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Improving the efficiency of organic light emitting devices by using co-host electron transport layer

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Abstract

By engineering a new cohosting system of tris(8-hydroxyquinoline) and 4,7-diphenyl-1,10-phenanthroline in the electron transport layer, the current efficiency of the organic light emitting diode is improved by 34% to 4.3 cd/A as compared to the device with a single host of Alq₃ as the electron transport layer. The maximum luminance is over 16000 cd/m² at the bias of 22V and the current of 475mA/cm², which is ~73% higher than the single host Alq₃ device without optimizing the layer thickness. The reasons for the improvement will be investigated. The results strongly indicate that the knowledge of bulk conductivity engineering of organic n-type transporters shows practical significance in OLED applications.

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Keywords: *Tris* (8-hydroxyquinoline) aluminum, 1,10-phenanthroline, mobility, electron transport, co-host

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Introduction

Tris (8-hydroxyquinoline) aluminum (Alq_3) has been commonly used as an organic light-emitting host as well as electron transport layer (ETL) in organic light-emitting diodes (OLEDs). However, due to its lower electron mobility of $10^{-6} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ [1-3], Alq_3 is one of the key sources responsible for the undesirable predominant voltage drop in Alq_3 -based devices [4]. Furthermore, the operation lifetime of Alq_3 based devices is reduced due to the unbalanced charge accumulation originated from an excessive hole injection into the Alq_3 layer [5]. Extra amount of holes also generate non-emissive cationic species in the Alq_3 and result in the formation of dark spots [6]. Hence, enhancement of electron conduction in the ETL of OLEDs is a critical approach to both lowering the driving voltage and improving the operation lifetime. Recently, although some electron transporting materials such as phenanthroline and oxadiazole compounds were suggested for replacing the Alq_3 as the ETL due to their improved drift electron mobility [7-10], the device reliability including film stability and thermal endurance have not yet fulfilled the requirement of commercial display application [10-11]. The highly reactive metal (e.g. Li) doping reported by Kido *et al* [12] is an alternative means to enhance the device electron conduction, although photoluminescence quenching occurs if the doped region exceeds 30 nm.

In our study, we establish an organic based co-host composite to replace the traditional single host ETL. The results conclude that our approach improves the electron conductivity on the ETL without affecting the film stability, resulting in an improved current efficiency of about 34 % through employing a ET composite of phenanthroline :

hydroxyquinoline. It is important to note that 4,7-diphenyl-1,10-phenanthroline (BPhen) is chosen for mixing with Alq₃ to form the co-host ETL since it possesses a considerable high electron mobility ($\sim 5 \times 10^{-4} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) among the electron-dominant materials [10]. This report focuses mainly on the device aspect by applying the knowledge of bulk conduction presented previously. The composition of the BPhen:Alq₃ co-host electron transporter based devices will be investigated.

Experimental

The indium-tin-oxide (ITO) substrates with a size of $25 \times 25 \text{ mm}^2$ and resistivity of 80 ohm/square were cleaned prior to loading into the evaporation chamber through scrubbing by detergent and soaking into de-ionized (DI) water for 10min in each step. The evaporation chamber is operated at $\sim 10^{-7}$ Torr. The substrates were then immersed into ultrasonic bath of DI water, ethanol and acetone for 20 min for each solvent. The solvent cleaned ITOs were further treated in UV ozone for 20 min.

The organic materials used in this study were shown in Fig. 1. They were Alq₃ acted as ETL and light emitting layer (EML), *N,N'*-bis(1-naphthyl)-*N,N'*-diphenyl-1,1'-biphenyl-4,4'-diamine (NPD) as a host transport layer (HTL), BPhen as ETL, and copper phthalocyanine (CuPc) for stabilizing the ITO surface and LiF as the electron injection layer (EIL). The materials were purified by gradient sublimation prior to thin-film coating when BPhen was used without further purification. The deposition rate for CuPc, Alq₃, NPD, BPhen was typically 1.0-2.0 Å/s. The deposition rates of LiF and Al were 0.1 Å/s and 10-15 Å/s, respectively. Film thickness was monitored *in situ* using the quartz

crystal monitor and *ex situ* by a stylus profilometer (Tencor α -step 500). The emission area of devices is 4.57 mm².

To investigate the contributions of the co-host electron transport composite, three devices containing different ETLs have been prepared: (i) device **A** – Alq₃; (ii) device **B** – BPhen; and (iii) device **C** – *n* wt% BPhen-doped Alq₃. The undoped EML device has a structure: ITO / CuPc (15 nm) / NPD (45 nm) / Alq₃ (EML, 20 nm) / ETL (60 nm) / LiF (0.5 nm) / Al as shown in Fig. 1(b). The devices were characterized in room ambient without encapsulation. The current density-voltage-luminance (J-V-L) characteristics of the devices were measured with International Light IL1400A Photometer and a programmable Keithley source meter 2400.

Results and discussion

Fig. 2 shows the luminous efficiency of undoped devices with various compositions of ET hosts incorporating BPhen and Alq₃. In the typical device using Alq₃ as the ETL (device **A**), the efficiency is ~3 cd/A at 20 mA/cm², similar to the reported values [13]. Replacing ETL by BPhen (device **B**), the efficiency is significantly enhanced by ~34 %, to 4.3 cd/A. The time-of-flight results indicate that under an applied electric field of ~1 MV/cm, the electron mobility of Alq₃ has a value of ~10⁻⁶ cm²V⁻¹s⁻¹ [1-3] while that of BPhen exhibits a much higher electron mobility of 5 × 10⁻⁴ cm²V⁻¹s⁻¹ [10]. With higher electron drift mobility, BPhen-type ETL possesses better electron conduction and thus a higher charge recombination in the Alq₃ based light-emitting devices. As a result, electron drift mobility difference between BPhen and Alq₃ can be

seen as responsible for the dramatic change in the device's luminous efficiency. In addition, when the BPhen concentration is increased in the BPhen:Alq₃ co-host, the efficiency increases. It suggests that more electrons are transferred into the EML which will be discussed in detail later. Consequently, BPhen:Alq₃ forms a promising electron transporting layer in Alq₃-based OLEDs.

It is worth to note that while the device can still benefit from the co-host approach to maximize the efficiency at the BPhen composition of > 50 wt%. It would encounter the problem of re-crystallization with its ETL containing a pristine or heavily-doped Bphen [14]. Therefore, in the following discussion, we will focus on improvement of the device performance by employing 50 wt% BPhen:Alq₃ co-host structure.

Two testing devices with difference in ETL only: device **A** – a single Alq₃ host and device **C** – a 50 wt% BPhen:Alq₃ co-host have been investigated. Their device performances are shown in Fig. 3. As illustrated in the current-voltage (I-V) curves of Fig. 3(a), the driving voltage required for the device to operate at a current of 20 mA/cm² is reduced from 14.3 to 10.8 V by introducing a BPhen:Alq₃ co-host ETL. Meanwhile, the corresponding luminance increases from 580cd/m² to 760 cd/m², as shown in Fig. 3(b). Without thickness optimization, the maximum luminance of the co-host device is over 16,000 cd/m² at a current of 475 mA/cm² and bias of 22V; this is ~73 % higher than the single Alq₃ host device.

Besides the merit of the high drift mobility of BPhen, the noticeable achievement of the BPhen:Alq₃ co-host is also contributed by the similar electron hopping along their lowest unoccupied molecular orbitals (LUMOs). The LUMO of Alq₃ and BPhen are similar with a value of 3eV [15]. Therefore, it is believed that electrons hop freely at both Alq₃ and BPhen in the co-host device as shown in Fig. 4 while in a single host device, electrons hop merely along the LUMO in Alq₃. Moreover, no shifting to higher operating voltage in the I-V characteristics has been observed in the devices with different compositions of BPhen:Alq₃. This also implies that electron trapping due to the introduction of BPhen does not occur when the BPhen concentration is beyond 10 wt%.

In addition, the transport of electrons can be adjusted by varying the weight ratio of BPhen:Alq₃ in the mixed co-host ETL layer. By increasing the BPhen:Alq₃ weight ratio, the probability of hopping via the LUMO of BPhen will be increased, resulting in the increase of electron mobility in co-host ETL. Our results show that the current efficiency improves through increasing the weight ratio of BPhen, although the high concentration of Bphen causes the trend of crystallization of BPhen [14] when the weight ratio increases beyond the 50%BPhen : 50%Alq₃. Furthermore, the largely negative highest occupied molecular orbital (HOMO) bands of BPhen with a value of ~6.4eV in the co-host ETL matrix effectively blocks the holes from penetrating through BPhen:Alq₃ co-host ETL, and confines excitons in EML. Consequently, the performance of the devices with co-host ETL can also be improved as compared to that of conventional single Alq₃ devices through better balance of the number of holes and electrons and confining the excitons within the EML.

Conclusion

A new method of co-hosting hydroxyquinoline with phenanthroline derivative for improving the electron mobility of ETL has been introduced in this paper. Thanks to (a) the superior electron mobility of BPhen, (b) free electron hopping in ETL due to similar LUMO value of both Alq₃ and BPhen, and (c) high HOMO value of BPhen for reducing the hole injection into the ETL, the current efficiency of the cohost device is increased by 34% to 4.3 cd/A as compared to the conventional structure with single host of Alq₃ when the weight ratio of BPhen:Alq₃ is 50:50. The further increment of BPhen content will cause the trend of crystallization of BPhen at high concentration. Without any optimization of the layer thickness, the maximum luminance of the co-host device is over 16,000 cd/m² at a current of 475 mA/cm² and bias of 22V, which is ~73 % higher than the single Alq₃ host devices.

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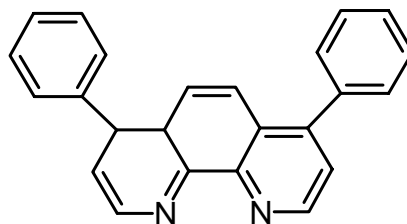
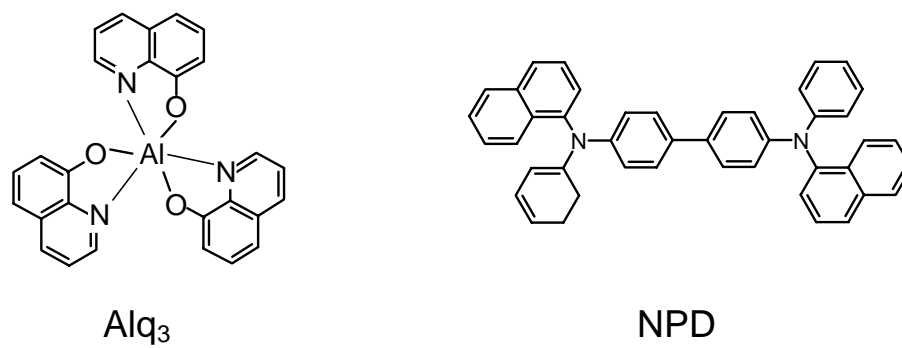
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Fig. 1 (a) Molecular structures of Alq₃, BPhen, and NPD. (b) Device configuration: ITO /CuPc(15nm)/NPD (45nm)/Alq₃(20nm)/ETL (60nm)/LiF (0.5nm)/Al (150nm).

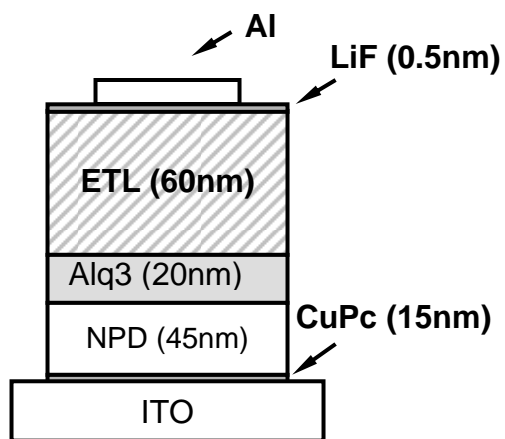
Fig. 2 Comparison of luminous efficiency of undoped devices driven at 20 mA / cm². The ETL consists of a composite layer of BPhen and Alq₃. The total thickness of Alq₃ is 700 nm.

Fig. 3 Plots of (a) Current-voltage and (b) Luminance-current of devices without doping the EML: Device A (close circle) contains a single ETL. Device B (close triangle) contains a co-host ETL (50 wt% BPhen mixed Alq₃).

Fig. 4 Energy diagram of (a) undoped and (b) 50 wt% Bphen:Alq₃ cohost devices. Shaded area indicates the Alq₃ based EML.



(a)



ETL: BPhen, BPhen:Alq₃, and Alq₃

(b)

Fig. 1

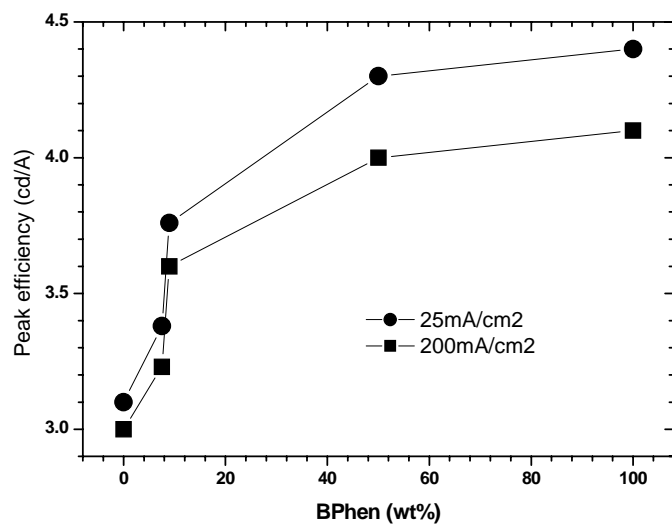


Fig. 2

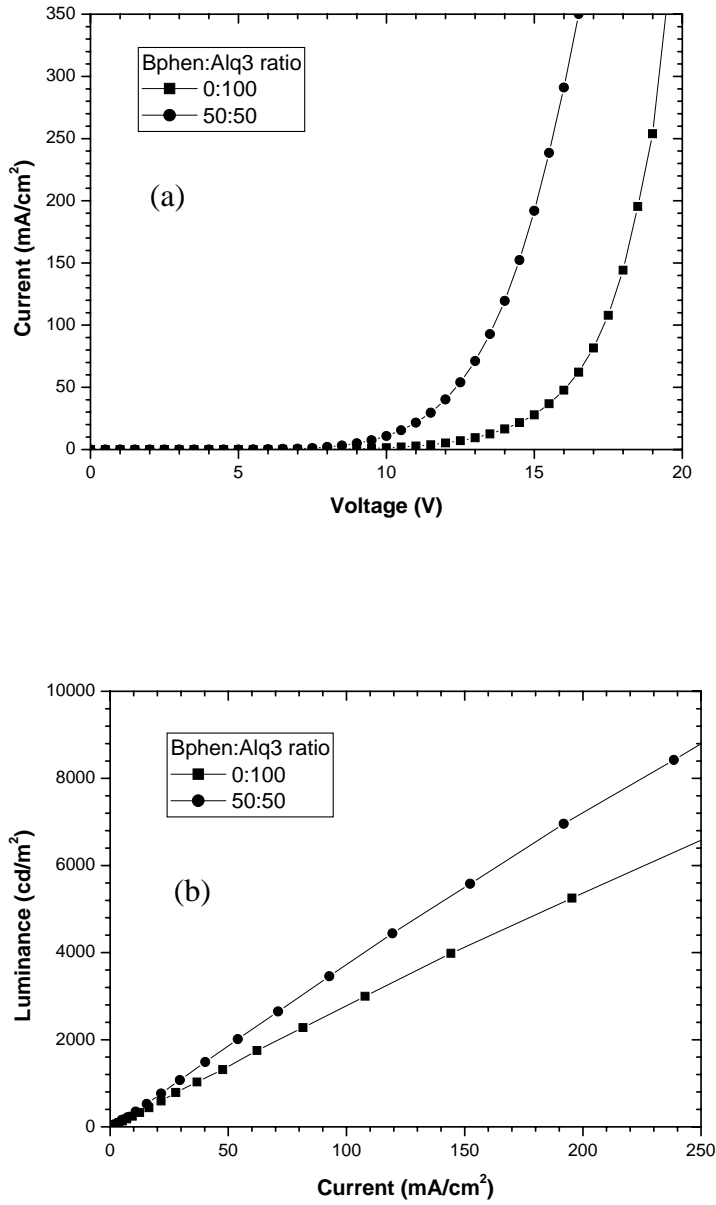


Fig. 3

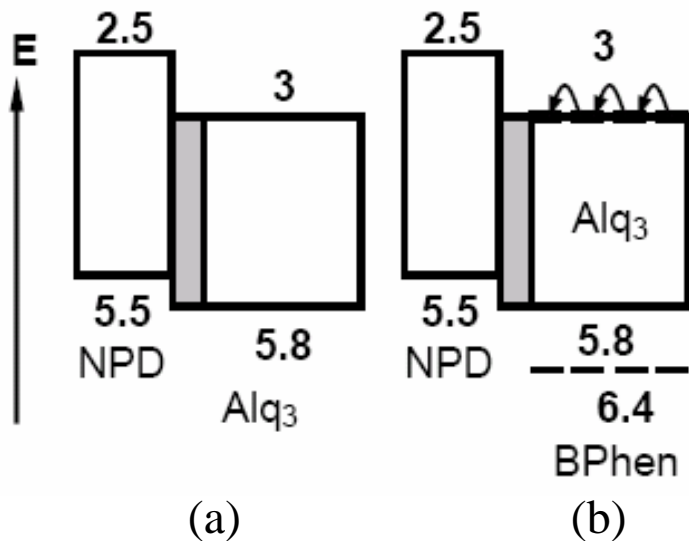


Fig. 4