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# Molecular Crystals and Liquid Crystals

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# IMPROVED CHARACTERISTICS OF ORGANIC LIGHT EMITTING DIODES WITH COEVAPORATED AL-ALKALINE METAL CATHODE

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#### IMPROVED CHARACTERISTICS OF ORGANIC LIGHT EMITTING DIODES WITH COEVAPORATED AL-ALKALINE METAL CATHODE

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The cathodes using co-evaporated Al-CsF, Al-KF, and Al-NaF composites are adopted to enhance the electrical and the optical properties of organic light emitting diodes (OLEDs). The hole transport layer (HTL), 50 nm thick N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) and the electron transport layer (ETL), 50 nm thick tris(8-hydroxy-quinoline) aluminum (Alq<sub>3</sub>) were deposited under the base pressure of  $1.6\times10^{-6}$  Torr. In depositing Al-CsF, Al-KF, and Al-NaF composite contacts, the mass ratio of alkaline metals is varied  $1\sim10$  wt%. OLEDs with alkaline metal-doped cathode have luminance of about 35,000 cd/ $m^2$ , and external quantum efficiency of about 1.35%. Alkaline metal tends to diffuse into organic layer and then form AlF<sub>3</sub> with the alkaline metal-doped surface region.

Keywords: alkaline metal; composite contacts; current-voltage characteristics; organic light emitting diode

#### INTRODUCTION

Since C. W. Tang and S. Van Slyke showed highly efficient and bright electroluminescence using multi-layer organic thin films in 1987 [1], OLEDs have been paid much attention due to wide viewing angle, high luminous efficiency and possibility of very thin devices [2]. It is one of the most critical research issues to obtain appropriate device reliability and efficiency. The low work function materials have been used as a cathode to improve the electron injection into OLEDs [1–5]. However, OLEDs using low work function metal cathodes exhibited inferior stability [6]. The coevaporated cathode structure is applied to enhance OLED performances

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and stability. Alkaline metals (CsF, KF, and NaF) and Al were used. Especially, CsF can be thermally deposited at lower temperature compared with LiF [6] so that the CsF could prevent the heat exposure of the organic layers during the evaporation of the metals [7]. In this paper, the effects of co-evaporated cathodes on the performances of OLEDs were investigated.

#### **EXPERIMENTAL DETAILS**

Glass substrates coated with ITO(the sheet resistance of ITO,  $20\,\Omega/\Box$ ) were cleaned in an ultrasonic bath of acetone, followed by isopropyl alcohol, and then D. I. water. The fabricated devices have a double layer structure, which consists of HTL and ETL. The 50 nm thick TPD and 50 nm thick Alq<sub>3</sub> were deposited under the base pressure of  $1.6\times10^{-6}$  Torr. In depositing the Al-CsF, Al-KF, or Al-NaF composite contacts, the mass ratio of alkaline metal was varied from 1 to 10 wt%. Different type devices were also fabricated to study decomposition of CsF during thermal evaporation and find the order of improved conductivity and trap number. The thickness of Alq<sub>3</sub> layer is  $40\sim50\,\mathrm{nm}$  between the electrodes.

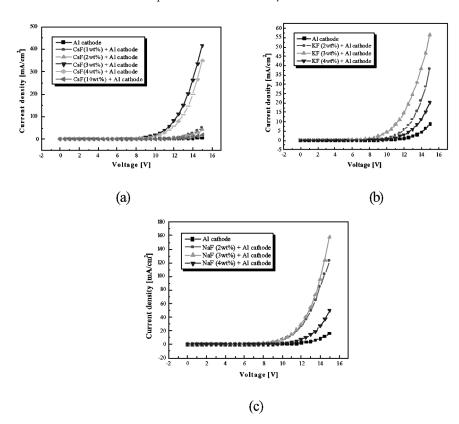
#### RESULTS AND DISCUSSIONS

#### 1. J-V and L-V Characteristics

The current density and luminance intensity of the devices with different cathode compositions are shown in Figure 1 and Figure 2. The devices with co-evaporated cathodes show superior performance to the devices based on Al cathode. At all the devices with co-evaporated cathodes, low operation voltages were obtained, which is attributed to the efficient charge injections. It has been reported that Cs, K, and Na is evaporated onto  $Alq_3$  and diffused uniformly into  $Alq_3$  so that the Fermi level moves towards the LUMO level of  $Alq_3$  [8]. The device with Al-CsF(3 wt%) cathode has the luminance of  $7,000 \, \text{cd/m}^2$  at  $100 \, \text{mA/cm}^2$ .

# 2. Power Efficiency and External Quantum Efficiency

The power efficiencies of the devices with the different cathode compositions are shown in Figure 3. Higher power efficiencies for the devices with co-evaporated cathodes compared with the devices with Al cathode are observed. Using the Al-CsF composite cathode, the highest external quantum efficiency of 1.35% at 11 V was achieved. It is likely that CsF



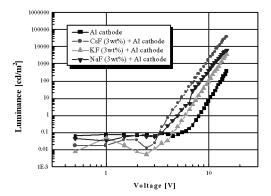
**FIGURE 1** The current density-voltage characteristics of organic light-emitting diodes with co-evaporated cathode (a) using Al-CsF cathode (b) using Al-KF cathode (c) using Al-NaF cathode.

reacts with Al and forms  $AlF_3$  compound, and cesium acts as a dopant to build up cesium-doped surface region, these results lead to improve electron injections and hence device performance [9].

## 3. Carrier Injection Process

Fowler-Nordheim tunneling equation is used to calculate the effective barrier height at the interface between cathode and organic layer. Fowler-Nordheim tunneling equation is expressed as

$$J = \frac{A^* T^2}{\phi_B} \left(\frac{qF}{\alpha kT}\right)^2 \exp\left[-\frac{2\alpha \phi_B^{3/2}}{3qF}\right] \tag{1}$$

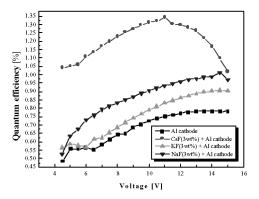


**FIGURE 2** The luminance-voltage characteristics of organic light-emitting diodes with co-evaporated cathode.

CsF molecules undergo decomposition when contacted with Al atoms. The energy level of organic layer depends on the diffusion of cesium into the organic layer. The electron from fluorine was transferred back to the cesium atom and an  $Alq_3$  gets transferred extra charge [8]. As a result, the contact formed at the organic/cathode interface becomes ohmic [7]. The turn-on voltages and the barrier heights of the devices with Al-CsF cathodes are detailed in Table 1.

# 4. J-V Characteristic of Electron-only Devices

To study decomposition of CsF during thermal evaporation, the devices with different structures were fabricated; Mg:Ag/Alq<sub>3</sub>(40 nm)/Al-CsF (3 wt%)(D1) and Al-CsF(3 wt%) /Alq<sub>3</sub>(40 nm)/Mg:Ag(D2).



**FIGURE 3** The external quantum efficiency of organic light-emitting diodes with co-evaporated cathode.

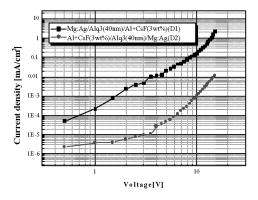
TABLE 1	Calculated	Tunneling Onse	t Voltage and	Barrier	Height for an	ITO/TPD/
Alq <sub>3</sub> /Al+C	sF Using Ed	ą. (1)				

	Tunneling onset voltage [V]	Barrier height [eV]	
Al cathode	8.5	0.26	
Al+CsF(1 wt%)	7.5	0.25	
Al+CsF(2 wt%)	7.0	0.28	
Al+CsF(3 wt%)	6.5	0.22	
Al+CsF(4 wt%)	6.0	0.24	
Al+CsF(10 wt%)	8	0.26	

The current densities of the electron-only devices with different stacking order are shown in Figure 4. It is shown that the diode D1 exhibited the enhanced electron injection into the  $Alq_3$  layer at all bias. The D1 device shows about 100 times larger current density than that of the D2 device at 10 V. During CsF evaporated onto organic layers, cesium can diffuse into organic layer. Consequently, the ohmic contact was formed between an organic layer and metal cathode, where cesium donates free electrons into the  $Alq_3$  layer. Therefore it can be stated that the penetration and the diffusion of cesium enhanced the electron injection [7–8].

### 5. The Characteristics of Electrical Properties

To observe in order to indentify conductivity and make a comparison between trap number, the devices with different structures were fabricated; Mg:Ag/Alq<sub>3</sub>(50 nm)/Al(D3) and Mg:Ag /Alq<sub>3</sub>(50 nm)/Al-CsF (3 wt%)(D4). The Figure 5 is shown I-V characteristics of the device with



**FIGURE 4** The current density of D1 and D2 device with co-evaporated electrode.

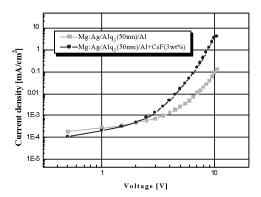


FIGURE 5 The current density of D3 and D4 devices.

different cathode. CsF molecules undergo decomposition when contacted Al atoms. The energy level of organic layer depends on the diffusion of cesium into the organic layer. So, the D4 is 30 times higher current density than the D3. The measured conductivity of D3 is 0.26  $\Omega^{-1}$ cm<sup>-1</sup> and that of D4 is 34.84  $\Omega^{-1}$ cm<sup>-1</sup> from Figure 5. The conductivity of D4 is about 130 times higher than that of D3. And the equation finding the number of trap is expressed as

$$V_{\Omega} = \frac{qd^2H_b'}{\varepsilon} \left(\frac{n_0}{N_c}\right)^{1/l} \left(\frac{l+1}{1}\right) (l+1^{2l+1})^{l+1/l}$$
 (2)

where  $H_b'$  is the density of traps and  $n_0$  is the density of thermally generated free carriers and  $N_c$  is LUMO density of states. The number of D3's trap is  $3.27 \times 10^{19}$  cm<sup>-3</sup> and that of D4's trap is  $8.66 \times 10^{18}$  cm<sup>-3</sup>. The conduction of the D4 is better than that of the D3, because Cs atoms diffuse Alq<sub>3</sub> and fill the trap of Alq<sub>3</sub>. The device with co-evaporated cathode, as a result, the contact formed at the organic/cathode interface becomes ohmic, was enhanced electrical properties [7].

#### SUMMARY

In this work, we have investigated the electrical properties of the OLEDs with the co-evaporated Al-alkaline metal cathodes. Upon the investigations, it is revealed that co-evaporated cathode structures enhance the electron injections and luminance intensity, and reduce the driving voltage, which is due to the fact that cesium ions tend to diffuse into organic layer and form  $AlF_3$  by reacting with Al and the alkaline metal forms a sort of metal-doped surface region. Therefore, the contact becomes nearly ohmic at the

organic/metal interface [8]. Consequently, it has been confirmed that co-evaporated cathode is suitable for enhancing the OLED performances.

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