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Publisher: Taylor & Francis

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Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl20>

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Version of record first published: 17 Sep 2012.

To cite this article: Tomofumi Ikeda, Hirotake Kajii & Yutaka Ohmori (2012): Characteristics and Transient Electroluminescence of Alq₃-based Organic Light-Emitting Diodes with LiF:Al Alloy Cathode, Molecular Crystals and Liquid Crystals, 567:1, 143-148

To link to this article: <http://dx.doi.org/10.1080/15421406.2012.703437>

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Characteristics and Transient Electroluminescence of Alq₃-based Organic Light-Emitting Diodes with LiF:Al Alloy Cathode

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The emission characteristics and transient electroluminescence (EL) of organic light-emitting diodes (OLEDs) using tris(8-hydroxyquinolino)aluminum (Alq₃) as an emissive layer with LiF/Al cathode and LiF doped Al composite cathode (LiF:Al alloy cathode) were investigated. A device with LiF:Al (1:10) alloy cathode showed the maximum current efficiency of 4 cd/A. The emission characteristics of this device were almost the same as that with LiF/Al cathode. Rise time of the device with LiF:Al alloy cathode was shorter than that of a device with LiF/Al cathode, because the waveform rounding of current density occurred for the device with LiF/Al cathode.

Keywords Organic light-emitting diodes; transient electroluminescence; Alq₃; LiF

1. Introduction

Efficient charge injection is essential to improve the characteristics of organic light-emitting diodes (OLEDs). The interface of organic layer/electrode is one of important factors. For efficient charge injection, low work function metals, such as lithium, calcium, and magnesium, have been used. But the use of them results in unreliable OLEDs due to the reactive nature of these materials. In order to achieve stability and retain a performance, a thin film of an insulator, such as fluoride or oxide of alkali metal or alkali earth metal, deposited between the organic layer and the Al cathode has been used commonly. It is well-known that LiF-buffered Al cathode improves the efficiency of electron injection from the cathode and depends on the thickness of the LiF buffer layer. However, due to insulation properties of LiF, the thickness is required to be maintained 0.5 nm ranges so as not to increase driving voltage.

In this study, we investigated the emission characteristics and transient electroluminescence (EL) of OLEDs based on tris(8-hydroxyquinolino)aluminum (Alq₃) as an electron transport and emissive layer with LiF/Al cathode and LiF doped Al composite cathode (LiF:Al alloy cathode), respectively.

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2. Experimental

OLEDs were fabricated by using organic molecular beam deposition system at a background pressure of 10^{-5} Pa. The typical device structure was indium-tin-oxide (ITO)/4,4'-bis[*N*-(1-naphthyl)-*N*-phenyl-amino]-biphenyl (α -NPD) (50 nm)/Alq₃ (50 nm)/cathode. The electrodes of LiF(0.5 nm)/Al(10 nm)/Ag(100 nm), LiF:Al alloy (10 nm)/Ag, Al(10 nm)/LiF (0.5 nm)/Ag, Al (10 nm)/Ag, and Li(2 nm)/Al (10 nm)/Ag were used as cathodes. LiF:Al alloy cathode was fabricated by codeposition of LiF and Al. The fabricated devices were sealed using glass plates with epoxy resin in an argon-filled glove box. The active areas were 4 or 0.3 mm².

The current density-voltage-luminance (J-V-L) characteristics were recorded using a 2000 multimeter (Keithley), a regulated DC power supply (Kenwood PW 36–1.5AD) and a luminance meter (Minolta LS-100). Transient EL was measured by applying square-wave voltage pulses generated by an HP8114A source (Agilent). The optical pulse was observed using a photomultiplier tube detector (Hamamatsu Photonics). The EL, voltage and current waves were monitored by a Lecroy104Xi oscilloscope. All of the measurements were carried out at room temperature.

3. Results and Discussion

First, we investigated the optimal concentration of LiF for the OLEDs of ITO/ α -NPD (50nm)/Alq₃ (50nm)/LiF:Al alloy (LiF concentration : 0–50 vol%) (10nm)/Ag (100 nm). Figure 1(a) shows the maximum luminance and maximum current efficiency of OLEDs with various LiF:Al alloy cathodes for different concentration of LiF. The maximum luminance and maximum current efficiency of an OLED with LiF nondoped cathode were approximately 4,000 cd/m² and 1.3 cd/A, respectively. On the other hand, for the devices with various LiF:Al alloy cathodes, the device performance was markedly increased. A device with the LiF:Al (1:10) alloy cathode (concentration of LiF: 10 vol%) showed the highest maximum current efficiency of 4 cd/A. The maximum current efficiency decreased with increasing concentration of LiF over 10 vol%. It is clear that the increase of insulation of LiF in the LiF:Al alloy cathode results in the decrease of electron injection into organic layer. Therefore, we selected the OLED with LiF:Al (1:10) alloy cathode for further investigation.

Figure 1(b) shows J-V-L characteristics of devices with various cathodes. The emission characteristics of the device with LiF:Al alloy cathode were almost the same as those with LiF/Al cathode. It indicates that LiF:Al alloy cathode can achieve efficient electron injection. Compared with the OLEDs with Al/LiF and Al cathodes, the maximum luminance and current efficiency of the OLED with Al/LiF were clearly increased. According to K. Seki et al. [1,2], for the deposition of LiF on the Alq₃ layer, the dipole of LiF affects bending the band of the Alq₃ layer to reduce the injection barrier. These results suggested that the formed dipole of LiF near the interface of Alq₃/Al leads to achieve the efficient electron injection.

Next, we investigated the transient characteristics of OLEDs with the active area of 0.3 mm² under the application of rectangular-shaped voltages (Frequency: 100 Hz, Pulse width: 100 μ s). Figure 2(a) shows transient EL characteristics of OLEDs with LiF/Al, LiF:Al alloy, Al/LiF, Al, and Li/Al cathodes. Rise (Decay) time is defined as the time required to change the optical response from 10 (90) to 90% (10%) of its total intensity change. For all devices, the decay time was almost the same because the time constant

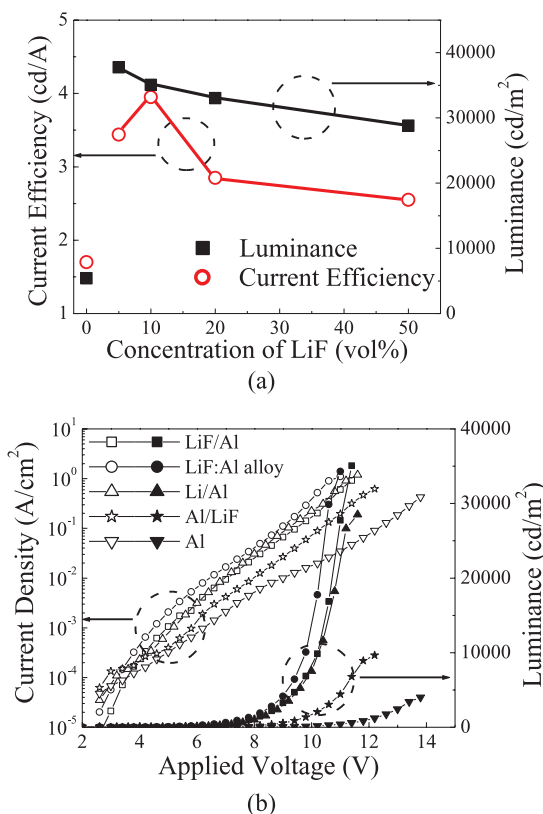


Figure 1. (a) The maximum luminance and the maximum current efficiency of OLEDs with various LiF:Al alloy cathodes for different concentration of LiF. (b) J-V-L characteristics of OLEDs with LiF(0.5 nm)/Al(10 nm), LiF:Al(1:10) alloy (10 nm), Li(2 nm)/Al(10 nm), Al(10 nm)/LiF(0.5 nm), and Al(10 nm) cathodes, respectively.

of resistance and capacitance, and the delayed EL from residual carriers were mainly contributing factors. By contrast, for all devices except the devices with LiF/Al and Li/Al cathodes, the rise time decreased with increasing the current density in Fig. 2(a). For a device with LiF/Al cathode, the waveform rounding of transient EL was observed as shown in Fig. 2(b). Therefore, compared with the devices with LiF:Al alloy and LiF/Al cathodes, the rise time of a device with LiF:Al alloy cathode was shorter than that of a device with LiF/Al cathode at high current density. This is because the waveform rounding of current density occurred for a device with LiF/Al cathode as shown in Fig. 2(b).

In Fig. 3(a), the waveform rounding of transient EL signals of both the devices with Al/LiF and Al cathodes was not observed, and were almost the same as that of a device with LiF:Al alloy cathode. On the other hand, the waveform rounding of transient EL signals of devices with LiF/Al and Li/Al cathodes occurred similarly, as shown in Fig. 3(b). These results suggested that LiF or Li at the interface of Alq₃ and Al is attributed to the waveform rounding.

Dissociation of Li from deposited LiF at the interface of Alq₃ layer and Al layer has been reported [3,4,5]. On the devices with LiF/Al and Li/Al cathodes, the residual

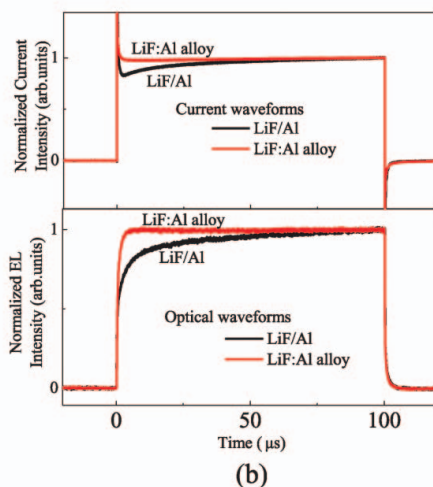
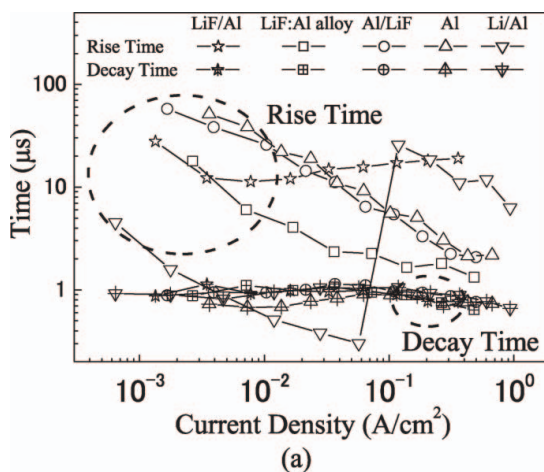


Figure 2. (a) The applied pulse voltage dependence of the rise and decay times for the devices with LiF/Al, LiF:Al (1:10) alloy, Li/Al, Al/LiF, and Al cathodes and (b) typical transient current densities and EL signals of devices with LiF/Al and LiF:Al alloy cathodes at current density of 0.1 A/cm².

charge carriers in Alq₃ near the interface increased because the diffusion of deposited Li or dissociated Li from LiF into organic layer occurred and the diffused Li acted as trap. Thus, the waveform rounding of transient response curves of devices with Li/Al and LiF/Al cathodes occurred. The reasons why response waveforms of a device with LiF:Al alloy cathode improved and achieved shorter transient electroluminescence are because Al acted as the blocking layer of diffusion of LiF and less LiF existed at the interface, compared with a device with LiF/Al cathode owing to codeposition of LiF and Al.

Rise time of a device with Li/Al cathode did not decrease with increasing current density. This is because the overshoot of transient EL signal leads to the shorter rise time as shown in Fig. 3(b). This overshoot is due to any capacitance in this device because it vanished when applied positive base voltage as shown in the inset of Fig. 3(b).

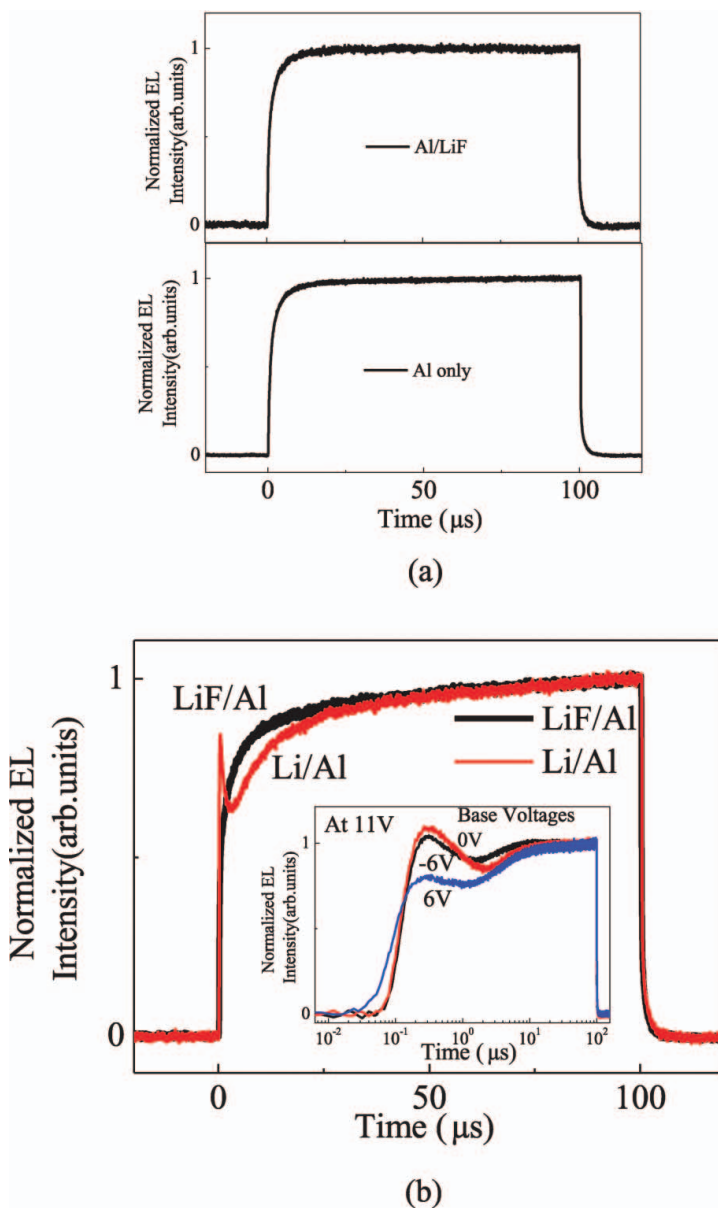


Figure 3. Typical transient EL signals of the devices with (a) Al/LiF and Al cathodes, and (b) LiF/Al and Li/Al cathodes at 0.1 A/cm^2 . Inset of Fig. 3(b): the transient EL signals of a device with Li/Al cathode when applied three base voltages of -6, 0, +6 V at the applied voltage of 11 V.

4. Conclusions

In this study, we investigated the effects of LiF on the emission characteristics and transient electroluminescence of Alq_3 -based OLEDs with LiF/Al cathode and LiF:Al alloy cathode, respectively. The maximum luminance of $3.4 \times 10^4 \text{ cd/m}^2$ and the maximum current

efficiency of 4 cd/A were achieved for a device with LiF:Al (volume ratio 1:10) alloy cathode, and these emission characteristics were almost the same as those of a device with LiF/Al cathode which is a typical structure of cathode of OLEDs. Furthermore, under application of rectangular-shaped voltage, a device with LiF:Al alloy cathode achieved shorter rise time than that of a device with LiF/Al cathode at high current density. At 0.1 A/cm², waveform rounding of transient current and optical curves of a device with LiF/Al cathode was observed due to the residual charge carriers in organic layer owing to the diffusion of dissociated Li from LiF into organic layer, whereas, the waveform rounding of transient curves of a device with LiF:Al alloy cathode was not observed.

Acknowledgements

Part of this work was supported by Grant-in-Aid for Scientific Research, from the Ministry of Education, Culture, Sports, Science and Technology, Japan. This research was also partially supported financially by a grant for the Osaka University Global COE Program, "Center for Electronic Devices Innovation", and a Grant in Aid of Special Coordination Funds for Promoting Science and Technology.

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