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Improved Efficiency and Lifetime of Organic Light-Emitting Diode with Lithium-Quinolate-Doped Electron Transport Layer

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In this letter, we analyze the device performance of organic light-emitting diodes (OLEDs) with a lithium-quinolate (Liq)-doped electron transport layer (ETL). When the ETL:Liq doping ratio is 1 : 3, the luminous efficiency of the OLED is 7.1 cd/A; that of an OLED with a nondoped ETL is 5.3 cd/A. The corresponding OLED lifetime is improved by threefold that with a nondoped ETL, owing to the improved hole-electron charge balance and electrical stability of the Liq-doped ETL film. Additionally, when the Liq-doped device was combined with a Mg/Al cathode, the OLED produced had a longer lifetime than the other tested devices. © 2009 The Japan Society of Applied Physics

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1. Introduction

In 1987, Tang and VanSlyke developed efficient organic light-emitting diodes (OLEDs).¹⁾ Since then, these devices have been studied intensively for their potential application in flat panel displays because of their viewing angle, wide color gamut, low cost, and fast response time. However, power consumption and stability are major issues in the use of OLEDs, which are challenges not shared by other competitive display technologies such as liquid crystal displays. In general, the mobility of an electron is less than that of the holes in an organic film. Therefore, an excess of holes in the emission layer generates nonemissive cationic species, resulting in luminance decay and the formation of dark spots.^{2,3)} Consequently, balancing the holes and electrons in an emission layer is a well-known method for improving the power consumption and stability of OLEDs. To achieve a balanced injection of carriers, electron and/or hole transport layers with either a metal or organic dopant with a high carrier mobility have been studied extensively.⁴⁻⁶⁾ A typical n-type doped electron transport layer (ETL) is a highly reactive metal with a low work function doped in an organic material.^{7,8)} Although highly reactive metals such as Li, Cs, Mg, and Ca are very effective in improving carrier injection, such metals are not stable in air and require protection during device fabrication.

In this study, we developed an n-type doped ETL to enhance the performance of conventional devices. For improving OLEDs, we use lithium quinolate (Liq) as an ETL doping material, since it is one of the organometallic complexes that have been demonstrated to assist electron injection owing to the low binding energy between Li and the quinolate ligand.⁹⁻¹¹⁾

2. Experimental Methods

OLEDs were fabricated in accordance with the following specifications applied sequentially; a hole injection layer of 5-nm-thick LGC101 (LG Chem), a hole transport layer of 100-nm-thick *N,N*-bis(1-naphthyl)-*N,N'*-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB), a blue emitting layer of 30-nm-thick ABH034 (SFC) with 4% SD1 (SFC), an electron transport layer of 20-nm-thick LGC201 (LG Chem), and an electron injection layer of 1-nm-thick Liq. A 100-nm-thick

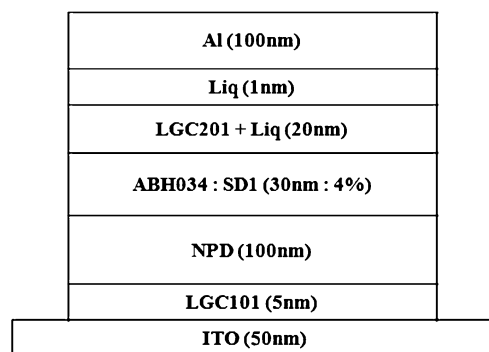


Fig. 1. Schematic structure of the fabricated OLED device with Liq-doped ETL.

aluminum layer was used as the cathode electrode. All the organic layers and the cathode were deposited successfully onto an indium tin oxide (ITO)-coated glass substrate by thermal evaporation at a base pressure of 10^{-7} Torr. To investigate the effect of the Liq-doped ETL, five devices containing different ratios of ETL:Liq were prepared. Devices A, B, C, D, and E corresponded to ETL:Liq mixed ratios of 1 : 0, 3 : 1, 1 : 1, 1 : 3, and 1 : 5, respectively. Figure 1 shows the structure of an OLED fabricated on a glass substrate with a Liq doped ETL. All the devices were encapsulated in a glass-to-glass epoxy sealed package with desiccant and then treated by thermal annealing at 90 °C for 60 min. The current density–voltage–luminance (*J–V–L*) characteristics of the devices were measured using a Keithley 236 and a PR-705 spectrophotometer (Photo Research). All the devices were encapsulated in a glove box under nitrogen atmosphere prior to measurement.

3. Results and Discussion

Figure 2 shows an energy level diagram of the proposed OLED. The energy band diagrams of synthesized organic materials were obtained from the UV absorption edges and the highest occupied molecular orbital (HOMO) energy level from the photoelectrometer AC-2 measurement. As shown in Fig. 2, the lowest occupied molecular orbital (LUMO) energy level of the ETL and Liq are 2.8 and 3.15 eV, respectively. A Liq-doped ETL can be expected to exhibit a lower electron barrier height; therefore, the electrons can more easily be injected from the cathode to the emission

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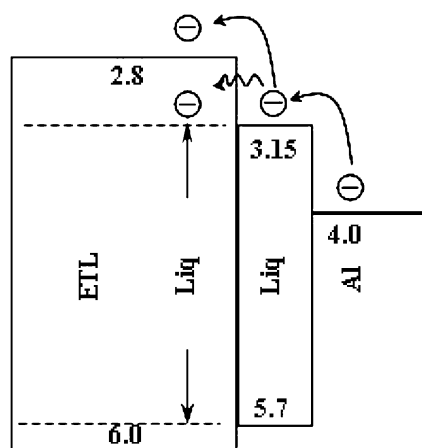


Fig. 2. Schematic energy-level diagram of Liq-doped ETL.

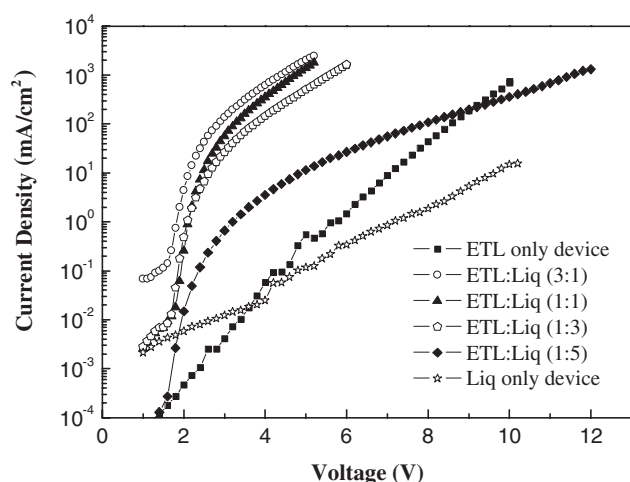
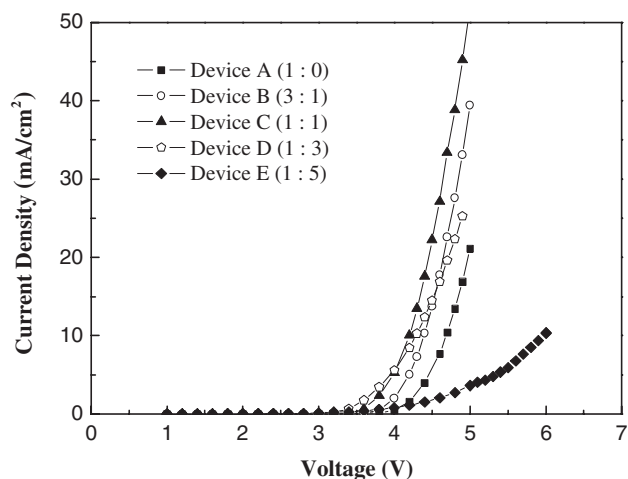


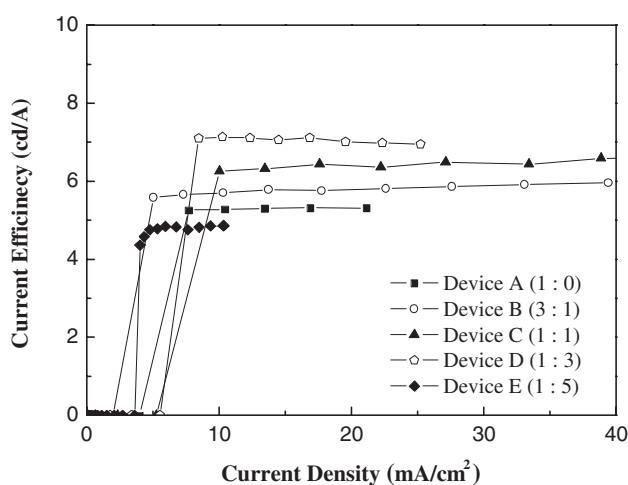
Fig. 3. Dependence of the typical current density–voltage (J – V) characteristics of Liq-doped electron-only devices.

layer. To compare the electron conduction characteristics of the Liq-doped ETLs, an electron only device composed of glass substrate/ITO (50 nm)/nondoped ETL (50 nm) or Liq-doped ETL (50 nm)/Al (100 nm) was fabricated. As shown in Fig. 3, a Liq single layer has a low electron current flow compared to with an ETL single layer. However, the data clearly show that the electron only device with a Liq-doped ETL has a high electron current flow compared with an electron only device with a nondoped ETL. As shown in the energy band diagram (Fig. 2), easier electron injection from the cathode is theoretically expected for the Liq-doped ETL than for the nondoped ETL owing to the low LUMO level of Liq. However, the electron current flow from the cathode to the EML in a heavily Liq-doped ETL may be more difficult than that from the cathode to the EML in a Liq-doped ETL owing to the low electron transporting properties of Liq. These results suggest that an optimal Liq doping ratio would enable the production of OLEDs with excellent performance.

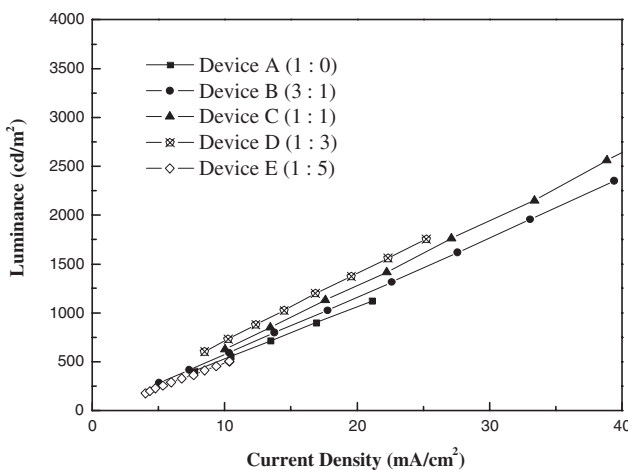
Figure 4 shows the J – V – L characteristics of five different devices. Device A, with a nondoped ETL, exhibited an operation voltage of 5.0 V at 20 mA/cm². In contrast, the devices with a Liq-doped ETL layer showed significantly reduced operation voltages. For example, devices B, C, and D showed operation voltages of 4.6, 4.4, and 4.7 V at



(a)



(b)



(c)

Fig. 4. (a) Current density–voltage, (b) current efficiency and (c) current density–luminance characteristics of the OLEDs with different Liq doping ratios.

20 mA/cm², respectively. These results demonstrate that the Liq-doped ETL generated an improved driving voltage owing to the lowered electron barrier. However, device E, with a heavily Liq-doped ETL, exhibited a considerably higher driving voltage than that of the device with a nondoped ETL. These results may be interpreted to indicate

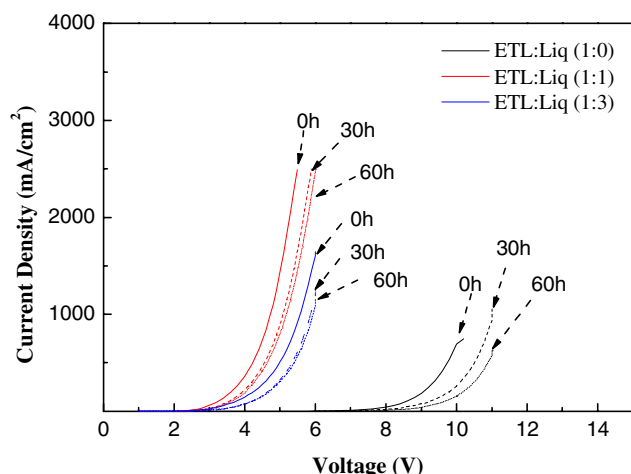


Fig. 5. (Color online) Effects of the ETL:Liq doping ratio on the current density–voltage characteristics of electron-only devices driven under a constant current of 80 mA/cm².

that a heavily Liq-doped ETL interferes with electron transport to the emission layer because it has a low electron transporting ability. The current efficiency–current density characteristics of the OLEDs with Liq-doped and nondoped ETLs are shown in Fig. 2(b). Devices B, C, and D with Liq-doped ETLs exhibited improved current efficiency (5.8–7.1 cd/A at 20 mA/cm²) compared with that of A device (5.3 cd/A), which had a nondoped ETL layer. In particular, device D showed markedly higher current efficiency and higher luminance (at 20 mA/cm²) than any other device, which can probably be attributed to the improvement in the balance of the hole–electron injection process in the emission layer.

Figure 5 shows the change in the J – V characteristics of electron-only devices before and after an electrical stress of 80 mA/cm² of constant current was applied. In the case of a non-doped ETL, the J – V performance is severely decayed by electrical stress after 60 h. In contrast, the Liq doped ETLs show no significant change even after 30–60 h. In particular, the electron-only device with an ETL:Liq doping ratio of 1 : 3 exhibited the highest electrical stability of all the devices tested in this experiment. This result clearly implies that doping of Liq in the ETL is very effective in suppressing ETL degradation, hence resulting in a significant improvement in the lifetime of the OLED.

Figure 6 shows the lifetime of devices depending on variations in the Liq doping concentration. At doping concentrations below 1,500 cd/m², devices A, B, and C showed half lifetimes of 500, 780, and 770 h, respectively. Interestingly, the lifetime of device D was threefold that of device A which had a nondoped ETL. Two explanations exist for this observation. Firstly, the devices with Liq-doped ETLs possessed comparatively better electrical stability. Secondly, the devices with Liq-doped ETLs exhibited comparatively high power efficiency; therefore, less heat would be generated during the operation of these devices. In addition, a Liq electron injection layer has also been tried in combination with a Mg/Al cathode. When a Mg (5 nm)/Al (100 nm) cathode was deposited on the organic layer, the OLEDs produced a longer lifetime which was better than that of any other device tested in the experiments. Recent

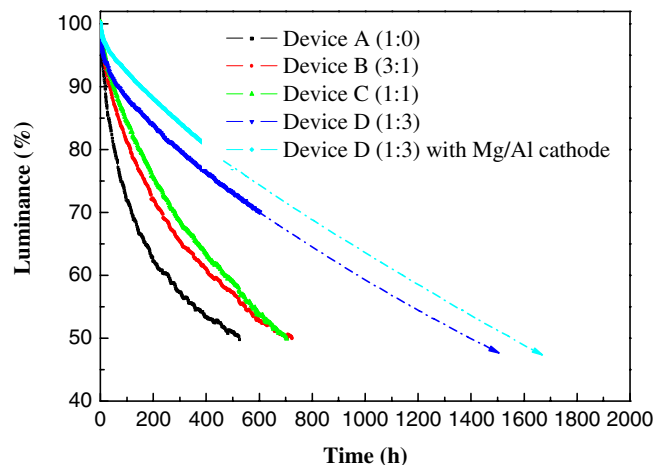


Fig. 6. (Color online) Normalized luminance–operating time characteristics of OLEDs with different Liq doping ratios.

theoretical studies have shown strong interaction between magnesium and organic materials at high temperature.¹²⁾ At room temperature, the deposited Mg cathode is weakly bonded to the quinolate ligands of the Liq. However, once the device is heated, the diffusion of Mg into the Liq layer takes place. As a result, Mg atoms insert into Liq molecules and bond with oxygen atoms in the quinolate ligand. It is probable that the relatively excellent lifetime of device D with a Mg/Al cathode is mainly caused by the interface reaction between Mg and the organic layer through thermal annealing at 90 °C for 60 min.

4. Conclusions

In this study, we have demonstrated a highly efficient and stable device prepared with a Liq-doped ETL. When the ETL:Liq doping ratio in a Liq-doped ETL is 1 : 3, the OLEDs exhibit a maximum current efficiency of 7.1 cd/A and the longest lifetime observed was 1500 h. The performance of this device illustrates improved hole–electron balance in the emissive layer due to a low electron barrier as well as the improved electrical stability of the ETL. Additionally, the Mg/Al cathode structure exhibited excellent stability owing to improved interface adhesion of the cathode and the organic layer through thermal annealing.

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