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Enhancement of Luminance Efficiency in Green Organic Light-Emitting Devices Utilizing a Cesium Nitrate/Lithium Quinolate Electron Injection Layer

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The current density and the luminance efficiency of the organic light-emitting devices (OLEDs) with a cesium nitrate (CsNO₃)/lithium quinolate (Liq) electron injection layer (EIL) were larger than those of the OLEDs with a Liq EIL. Residual Cs ions or cesium oxides in the OLEDs decreased the electron affinity of the cathode electrode, resulting in an improvement of the electron injection efficiency and the luminance efficiency. Electron only devices, which are elements being able to flow only electrons, showed that the electron injection magnitude was increased due to the insertion of the CsNO₃ EIL into OLEDs.

Keywords: organic light-emitting device; double electron injection layer; luminance; CsNO₃; Liq; electron only devices

Introduction

Organic light-emitting devices (OLEDs) have become particularly interesting because of their promising applications in next-generation full-color displays and lighting sources [1–5]. Moderate controls of carrier injection and transport for the efficient carrier recombination and for the balance of the electrons and the holes in the emitting layer (EML) are necessary to fabricate high-efficiency OLEDs [6, 7]. Because the hole mobility of almost all of organic layers is larger than their electron mobility, the increase of the electron injection is necessary to enhance the luminance efficiency of the OLEDs. Some works concerning the device properties and the luminance mechanisms of OLEDs with a lithium fluoride (LiF)/Al cathode, a lithium quinolate (Liq)/Al cathode, or a cesium fluoride/Al cathode have been conducted to enhance the luminance efficiency of the OLEDs [8–10]. While the metal ions with a low work function in the electron injection layer (EIL) reduce the electron injection barrier, they decrease the stability of OLEDs due to their high reactivity [11–13]. Cesium carbonates with a low work function have been used as an EIL in OLEDs to enhance their stability of the OLEDs [14–16]. However, codeposition process of organic materials with a low sublimation temperature in OLEDs and metal derivatives with a high sublimation temperature releases organic materials from the codeposited layer due to high thermal energy during the evaporation [17], resulting in the contamination of

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the organic layer due to the generation of carbon oxides. Because a cesium nitrate (CsNO_3) EIL containing nitrogen oxides does not contaminate organic layers, the device stability and the luminance efficiency of OLEDs with a CsNO_3 can be enhanced. Even though some works on the enhancement of the luminance efficiency in green OLEDs with various EILs have been performed, systematic studies concerning the electron injection mechanisms in OLEDs fabricated utilizing a CsNO_3 /Liq EIL are very important for improving the luminance efficiency of the OLEDs.

This paper reports data on the luminance efficiency enhancement in green OLED with a CsNO_3 /Liq EIL fabricated by using organic thermal evaporation. Current density-voltage, luminance-voltage, and luminance efficiency-current density measurements on OLEDs with a CsNO_3 /Liq EIL were carried out to investigate their electrical and the optical properties. Current density-voltage measurements on electron only devices (EODs), which are elements being able to flow only electrons, were performed to investigate the enhancement of the electron injection magnitude due to the insertion of the CsNO_3 EIL. The electron injection mechanisms related to the luminance efficiency enhancement of the OLEDs with a CsNO_3 /Liq EIL are described on the basis of the experimental results.

Experimental

The sheet resistance and the thickness of the indium-tin-oxide (ITO) thin films coated on glass substrates used in this study were $15 \Omega/\text{square}$ and 100 nm, respectively. The ITO-coated substrates were cleaned by using acetone and methanol at 35°C for 5 min, and then rinsed in de-ionized water thoroughly. After the chemically cleaned ITO substrates had been dried by using N_2 gas with a purity of 99.9999%, the surfaces of the ITO substrates were cleaned with an ozone gas generated by an UV radiation for 10 min. The four kinds of OLEDs used in this study were deposited on ITO-coated glass substrates. The OLEDs consisted of the following structures from the bottom: a glass substrate/an ITO anode electrode (100 nm)/a N,N'-di(1-naphthyl)-N,N'-diphenylbenzidine (NPB) hole transport layer (40 nm)/a tris(8-hydroxyquinoline) (Alq_3) emission layer (EML) (30 nm)/an Alq_3 or 4,7-diphenyl-1-10-phenanthroline (BPhen) electron transport layer (ETL) (30 nm)/a CsNO_3 EIL (5 nm) or no layer/a Liq EIL (0.5 nm)/an Al cathode (150 nm). The four kinds of EODs corresponding to devices I, II, III, and IV were deposited on ITO-coated glass substrates. The organic and metal layers were deposited at a system pressure of 3×10^{-7} Torr. After organic and metal layers were deposited on the ITO-coated on glass substrates, the OLEDs were encapsulated in a glove box containing N_2 gas under a H_2O concentration below 1 ppm. A desiccant material consisting of a barium-oxide powder was used to absorb the residual moisture and oxygen in the encapsulated device. The schematic diagrams of devices I, II, III, and IV and of their corresponding EODs are shown in Fig 1. The current-voltage characteristics of the OLEDs were measured by using a programmable electrometer with built-in current and voltage measurement units (M6100, McScience). The brightness was measured by using a brightness meter, and the EL spectrum was measured by using a luminance spectrometer (CS-1000A, Minolta).

Results and Discussion

The schematic diagrams of the OLEDs and their EODs used in this study are shown in Fig. 1. The Liq in the EIL of device I is mostly separated into the Li and quinolate ions due to the low binding energy of the Liq molecules, resulting in a diffusion of the Li ions into the Alq_3 ETL and an increase in the electron mobility of the ETL. However, the Li-combined

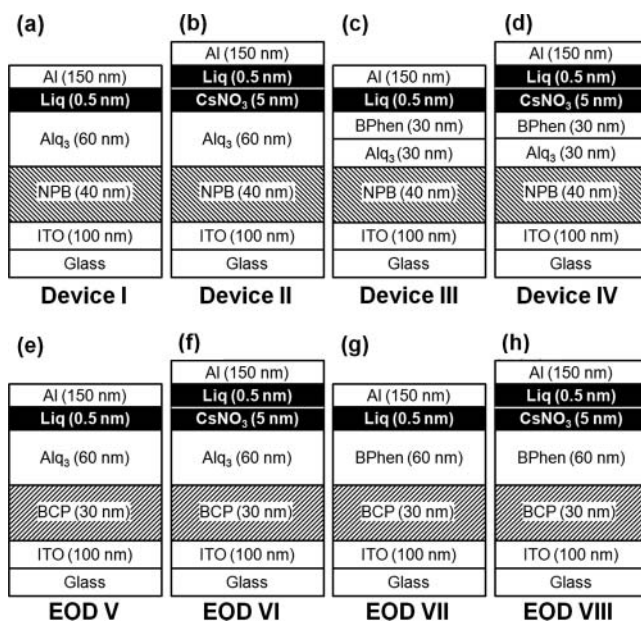


Figure 1. Schematic diagrams of the OLEDs of devices (a) I, (b) II, (c) III, and (d) IV and the EODs (e) V, (f) VI, (g) VII, and (h) VIII.

Alq₃ molecules in the EML can quench the excitons generated in the EML, resulting in a decrease in the luminance efficiency of the OLEDs. When the CsNO₃ EIL of the devices II and IV is thermally evaporated in a vacuum chamber, only Cs ions or cesium oxides remain in their EIL due to the generation of NO₂ and O₂ gases [18, 19]. Because the atomic weight of the Cs ion is larger than that of the Li ion, the diffusion length of the Cs ion is smaller than that of the Li ions, resulting in the decrease of the number of the quenching sites in comparison with that of the OLEDs with a Liq EIL. The combined CsNO₃ layer of devices II and IV isolated the Liq EIL from the Alq₃ or BPhen ETL, resulting in the decrease of the Li diffusion for the Liq EIL. While the EIL and the ETL of EODs V, VI, VII, and VIII are the same materials as devices I, II, III, and IV, the BCP hole blocking layer (HBL) in EODs substitutes to the EML and the HTL of the OLEDs. Because the BCP HBL of EODs at low voltages suppresses the hole injection from the ITO anode into the BPhen ETL, the current densities of EODs are dominantly attributed to the electrons injected from the EIL.

Figure 2 shows current densities as functions of the applied voltage of devices I, II, III, and IV. The current densities of devices II and IV are much larger than those of devices I and III due to the insertion of the n-type CsNO₃ EIL. Because almost all of the NO₂ and O₂ molecules in the CsNO₃ layer of devices II and IV outgas during evaporation, the Cs ions or cesium oxides form at the heterointerface between the Alq₃ ETL and the Liq EIL. The residual Cs ions or cesium oxides decrease the surface electron affinity of the cathode electrode. Because the dipole moments generated by the Liq and the CsNO₃ EILs increase the internal electric field of the EIL, the electron injection magnitude from the cathode into the ETL increases. Current densities of devices III and IV are much larger than those of devices I and II because the electron mobility of the BPhen ETL is larger than that of the Alq₃ ETL. While the Li atoms diffused into the ETL increase the quantity of the electron injection, the Li atoms diffused into the EML act as exciton quenching sites, resulting in a

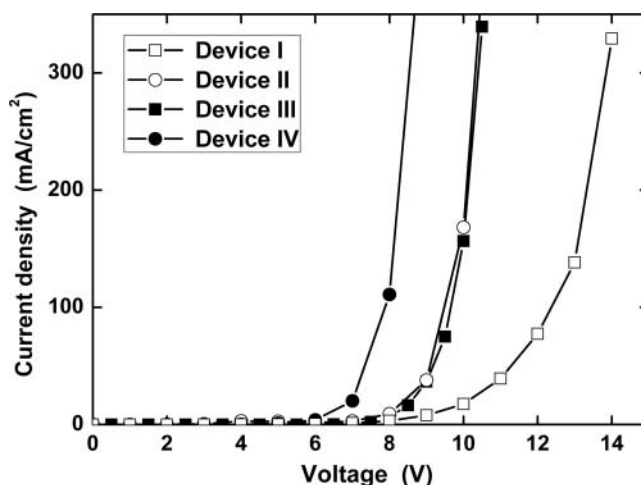


Figure 2. Current densities as functions of the applied voltage for devices I, II, III, and IV. Open rectangles, open circles, filled rectangles, and filled circles represent devices I, II, III, and IV, respectively.

decrease in the lifetime of the OLEDs. However, because the atomic weight of the Cs ions in the CsNO_3 layer in devices II and IV is larger than that of the Li atoms, the diffusion length of the Cs ions decreases, resulting in the decrease of the number of quenching sites. Furthermore, the BPhen ETL in device IV decreases an operating voltage due to the high electron mobility of the BPhen ETL.

Figure 3 shows the luminances as functions of the applied voltage of devices I, II, III, and IV. Luminances of devices II and IV are much larger than those of devices I and III due to the insertion of the CsNO_3 EIL. The maximum luminance of devices I, II, III, and IV are 8,675, 15,860, 10,310 and 20,050 cd/m^2 , respectively, and the turn-on voltages of devices I, II, III, and IV at 100 cd/m^2 are 8, 7.5, 7.5, and 6 V, respectively. Because Cs

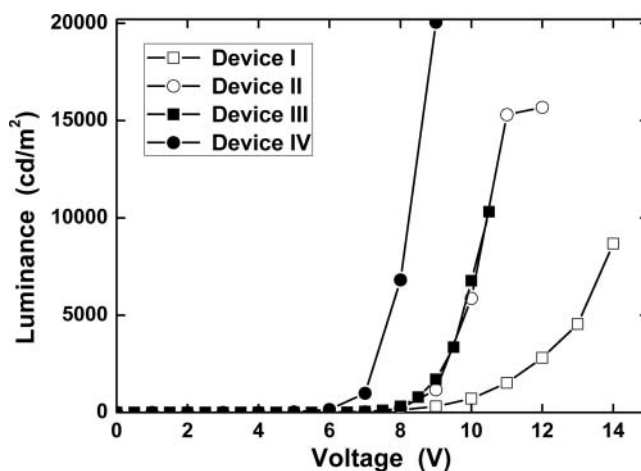


Figure 3. Luminance as functions of the applied voltage for devices I, II, III, and IV. Open rectangles, open circles, filled rectangles, and filled circles represent devices I, II, III, and IV, respectively.

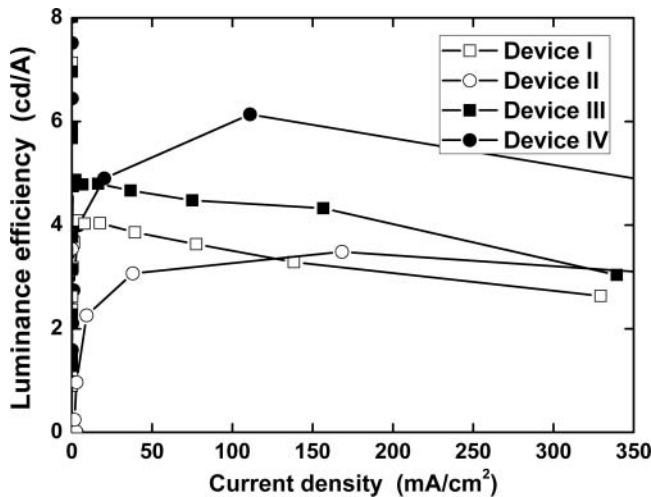


Figure 4. Luminance efficiencies as functions of the current density for devices I, II, III, and IV. Open rectangles, open circles, filled rectangles, and filled circles represent devices I, II, III, and IV, respectively.

ions or cesium oxides of the CsNO_3 EIL reduce the electron affinity of the surface of the cathode electrode, resulting in the decrease of the electron injection barrier for the Liq EIL, the electrons of the cathode electrode easily inject into the ETL, resulting in the luminance enhancement due to the achievement of more balance of electrons and holes in the EML.

Figure 4 shows the luminance efficiencies as functions of the current density for devices I, II, III, and IV. Because the electron mobility of the BPhen ETL in devices III and IV is much larger than that of the Alq_3 ETL in devices I and II, the luminance efficiencies of devices III and IV are much larger than those of devices I and II. The maximum luminance

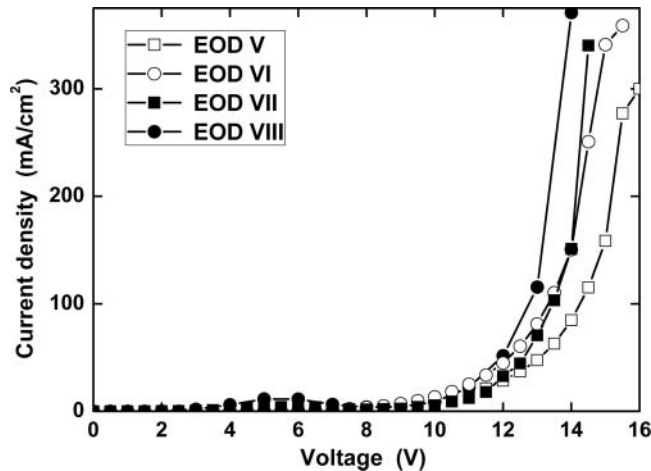


Figure 5. Current densities as functions of the applied voltage for EODs V, VI, VII, and VIII. Open rectangles, open circles, filled rectangles, and filled circles represent devices V, VI, VII, and VIII, respectively.

efficiencies of devices I, II, III, and IV are 4.08, 3.48, 4.87, and 6.13 cd/A, respectively. The luminance efficiency of the OLEDs is significantly affected by using Cs ions, as shown in Fig. 4. The luminance efficiency of the OLEDs without Cs ions decreases with increasing current density, resulting in the generation of the exciton quenching due to the Joule heating.

Figure 5 shows the current densities as functions of the applied voltage for EODs V, VI, VII, and VIII. The current densities of EODs VI and VIII are much larger than those of EODs V and VII. Because the hole injection in EODs is suppressed by the insertion of the BCP HBL, the difference between the current densities of EODs is dominantly attributed to the difference between the numbers of electrons injected from the cathode. The decrease of the operating voltages between devices I and II or between devices III and IV originates from the insertion of the CsNO₃ EIL. The existence of the Cs ions or cesium oxides in the CsNO₃ EIL results in the decrease of the operating voltages due to the decrease of the injection barrier of the EIL. This result indicates that the electron injection magnitude was increased due to the insertion of CsNO₃ EIL into OLEDs.

Conclusions

The electrical and the optical properties of OLEDs utilizing a CsNO₃/Liq EIL were investigated. The current density and the luminance of devices with a CsNO₃/Liq EIL were much larger than those of devices I and III. Because Cs ions or cesium oxides of the CsNO₃ EIL reduced the electron affinity of the surface of the cathode electrode, resulting in the decrease of the electron injection barrier for the Liq EIL, the number of the injected electrons in the EML increased, resulting in an enhancement of the luminance efficiency due to an achievement of more balance of electrons and holes. EODs showed that the electron injection magnitude was increased due to the insertion of CsNO₃ EIL into OLEDs. The electron injection mechanisms of the OLEDs with a CsNO₃/Liq EIL are described on the basis of the experimental results. These results can help improve understanding of the electron injection mechanisms correlated to the luminance efficiency enhancement in OLEDs utilizing a CsNO₃/Liq bilayer EIL.

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