

# Blending effect on small molecule based OLED

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Organic light-emitting diodes (OLEDs) were fabricated containing guest molecule of Tris(8-hydroxyquinoline) aluminium ( $\text{Alq}_3$ ) blend with host molecules of N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) and 2-(4-biphenyl)-5-phenyl-1,3,4-oxadiazole (PBD) small molecules. Optical, photoluminescence (PL), electroluminescence (EL) and impedance properties were investigated with respect to blend systems. Optical energy gap and PL intensity obtained increased in blend systems attributed to high energy transfer from host to guest molecules. Luminance and current efficiency were enhanced for blended OLEDs as compared to that of pure  $\text{Alq}_3$ , related to high accumulation of charge carrier and exciton recombination in guest. Impedance spectra show conductivity of OLEDs was improved by blending technique.

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**Keywords:** OLED, Small molecules, Optical, Luminescence, Electrical properties

## 1. Introduction

Since Tang and VanSlyke have reported the first organic light emitting diode (OLED) using a small molecule of  $\text{Alq}_3$  [1], many approaches have been carried out to improve the performance of OLED. Blending method has been intensively used in fabrication of solution-processed based OLED [2-8] in order to improve the performance of an OLED such as to modify the turn on voltage [9, 10], current and luminance efficiency [11]. Usually, an electron transporting material is blended with a hole transporting material to promote a more balance carrier injection and transportation in the organic layer [12,13].

In this work, we introduce a new ternary system comprised of a blend of TPD and PBD host-materials with  $\text{Alq}_3$  guest-molecules for single layer OLED. Interestingly, the devices were prepared in open air environment to simplify the fabrication method towards a simpler and cheaper OLED production. Significant improvement on the performance of OLED was investigated upon blending.

## 2. Experimental

$\text{Alq}_3$ , TPD and PBD which used as organic blend of the emissive layer were purchased from Sigma Aldrich. Fig. 1 shows the molecular structure of the materials. The organic blend solution was prepared by dissolving the  $\text{Alq}_3$ , TPD: $\text{Alq}_3$  and TPD:PBD: $\text{Alq}_3$  in chloroform with the weight ratio of 1, 1:1 and 1:1:1, respectively. Single layer OLED was constructed as ITO/PEDOT:PSS/Blend/Al where ITO and PEDOT:PSS acted as the bulk anode layer,  $\text{Alq}_3$ -organic blend as the emissive layer and aluminium (Al) as the metal cathode. Glass coated with ITO layer was patterned and ultrasonically cleaned in Decon<sup>TM</sup>, followed by deionized

water, acetone and isopropanol, and finally dried with nitrogen purge. Buffer layer of PEDOT:PSS was then spin-coated onto the substrate. Thereafter, an emissive layer with thickness of 80 nm was deposited by the spin-coating method. Finally, a 100 nm thick of Al electrode was thermally deposited to define an active area of  $2.6 \times 10^{-5} \text{ m}^2$ . Fig. 2 shows the structure of the fabricated device.

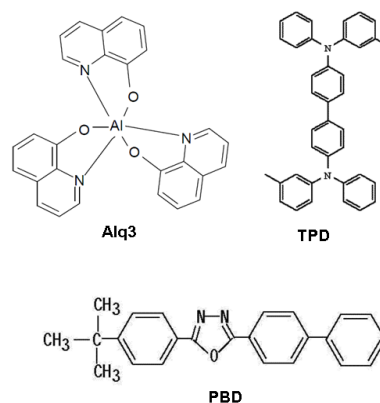


Fig. 1. Molecular structure of  $\text{Alq}_3$ , TPD and PBD.

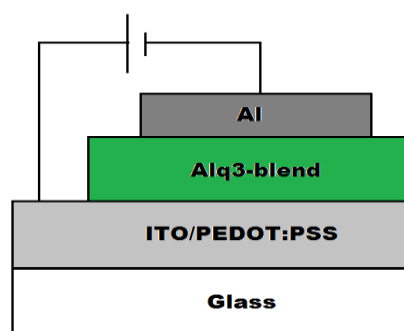


Fig. 2. OLED structure used in this work.

Optical absorption and photoluminescence characteristics were obtained by UV-Vis-NIR (Jasco-V50) and luminescence spectrometer (Perkin Elmer LS 50B), respectively. The thickness of the thin film device was measured by profilometer (KLA Tencor 6). Electroluminescence characteristics were measured using chroma meter CS-200 (Konica Minolta) powered by source measurement unit (Keithley 2400). Impedance measurements were carried out using a Solatron 1260 Impedance analyzer. All the devices were characterized in open air environment without encapsulation.

### 3. Results and discussion

#### 3.1 Optical and photoluminescence properties of blend thin film

Fig. 3 shows normalized UV-Vis absorption spectra of TPD and PBD in thin films form taken at room temperature environment. All materials have their own maximum absorption peak. The most absorption peak for TPD and PBD are around 354, 292 and 392 nm, respectively. Fig. 4 shows the normalized UV-Vis absorption spectra for the different blend systems measured in the thin film form. The maximum absorption peak for the pure  $\text{Alq}_3$  is obtained at 386 nm while for TPD: $\text{Alq}_3$  and TPD:PBD: $\text{Alq}_3$  blend thin films, the maximum absorption peaks are observed shift towards shorter wavelength of 354 and 308 nm, respectively. For the TPD:PBD: $\text{Alq}_3$  blend, the absorption peak of oxadiazole moieties is observed in the range of 275–350 nm as similar to the reported one by Xu et al. [14]. This indicates that the oxygen and nitrogen atoms in the oxadiazole moiety are strongly bonded and do not form a coordinate bond with the aluminium from  $\text{Alq}_3$  molecule. It is suggested that the absorption peak of the blend system is related to the strong absorption intensity of the host-molecules that overlaps with the absorption spectra of  $\text{Alq}_3$  without molecular interaction.

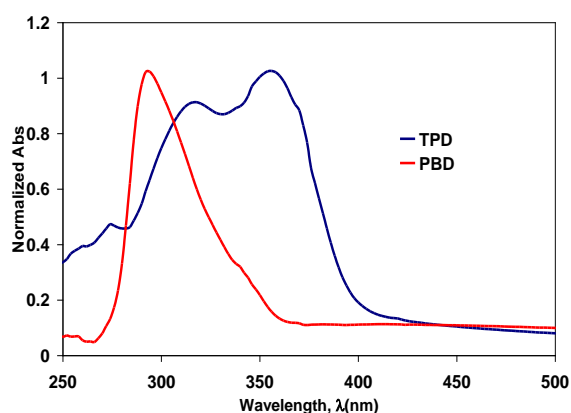


Fig. 3. Normalized UV-Vis absorption of TPD and PBD in thin film form.

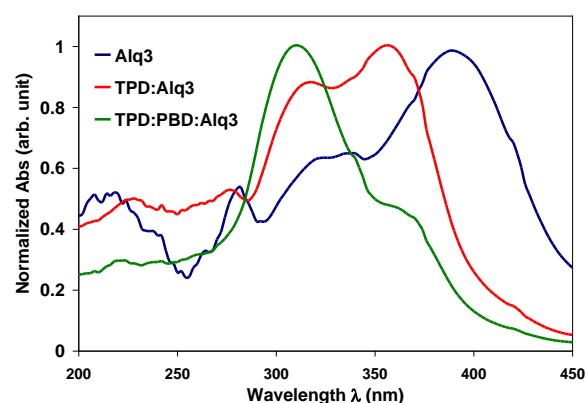


Fig. 4. Normalized UV-Vis absorption of the blend thin film.

Based on this Fig. 5 and 6 shows the Tauc plot of TPD and PBD, and the blend thin film, respectively. From the Tauc plot, the energy gap of TPD, PBD,  $\text{Alq}_3$ , TPD: $\text{Alq}_3$  and TPD:PBD: $\text{Alq}_3$  thin films are obtained to be 3.12 eV, 3.50 eV, 2.79 eV, 3.05 eV and 3.15 eV, respectively. The energy band of TPD and PBD are higher compared to the  $\text{Alq}_3$  as their absorption spectra are in the lower wavelength region. These results shows that the energy gap increased with increase in number of element in such a blend which related to high band gap of TPD and PBD molecules that overlap with energy gap of  $\text{Alq}_3$ . Based on the obtained energy gap, energy diagram of OLED blend is illustrated in Fig. 7.

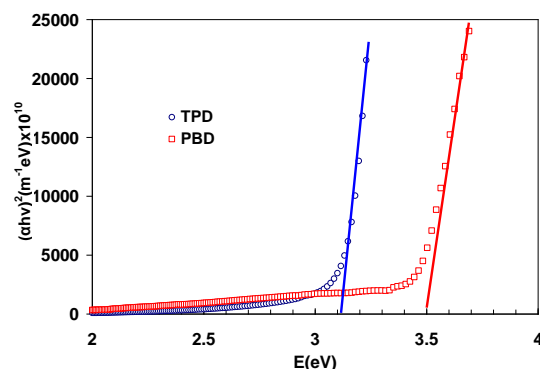


Fig. 5. Tauc plot of TPD and PBD.

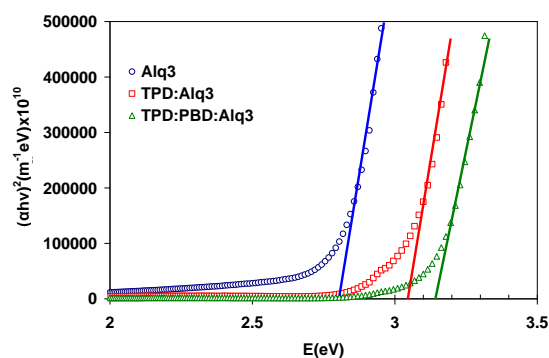


Fig. 6. Tauc plot of the blend thin film.

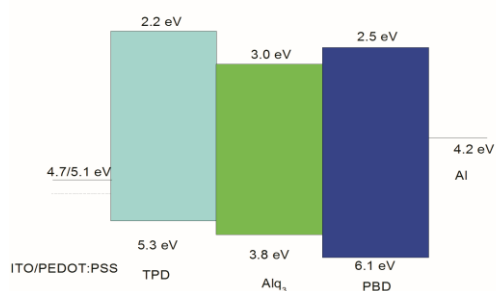


Fig. 7. Energy band diagram of the blend OLED.

Fig. 8 shows normalized PL spectra of the TPD and PBD in thin film form. The PL peak for TPD and PBD was centred at 415 and 374, respectively. Fig. 9, shows PL spectra of the blend thin film. It is shown that the intensity increases with the increase in the host-molecules added to the system. However, the PL peaks position for such blend remains at the almost same as the Alq<sub>3</sub> one. Please be noted that these samples have been excited at 280 nm, which covers the highest absorption energy of TPD and PBD. These PL results can be related to the efficient energy transfer from hosts to guest molecule. Emission only occurs at Alq<sub>3</sub> guest, and not TPD and PBD host which is in agreement to the reported review by Neghabi et al. [15], since a high energy gap in the blend thin films, therefore allows the hosts to absorb more photon energy and transfer them efficiently to the guest Alq<sub>3</sub>.

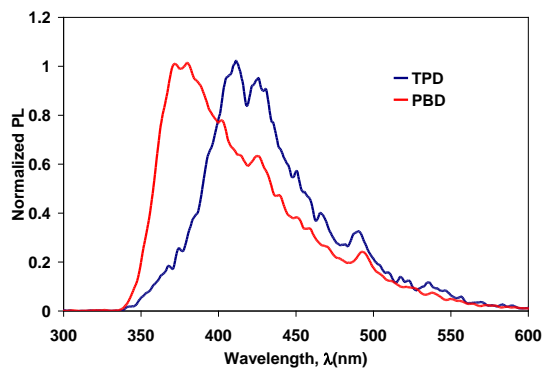


Fig. 8. Normalized PL spectra of TPD and PBD in thin film.

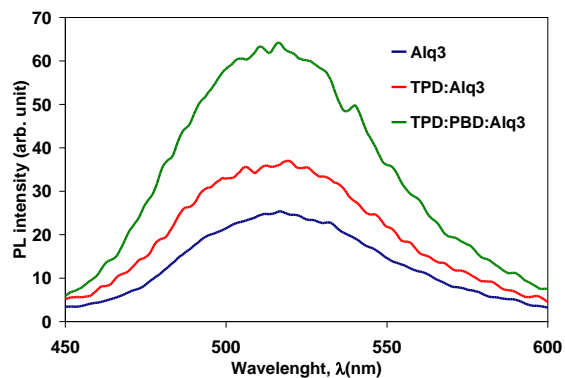


Fig. 9. PL spectra of the blend thin film.

### 3.2 Electroluminescence study

The current density-voltage ( $J$ - $V$ ) characteristics of the devices are shown in Fig. 10. It is observed that turn-on voltage reduces while current density increases with the increase in the blending elements in the emissive layer, which indicates that carrier injection and transportation have been enhanced by this blending technique. From Fig. 10, the anomalous  $J$ - $V$  characteristic is observed at low driven voltage range from 2.5 to 6.0 V for TPD:PBD:Alq<sub>3</sub> device, which is known as negative differential resistance (NDR) [16]. The NDR effect in OLED doped system is related to guest-hopping sites (GHS) and phonon scattering [17]. It has been reported that molecular blends will allow the formation of GHS in blended system [18]. Thus, it is suggested that host-molecules have created the GHS in such a device and resulted in NDR effect.

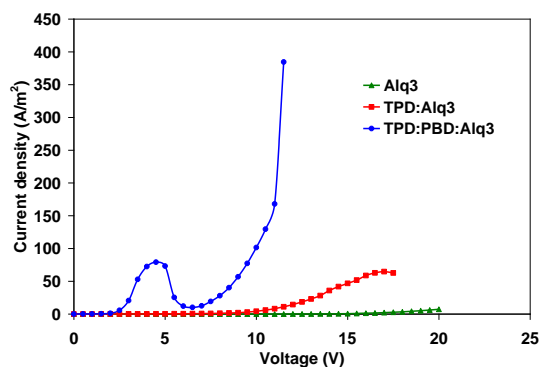


Fig. 10.  $J$ - $V$  spectra of the single layer OLED.

Fig. 11 shows the luminance-voltage ( $L$ - $V$ ) of the OLEDs with different blend systems. The significant improvement in the turn-on device (when  $L=1$  cd/m<sup>2</sup>) is obtained for TPD:PBD:Alq<sub>3</sub> blend (5 V) as compared to that of the Alq<sub>3</sub> device (15 V). The maximum luminance significantly improves from 8 cd/m<sup>2</sup> for Alq<sub>3</sub> OLED up to 347 cd/m<sup>2</sup> for TPD:PBD:Alq<sub>3</sub> blend OLED. A maximum current efficiency,  $\eta$  is obtained, which increases from 1.1 cd/A for Alq<sub>3</sub> to 2.7 cd/A for TPD:PBD:Alq<sub>3</sub> OLED. This indicates that both the TPD and PBD contribute to enhancement of the excitons formation in the Alq<sub>3</sub> molecule.

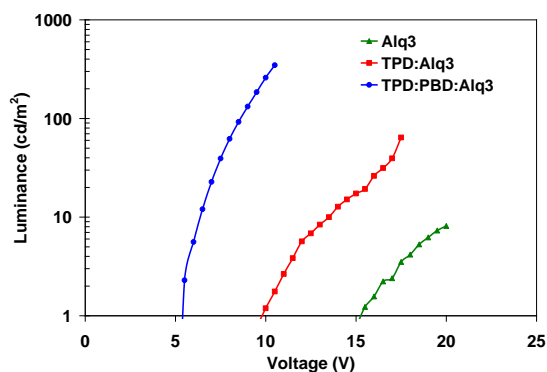


Fig. 11.  $L$ - $V$  plot of the OLED devices.

### 3.3 Impedance analysis

Fig. 12 (a) shows the Cole-Cole plots of the OLED devices with different blend systems at zero bias voltage in a large impedance scale. It is observed that the radius of Cole-Cole plot is reduced as the host molecules are blended into Alq<sub>3</sub> guest. This significant reduction of can be clearly seen in Fig. 12 (b). The value of device resistance obtained from simulation fitting is reduced from  $5 \times 10^5 \Omega$  for Alq<sub>3</sub> OLED to  $2.3 \times 10^3 \Omega$  for TPD:PBD:Alq<sub>3</sub> OLED. This reduction results in an increase in the effective conductivity up to magnitude of  $10^3$ . Host-molecules provide more conjugated bonds within the molecular structure which facilitate the carrier conductivity, thus enhance the transportation property in such a blend system. Table 1 shows the summary of device performances and impedance parameters for the OLED devices.

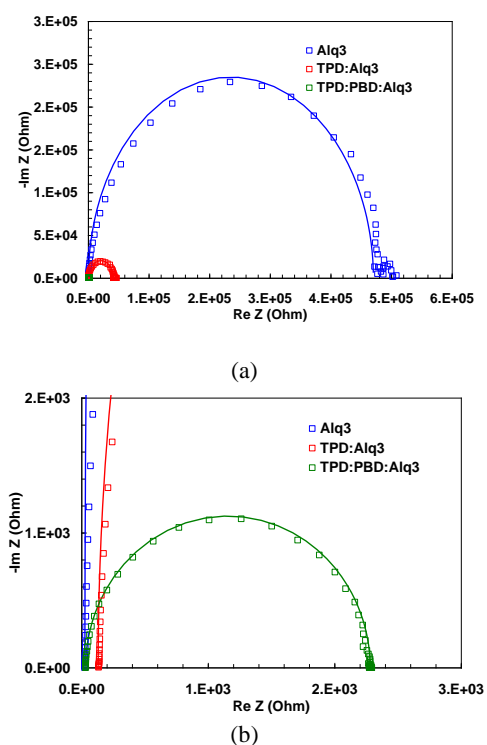


Fig. 12. Simulated (solid line) and experimental data (square box) of Cole-Cole plots of OLED device fabricated with different blends system at large (a) and low (b) impedance scales.

Table 1. Performances and impedance parameters of single layer OLED.

	Emissive layer		
	Alq <sub>3</sub>	TPD:Alq <sub>3</sub>	TPD:PBD:Alq <sub>3</sub>
$V_{on}$ ( $\pm 0.5$ V)	15.5	10.0	5.0
$V_{off}$ ( $\pm 0.5$ V)	20.0	17.5	11.5
$J_{max}$ ( $\pm 0.2$ A/m <sup>2</sup> )	7.3	62.8	384.6
$L_{max}$ (cd/m <sup>2</sup> )	8 $\pm$ 1	64 $\pm$ 4	347 $\pm$ 10
$\eta_{max}$ (cd/A)	1.1 $\pm$ 0.2	1.0 $\pm$ 0.3	2.7 $\pm$ 0.7
$R_p$ ( $\pm 0.5 \Omega$ )	$5.0 \times 10^5$	$4.5 \times 10^4$	$2.3 \times 10^3$
$\sigma_{eff}$ ( $\Omega^{-1}m^{-1}$ )	$(6 \pm 2) \times 10^{-9}$	$(7 \pm 3) \times 10^{-8}$	$(1.3 \pm 0.1) \times 10^{-6}$

### 4. Conclusions

The influences of host-molecules blend in Alq<sub>3</sub>-based OLED were investigated. The existence of TPD and PBD molecules in Alq<sub>3</sub>-based OLED has enhanced the charge carrier injection and transportation properties which resulted in a high maximum current efficiency of 2.7 cd/A. This new ternary system has offered a robust and effective approach to improve the performance of single layer OLED.

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