

Improved performance of organic light-emitting diodes with cesium chloride inside tris (8-hydroxyquinoline) aluminum

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ARTICLE INFO

Article history:

Received 26 February 2010

Received in revised form

22 July 2010

Accepted 29 September 2010

Available online 8 October 2010

Keywords:

Cesium chloride

Electron injection

Trap site

Insulating layer effect

ABSTRACT

A series of small molecular organic light-emitting diodes (OLEDs) based on tris (8-hydroxyquinoline) aluminum (Alq₃) was fabricated by varying thicknesses and positions of cesium chloride (CsCl) layer inside Alq₃ layer. Both luminance and efficiency are enhanced due to the improvement of electron injection when a CsCl layer was deposited between Alq₃ and aluminum (Al). For the insertion of the CsCl layer at the 10 nm position inside Alq₃ layer away from Al cathode, the enhanced current density and luminance are attributed to the reaction between diffused Al and Cs. And the efficiency and luminance are enhanced due to the trap sites induced by the CsCl layer at the distance of 20 and 30 nm away from the Al cathode. The current density and luminance of devices, in which various thicknesses of CsCl layer was inserted at 20 nm position inside Alq₃ layer away from the Al cathode, is affected by both hole trapped and insulating layer effects.

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1. Introduction

Organic light-emitting diodes (OLEDs) are extensively attracting tremendous interest due to their potential as solid-state lighting sources [1,2] as well as next generation technology for flat-panel display [3]. Much of recent effort has been devoted to optimizing OLED device performance, and to achieve this goal, balanced charge carriers need to efficiently recombine in the organic layer.

In typical heterojunction OLEDs, hole mobility is larger than that of electron and hole injection barrier is lower than electron injection barrier. Such devices have extra holes in the emission region and electrons are more or less consumed before reaching emissive layer, which decreases the device efficiency. In recent years, research on the effective electron injection in OLEDs has a significant progress for achieving a perfect balance between holes and electrons [4–10]. A buffer layer between electron transport layer (ETL) and aluminum (Al) cathode effectively lowered electron injection barrier and significantly enhanced performance of OLEDs.

It is reported that the electron injection can be improved by inserting cesium compounds between organic material and Al [11–18]. And cesium (Cs) was doped into the ETL material to serve as the *n*-type electron injection layer and it was effective to improve electron injection. That is, Cs has *n*-type doping effect when it

combined with electron transport materials [19–22]. As it is known, a better charge carrier balance would be reached owing to the enhancement of electron injection. Consequently, the device performance also can be improved.

Cesium chloride (CsCl), easily processable, is one of cesium compounds. It has a melting temperature of 645 °C and a sublimation temperature of 450–500 °C at a pressure of 1×10^{-4} Pa. Additionally, CsCl is stable under ambient condition at normal temperature. And, to the best of our knowledge, there are rare reports on the effect of CsCl in OLEDs. In this literature, we carried out systematic studies on the role of CsCl as the cathode modification buffer layer and inside tris (8-hydroxyquinoline) aluminum (Alq₃) layer in OLEDs. The results show that CsCl is an effective buffer layer between Alq₃ and Al for enhancing electron injection in OLEDs and that CsCl layer inserted inside Alq₃ layer can also improve the luminance and efficiency of OLED devices.

2. Experiment

To fabricate the devices, the ITO substrates with a sheet resistance of 20 Ω/□ were first cleaned using a routine procedure, included sonication in detergent, repeated rinsing in deionized water, acetone, and alcohol, and finally treatment with UV light and ozone for 10 min. And the cleaned substrates were then transferred to an evaporation chamber. The 40 nm N, N'-bis-(1-naphthyl)-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB, hole transport layer), 60 nm tris (8-hydroxyquinoline) aluminum (Alq₃, electron

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transport and emitter layer), 15 nm 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP, hole blocking layer) and CsCl varied from 0.3 to 2.0 nm were evaporated on the substrates in turn under a pressure 2×10^{-4} Pa, followed by the thermal deposition of Al from tungsten boats in an other chamber at a pressure of around 3×10^{-3} Pa. The thickness and deposition rate of the materials are monitored by a quartz–crystal oscillator during evaporation. The molecular structures of organic materials used in the experiment are referred to Ref. [23].

First, three sets of devices were fabricated with configurations as follows:

Set 1: ITO/NPB(40 nm)/Alq₃(60 nm)/CsCl(0.0, 0.3, 0.5 and 1.0 nm)/Al;

Set 2: ITO/NPB(40 nm)/Alq₃(60-*d* nm)/CsCl(1.0 nm)/Alq₃(*d* nm)/Al, where *d* = 10, 20 and 30 nm and the only-Al device of ITO/NPB(40 nm)/Alq₃(60 nm)/Al as reference;

Set 3: ITO/NPB(40 nm)/Alq₃(40 nm)/CsCl(0.5, 1.0, 1.5 and 2.0 nm)/Alq₃(20 nm)/Al.

In the Set 1 experiment, the effects of CsCl layer are explored by inserting various thicknesses of CsCl layer between Alq₃ and Al as an electron injection layer. In Sets 2 and 3, the effects of CsCl inside the Alq₃ layer are investigated by varying its position and thickness.

To further investigate the function of CsCl layer in OLEDs, other two sets of electron-only devices were prepared as follows:

Set 4: ITO/BCP(15 nm)/Alq₃(60-*d* nm)/CsCl(1.0 nm)/Alq₃(*d* nm)/Al, where *d* = 10, 20 and 30 nm and ITO/BCP(15 nm)/Alq₃(60 nm)/Al as a reference device;

Set 5: ITO/BCP(15 nm)/Alq₃(40 nm)/CsCl(0.5, 1.0, 1.5 and 2.0 nm)/Alq₃(20 nm)/Al.

The current–voltage–luminance (*I*–*V*–*L*) characteristics (the emissive size of devices was defined as 9 mm²) were measured using a Keithley 2410 Source Meter and a silicon photodiode calibrated by PR-650. The EL spectra were taken by a charge-coupled device (CCD) spectrometer. All measurements were carried out at room temperature under ambient conditions immediately after the devices have been fabricated.

3. Results and discussion

In order to determine how the CsCl electron injection layer functions, the first set of OLEDs has been fabricated with various CsCl thicknesses (0.3, 0.5 and 1.0 nm). The current density–luminance–voltage (*J*–*L*–*V*) and power efficiency–current density (η_p –*J*) characteristics are depicted in Fig. 1 (a) and (b) for devices with various CsCl thicknesses as an electron injection layer, respectively. All devices with CsCl as an electron injection layer exhibit an increased current density and luminance in forward bias compared to the only-Al device (without CsCl).

The maximum luminance (5435 cd/m²) of the bare Al device is the lowest among all devices since the injection barrier for electrons, taken as the difference between the lowest unoccupied molecular orbital (LUMO) of Alq₃ (~3.3 eV) [23] and the Fermi level of Al (*E*_F, 4.1 eV) [24], is rather high. However, for the devices with CsCl as an electron injection layer, the reaction of Cs and Al can be capable of reducing the work function of Al [14,16]. As a result, the barrier for electron injection is lowered. The resulting low electron injection barrier facilitates electron injection, which improves the charge carrier balance in the OLED and leads therefore to a superior performance. From Fig. 1, it can be seen that the optimum thickness of CsCl as an electron injection layer is determined to be about 0.5 nm. The device with 0.5 nm CsCl layer produces a luminance of

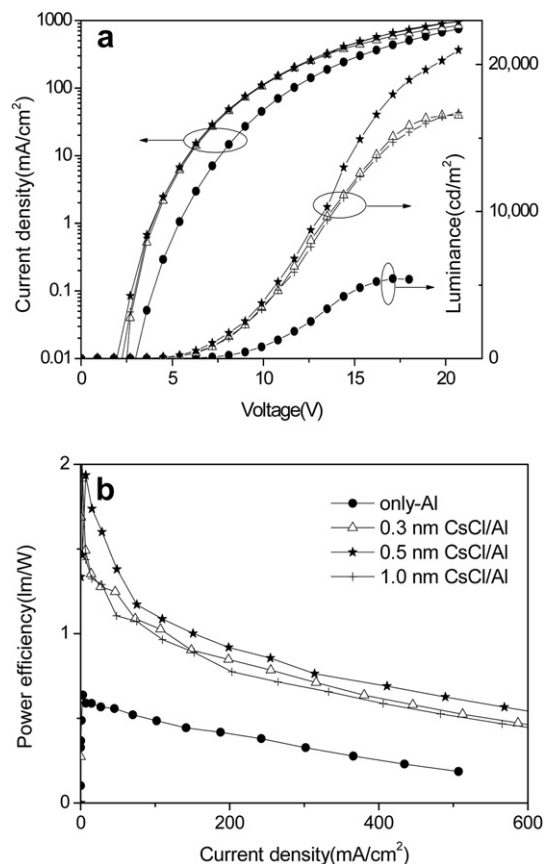


Fig. 1. (a) Current density–luminance–voltage (*J*–*L*–*V*) and (b) Power efficiency–current density (η_p –*J*) characteristics of devices with various CsCl thicknesses as an electron injection layer, respectively. Without CsCl layer (full circle), 0.3 nm (open up triangle), 0.5 nm (full star), 1.0 nm (cross).

above 20,000 cd/m². Correspondingly, the maximum power efficiency of 1.94 lm/W at the current density of 6.7 mA/cm² is three times as high as that of the only-Al device (0.64 lm/W @ 3 mA/cm²).

Then, the discussion of results for the devices with CsCl inside Alq₃ layer (Set 2) is followed. The characteristics of devices are shown in Fig. 2. From Fig. 2, both electron injection and luminance have been increased in ITO/NPB(40 nm)/Alq₃(60-*d* nm)/CsCl(1.0 nm)/Alq₃(*d* = 10 nm)/Al device compared to the reference device ITO/NPB(40 nm)/Alq₃(60 nm)/Al. Yet, the ITO/NPB(40 nm)/Alq₃(60-*d* nm)/CsCl(1.0 nm)/Alq₃(*d* = 20 and 30 nm)/Al devices do not show the enhancement of current density but demonstrate the enhanced luminance. The EL efficiency of all devices with the CsCl layer is enhanced by more than 80% compared to the reference device without the interlayer as shown in inset of Fig. 2(b).

For the device with CsCl at *d* = 10 nm position inside Alq₃ layer away from the Al cathode, the reaction between the diffused Al [25] and Cs reduces the effective barrier for electron injection. Therefore, the current density is increased and subsequently the luminance is also enhanced. If the distance of CsCl inside Alq₃ layer is more than 20 nm away from the Al cathode, the Al diffusion reaches the interlayer of CsCl no longer and the devices show luminance enhancement without enhanced injection, which is attributed to the trap sites induced by the CsCl layer, similarly the cases of Han [25] and our previous work [26]. The trap sites could trap holes leakage to the cathode and subsequently the current density is decreased. The trapped charges alter the field distribution inside OLEDs and enhance the recombination of devices.

The results are followed by varying thicknesses of CsCl at 20 nm position away from the Al cathode. A set of four types of devices

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