

## Development of efficient solution-processed red phosphorescent organic light-emitting diodes using carrier transport materials

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### ABSTRACT

We report on efficient solution-processed red emissive phosphorescent organic light-emitting diodes (PhOLEDs) employing bis[9-ethyl-3-(4-phenylquinolin-2-yl)-9H-carbazolato-*N,C*']iridium acetylacetonate (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and bis[9-(2-(2-methoxyethoxy)ethyl)-3-(4-phenylquinolin-2-yl)-9H-carbazolato-*N,C*']iridium acetylacetonate (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) doped into polymer host PVK, blended with electron transporting material OXD-7, and optimized concentration of carrier-transporting material TPD. The optimized PhOLEDs doped with 16 wt% carrier-transporting material shows a maximum luminance efficiency and power efficiency of 4.66 cd/A and 1.22 lm/W, which were increased by 40% and 20%, respectively, compared with that of reference device. The electroluminescence (EL) spectra exhibited the characteristic spectrum of (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) with its maximum at 630 nm and CIE (Commission International de L'Eclairage) coordinate of (0.66, 0.33).

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### 1. Introduction

Organic light-emitting diodes (OLEDs) are attractive as a next generation display due to their significant potential applications in solid state lighting source and flat-panel or flexible displays [1]. Recently, OLEDs using phosphorescent materials such as iridium or platinum complexes as emitting materials showed much higher efficiency than OLEDs fluorescent emitting materials [2–4]. In particular, phosphorescent OLEDs (PhOLEDs) materials open the possibility to develop highly efficient OLEDs since they are able to achieve emission from both singlet and triplet excited states. Therefore, an internal quantum efficiency of up to 100% is possible. Solution-processed OLEDs offer the potential for low fabrication cost, easy processing, and flexibility. Nevertheless it is more challenging to achieve good charge balance [5]. One effective way to achieve charge balance within emitting layer, incorporation of small molecules (molecular doping) into the fluorescent and phosphorescent emitting layers or synthesis of novel polymers has been used to improve the charge injection and transport [6–8]. To obtain high efficiency of PhOLEDs, the phosphorescent mate-

rials are usually doped as an emitting guest into a host material. The host materials of PhOLEDs should have a higher triplet excited state ( $T_1$ ) energy than that of the guest because of the need for  $T_1$  energy confinement on the guest [9]. Poly(*N*-vinylcarbazole) (PVK) is one of the most commonly used polymers in PhOLEDs due to its excellent film-forming properties, high glass transition temperature, wide band gap, and good hole mobility of  $\sim 10^{-5}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>, at typical electrical fields for OLEDs [10]. However, the electron mobility of PVK is rather poor. To achieve better balance of charge transport, most PVK-based PhOLEDs have appropriated an electron transporting component added to the emitting layer. One commonly used molecule is 1,3-bis[5-(4-*tert*-butylphenyl)-1,3,4-oxadiazole-2-yl]benzene (OXD-7) with the lowest unoccupied molecular orbital (LUMO) at  $-2.7$  eV and the highest occupied molecular orbital (HOMO) lower than  $-6.5$  eV [11].

In this work, we report on efficient solution-processed red emissive PhOLEDs, fabricated by doping a novel (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) or (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) as a phosphorescent dopant into host polymer matrix of PVK blended with an carrier-transporting material of *N,N'*-diphenyl-*N,N'*-(bis(3-methylphenyl)-[1,1-biphenyl]-4,4'-diamine) (TPD) and electron transporting material of OXD-7. It is well known that utilization of PEDOT:PSS leaves ca. 0.6 eV barrier for carrier injection to PVK. By the doping concentration of carrier-transporting material in emitting layer can promote the transport of carriers into emitting layer for increasing the probab-

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ity of carrier recombination. Consequently, the external quantum and luminance efficiency can be dramatically enhanced by as much as twofold comparing to that of using typical red emitters composed of host and dopant only.

## 2. Experimental

The PhOLEDs in this work were fabricated on indium tin oxide (ITO) covered glass substrates. The substrates were ultrasonically cleaned with detergent, deionized water, acetone, and isopropyl alcohol. Poly(3,4-ethylenedioxythiophene) doped with poly(styrene sulfonic acid) (PEDOT:PSS) (CLEVIOS P VP Al 4083) was spin coated onto the pre-cleaned patterned ITO glasses, yielding a layer with a thickness of 40 nm. The PEDOT:PSS layer was then baked at 150 °C for 20 min. In this work, novel red emissive Ir(III) complexes, (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) were synthesized by using procedures developed in our previous work [12]. PVK, OXD-7, TPD and (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) or (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) were dissolved separately in chlorobenzene. These solutions were then mixed with various ratios to give an appropriate weight percentages and spin coated on top of the PEDOT:PSS layer to obtain an emitting layer thickness of 70 nm. All solutions used in the PhOLED fabrication were filtered with 0.20 μm PTFE (hydrophobic) syringe filter. The doping concentrations of (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) or (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) were fixed to 6 wt%. The emitting layer was then annealed at 80 °C for 30 min. Finally, a 20 nm thickness of OXD-7 layer and 3 nm thickness of Ba as cathode and 100 nm thick layer of Al protection layer were then thermally evaporated vacuum ( $5 \times 10^{-6}$  Torr) subsequently on top of emitting layer. The film thickness was measured with an α-Step IQ surface profiler (KLA Tencor, SanJose, CA). To characterize the performance of PhOLEDs, the current density-voltage-luminance (*J-V-L*) characteristic were measured by using a current/voltage source meter (Keithley 236) and an optical power meter (CS-1000). All measurements were carried out at room temperature under ambient atmosphere without any encapsulation.

## 3. Results and discussion

Fig. 1(a)–(c) shows the molecular structures of (Et-Cvz-PhQ)<sub>2</sub>Ir(acac), (EO-Cvz-PhQ)<sub>2</sub>Ir(acac), PhOLED structure, and the energy band diagram of PhOLEDs, respectively. As shown in Fig. 1, the HOMO and LUMO energy levels of (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) observed for cyclic voltammetry measurement for the  $-5.02/-2.50$  and  $-5.03/-2.51$  eV, respectively. Such similarity in molecular structure of the Ir(III) complexes gives rise to very similar electro-optical property. The energy level diagram shows that the HOMO and LUMO energy levels of Ir(III) complexes lie above and below those of PVK host, respectively. Therefore, it is possible that Ir(III) complexes will trap both electrons and carriers within emitting layer. Moreover, the carrier block/electron transporting layer (OXD-7) was introduced for effective electron injection/transport and charge carrier balance within emitting layer. The LUMO energy level of OXD-7 was closely aligned to the LUMO level of Ir(III) complexes in the emitting layer. This good alignment of energy levels is important to efficient electron injection into the both transport molecules and Ir(III) complexes.

The UV-vis absorption spectra of Ir(III) complex solutions in chloroform and PL spectrum of PVK doped with OXD-7 as film state are shown in Fig. 2. As can be seen, the PL spectrum of host exhibits a maxima emission peak at 424 nm as excited by the light of wavelength 344 nm (at its maximum UV-vis absorption). These results show good overlap between emission spectrum of the host with the metal-to-ligand charge transfer (MLCT) absorption spectrum of the Ir(III) complexes, which ensures an efficient Förster

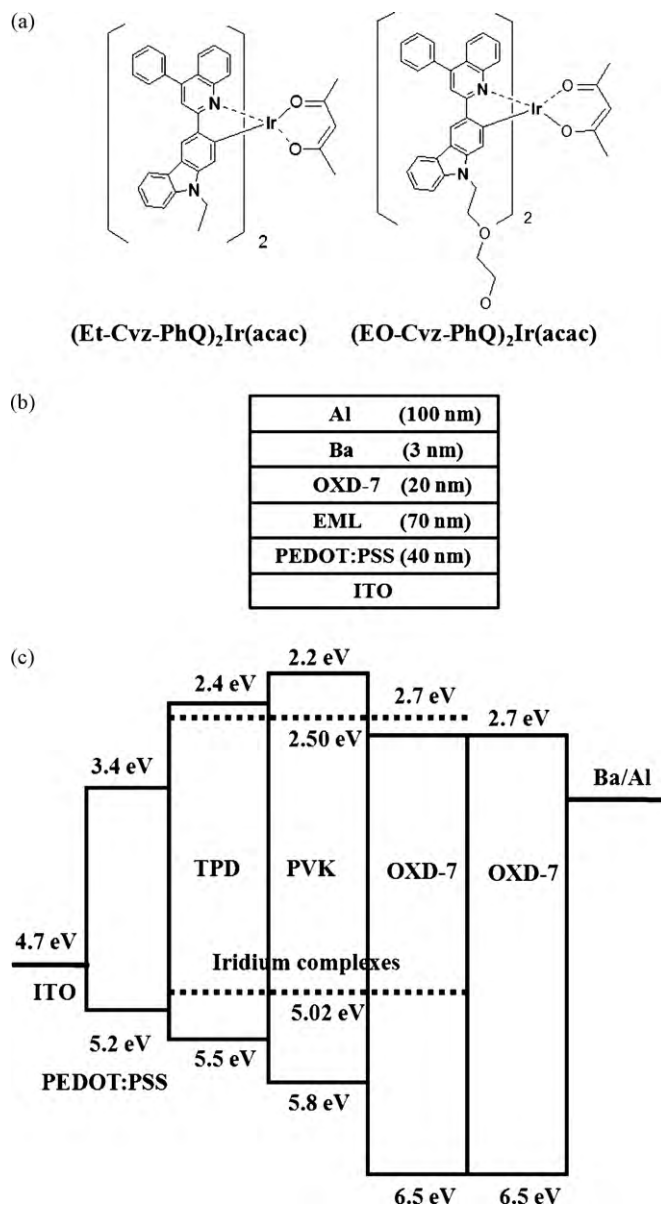


Fig. 1. Molecular structures of (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) (a), PhOLED structure (b), and energy level diagram (c).

energy transfer from host to guest and/or direct charge trapping of the Ir(III) complexes. In order to investigate the effect of doping carrier-transporting material in the performance of PhOLEDs, the doping concentration of 0, 4, 8, 12, 16 and 20 wt% for the carrier-transporting material were tested. As shown in the inset of Fig. 2, the EL spectra of PhOLEDs using (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) exhibited peaks corresponding to the dopant emission maximum of 630 nm with an excellent color purity at CIE coordinate of (0.66, 0.33) in the absence of host emission peak.

Fig. 3 shows the current density-voltage-luminance (*J-V-L*) characteristics of (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) for several doping concentration of carrier-transporting material in host. The maximum luminance of PhOLEDs using (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) and (EO-Cvz-PhQ)<sub>2</sub>Ir(acac) undoped carrier-transporting material in emitting layer were 1074 and 1014 cd/m<sup>2</sup>, respectively. However, the addition of increasing concentration of carrier-transporting material led to dramatic changes in the performances of PhOLEDs. For instance, turn-on voltage of the (Et-Cvz-PhQ)<sub>2</sub>Ir(acac) based on PhOLEDs reduces from 14 to 6 V.

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