Metal oxides, such as V₂O₅ [3], MoO₃ [5–7] and WO₃ [8,12], are used as the p-type side of the junction. The n-type layer is formed by doping an electron-transporting layer (ETL) with a low work function metal [6,9]. In order to explain the mechanism of the connecting unit, Tsutsui and Terai [3,4] proposed a concept of electric field-assisted bipolar charge spouting in organic solid films. But the influence of Alq₃:Mg thicknesses and MoO₃ thicknesses has not been reported. In this report, we give a discussion for the influence of Alq₃:Mg thicknesses and MoO₃ thicknesses. Moreover, we proposed some reasonable explanation for the cause of the influence by the connecting unit.

2. Experiments

The tandem OLEDs were grown on a glass substrate pre-coated with a 150 nm thick, 20 Ω sq⁻¹ ITO layer. Prior to organic-film deposition, the ITO-coated substrates were degreased with detergent solution and solvents, then dried

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in an oven, and finally treated in an ultraviolet–ozone chamber before loading into a vacuum evaporation system with a base pressure of under 4×10^{-6} Torr. The tandem OLEDs were fabricated with two individual emissive units, each consisting of a hole-transporting layer of NPB, an ETL of Alq₃, a hole-injecting layer of *m*-MTDATA and an exciton blocking layer of Bphen. The connecting units with a structure of Mg-doped Alq₃/MoO₃ separated the two emissive units. The concentration ratio of Alq₃:Mg was controlled at 4:1 wt%. The OLED architectures of the tandem devices and controlled are shown schematically in Fig. 1.

The electroluminescent (EL) spectra and Commission Internationale de l'Éclairage (CIE) coordinates of the devices were measured by using a PR650 spectrosan spectrometer. The luminance–voltage and current–voltage characteristics were measured simultaneously with a programmable Keithley 2400 voltage–current source. All measurements were carried out at room temperature under ambient conditions.

3. Results and Discussions

In order to get the optimal thickness of the Alq₃:Mg layer and MoO₃ layer in the connecting units for devices' performances, we firstly investigated the optimal thickness of the Alq₃:Mg layer. Several devices comprising of orange phosphorescent (OP) emissive units employing tri(8-hydroxyquinoline) aluminum (Alq₃):Mg/MoO₃ as connecting units were prepared to examine the relationship between thickness of connecting units and devices' performances. The used structure was ITO/OP/Alq₃: 20 wt% Mg (*X* nm)/MoO₃ (3 nm)/OP/LiF(0.8 nm)/Al (*X* = 10, 20, 30, 40 nm for devices A–D); Where OP emissive units are 4,4',4''-tris (3-methylphenylphenyl-lamino)-triphenylamine (*m*-MTDATA: 30 nm)/*N,N'*-bis-(1-naphthyl)-*N,N'*-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB: 20 nm)/CBP: 8% (F-BT)₂Ir(acac) (30 nm)/4,7-diphenyl-1,10-phenanthroline (Bphen: 20 nm)/Alq₃ (10 nm). [CBP and (F-BT)₂Ir(acac) stand for 4,4'

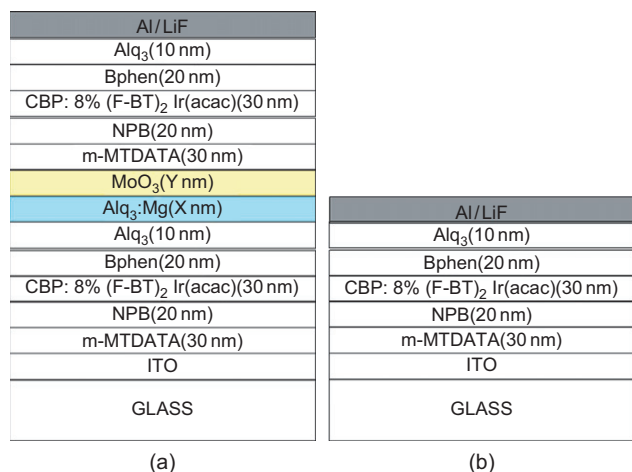


Fig. 1. Schematic diagram of (a) the controlled and (b) tandem devices.

*N,N'*dicarbazole-biphenyl and bis (2-(2-fluorophenyl)-1,3-benzothiazolato-*N,C2'*) iridium(acetylacetonate)]. The OLED architectures of tandem devices are shown schematically in Fig. 1(a). Here, we fixed the thickness of the MoO₃ layer as 3 nm and varied the thickness of the Alq₃:Mg layer, changing *X*, to determine the optimal thickness for Alq₃:Mg.

Fig. 2 depicts current density–current efficiency characteristics and current density–luminance (*J*–*L*) characteristics for the devices with different thicknesses of the Alq₃:Mg layer. In order to illuminate the function of Mg:Alq₃/MoO₃, a conventional control device with the structure of ITO/*m*-MTDATA (30 nm)/NPB (30 nm)/CBP:8% (F-BT)₂Ir(acac) (30 nm)/Bphen (20 nm)/Alq₃ (20 nm)/LiF (0.8 nm)/Al was fabricated firstly, as shown in Fig. 1(b). From Fig. 2(a), the current efficiency of device C is mostly twice over that of the control device. For instance, the current efficiency of the control device was 17.4 cd/A at 10 mA/cm²; and that of device C was 34.0 cd/A, approximately twice that of the control device. And a highest current efficiency was observed with 30 nm Alq₃:Mg, namely device C. Consistent with other works [2,5,6,8], all the tandem OLEDs show a doubled current efficiency. The maximum current efficiency of device C was

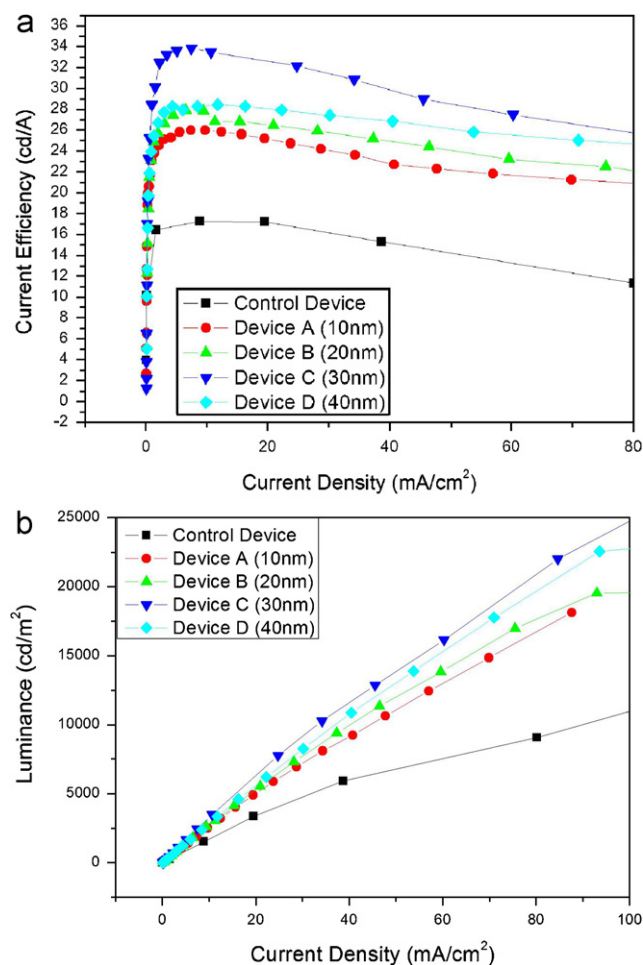


Fig. 2. (a) The current efficiency–current density–power efficiency; (b) the luminance versus voltage for devices A–D.

enhanced by at least 24% by the insertion of the connecting unit, compared with that of devices A, B and D. Moreover, the devices' performances had been improved with the thickness of the Alq₃:Mg layer changed from 10 to 30 nm; while with the thickness of the Alq₃:Mg layer changed from 30 to 40 nm, a reduction of devices' performances had been observed. Due to the results, we can infer that the thickness of 30 nm is appropriate for Alq₃:Mg to improve the organic devices' performances. The occurrence of electron transfer from the Mg to Alq₃ molecules [14,15], confirming efficient n-type doping, may contribute to the improvement for devices' performances. "Mobile electrons then reside on Alq₃ molecules in the form of Alq₃³⁻, and behave as a source for electron injection into adjacent emissive units in tandem OLEDs." [3,9]. With the increasing Alq₃:Mg thicknesses, more electrons are generated in the Alq₃:Mg layer and engaged in emitting light. But when the Alq₃:Mg thicknesses are too large, the balance of the number of holes and electrons in the emissive unit are destroyed in the same injection. Too thick an Alq₃:Mg layer cause holes to be transported from anode to cathode with difficulty. Therefore, less holes and more electrons are injected into the emitting units in the anode to cathode and anode to cathode direction, respectively. That is why current efficiency fell strongly. So higher performance for tandem devices can be obtained by a 30 nm Alq₃:Mg layer.

In order to find the correlations further between connecting units and device performance, we fixed the thickness of the Alq₃:Mg layer as 30 nm and varied the thickness of MoO₃. The structure we prepared for the purpose was ITO/OP/Alq₃:20 wt% Mg (30 nm)/MoO₃ (Y nm)/OP/LiF (0.8 nm)/Al (Y=1, 3, 5, 10, 15, 25 nm for devices E–J), where devices A–D we mentioned above and devices E–J come from different batches. Fig. 3 showed current efficiency–current density, power efficiency–current density and the *J–L* characteristics for the devices with different MoO₃ thicknesses, namely *Y*. It was found that different MoO₃ thicknesses in tandem OLEDs influenced the current efficiency, power efficiency and luminance for tandem OLEDs strongly, as shown in Fig. 3(a) and (b). We found that there is at least 14% performance enhancement, when we compare device F with any other device we mentioned above.

In Fig. 3, the highest current efficiency was obtained for device F with a 3 nm MoO₃ layer; and current efficiency was very low for device E with a 1 nm MoO₃ layer. The same trend of power efficiency for devices E–J also could be observed (see inset, Fig. 3(b)). According to the above phenomenon, because the MoO₃ layer had a good transmittance [6], the luminance reduction with increasing MoO₃ thicknesses can mainly be attributed to the decreasing number of radiated excitons in the emitting unit. Furthermore, the holes generated within the Alq₃:Mg layer were injected into the above emitting unit via MoO₃. When the MoO₃ thickness was 1 nm, the MoO₃ is not thick enough to inject holes well, which contributed to the poor performance of device E. The device performance reduces with the increasing MoO₃ thicknesses from 3 to 25 nm, which was attributed to the

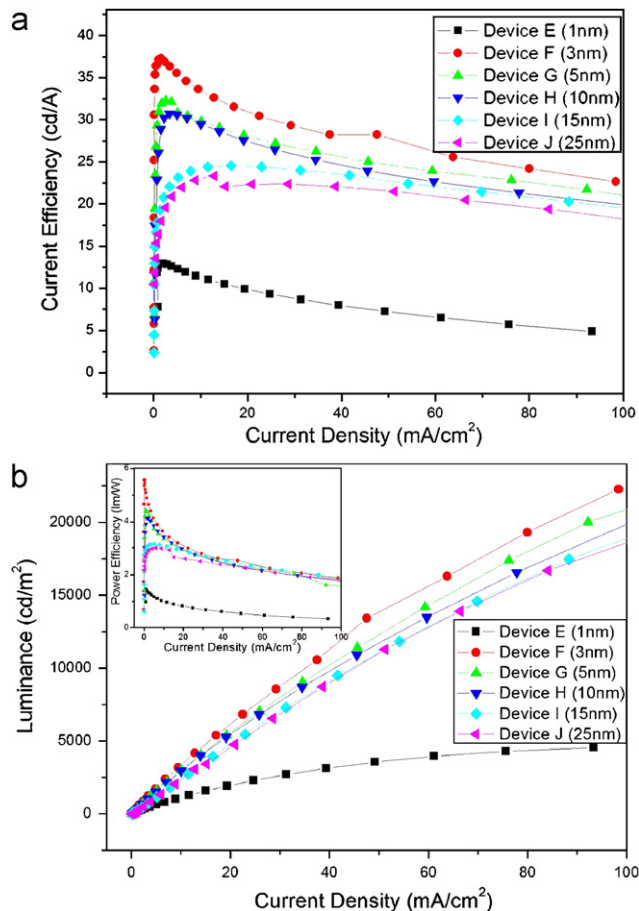


Fig. 3. (a) The current efficiency versus current density; (b) the luminance versus current density for devices E–J. Inset: the power efficiency versus current density for devices E–J.

fact that the injection of holes via MoO₃ will become more difficult because MoO₃ is a dielectric after all, namely it has a bad charge transporting ability. So we can conclude that a 3 nm MoO₃ layer is an optimal thickness for improving devices' performance and charge transporting ability.

4. Conclusion

In summary, connecting units in tandem OLEDs will influence the performance of OLEDs strongly. An optimal thickness for the Alq₃:Mg layer and MoO₃ layer can enhance current efficiency and power efficiency, respectively. The optimal thickness for the Alq₃:Mg layer is 30 nm, and that for the MoO₃ layer is 3 nm. Our results indicate that the optimal thicknesses of connecting units improve the performance of OLEDs, making it suitable for commercial applications, especially for the development of light sources.

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References

- [1] T. Matsumoto, T. Nakada, J. Endo, K. Mori, N. Kavamura, A. Yokoi, J. Kido, Society for Information Display (SID) International Symposium Digest of Technical Papers, Baltimore, MD, 2003. p. 979.
- [2] L.S. Liao, K.P. Klubek, C.W. Tang, *Appl. Phys. Lett.* 84 (2004) 167.
- [3] T. Tsutsui, M. Terai, *Appl. Phys. Lett.* 84 (2004) 440.
- [4] T. Tsutsui, M. Terai, *Appl. Phys. Lett.* 90 (2007) 083502.
- [5] C.W. Chen, Y.J. Lu, C.C. Wu, E.H.E. Wu, C.W. Chu, Y. Yang, *Appl. Phys. Lett.* 87 (2005) 121–241.
- [6] H. Kanno, R.J. Holmes, Y. Sun, S.K. Cohen, S.R. Forrest, *Adv. Mater.* 18 (2006) 339.
- [7] Ping Chen, et al., Stacked white organic light-emitting devices comprising of blue fluorescent and orange phosphorescent emissive units, unpublished.
- [8] C.C. Chang, J.F. Chen, S.W. Hwang, C.H. Chen, *Appl. Phys. Lett.* 87 (2005) 253–501.
- [9] Mei-Yee Chan, Shiu-Lun Lai, Kit-Ming Lau, Man-Keung Fung, Chun-Sing Lee, Shuit-Tong Lee, *Adv. Funct. Mater.* 17 (2007) 2509.
- [10] Fawen Guo, Dongge Ma, *Appl. Phys. Lett.* 87 (2005) 173510.
- [11] Ping Chen, Wenfa Xie, Jiang Li, Tao Guan, Yu Duan, Yi Zhao, Shiyong Liu, Chunsheng Ma, Liying Zhang, Bin Li, *Appl. Phys. Lett.* 91 (2007) 023505.
- [12] Hongmei Zhang, Yanfeng Dai, Dongge Ma, Hongmei Zhang, *Appl. Phys. Lett.* 91 (2007) 123504.
- [13] Chieh-Wei Chen, Yin-Jui Lu, Chung-Chih Wu, Elbert Hsing-En Wu, Chih-Wei Chu, Yang Yang, *Appl. Phys. Lett.* 87 (2005) 241121.
- [14] A.J. M_kinen, M. Uchida, Z.H. Kafafi, *Appl. Phys. Lett.* 82 (2003) 3889.
- [15] S.T. Lee, Y.M. Wang, X.Y. Hou, C.W. Tang, *Appl. Phys. Lett.* 74 (1999) 670.