Polymer Electrophosphorescent Light-Emitting Diode Using Aluminum Bis(2-Methyl-8-Quinolinato) 4-Phenylphenolate as an Electron-Transport Layer

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***Abstract*—The characteristics of organic light-emitting diodes depend critically on the arrangement and choice of the constituent organic layers. Diodes constructed using poly(vinylcarbazole) doped with phosphorescent *fac* tris(2-phenylpyridine) iridium (III) $[\mathrm{Ir(ppy)_3}]$ as the polymer hole-transport layers and aluminum (III) bis(2-methyl-8-quinolinato) 4-phenylphenolate as hole-blocking and electron-transport layers were investigated. The peak efficiencies of the diodes were sensitive to the concentration of $\mathrm{Ir(ppy)_3}$. With an optimal 2 wt% concentration, an effective external quantum efficiency of 10% photons/electron, a luminance power efficiency of 7.3 lm/W, and a low turn-on voltage of 6 V were obtained.**

***Index Terms*—BAlq, electro-phosphorescence, polymer organic light-emitting diode.**

I. INTRODUCTION

RECENTLY, nearly 100% [1]–[3] internal emission quantum efficiency has been obtained by harvesting both singlet and triplet excitons in organic light-emitting diodes (OLEDs) doped with phosphorescent emitters containing heavy metals. The strong spin-orbit coupling of a heavy metal enhances intersystem crossing and mixes the singlet and triplet states. Effective external quantum efficiency (η_{EQE}) of 19% photons/electron and luminance power efficiency (η_P) of 70 lm/W have been demonstrated in OLEDs based on small molecures [2], [3]. An important advantage of OLEDs based on polymers [4] over those based on small molecules is their compatibility with solution processing. This potentially allows a lower cost of production using room temperature spin-coating, screen-printing, or ink-jet printing techniques.

Phosphorescent emission dopants have also been applied to polymer-based OLEDs [5]–[14]. Using a single 100–200-nm-thick tris[9,9-dihexyl-2-(pyridinyl-2′) fluorene] iridium(III) doped blend of hole-transporting poly(9-vinylcarbazole) (PVK) and electron-transporting 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole, Gong *et al.* [12] demonstrated a high η_{EQE} of 10% photons/electron at an optimal doping concentration of 0.3 wt%. However, the turn-on voltage was relatively high, resulting in a low η_P of 2.5 lm/W. Using a double-layer structure of PVK doped with *fac* tris(2-phenylpyridine) iridium(III)— $\mathrm{Ir(ppy)_3}$ as the hole-transport layer and 1,3-bis[(4-tert-butylphenyl)-1,3,4-oxadiazolyl] phenylene as the electron-transport layer, Yang *et al.* [13] reported a higher η_P of 5.8 lm/W but a lower η_{EQE} of 7.5% photons/electron at a doping concentration of 6.8 wt%. Adding an extra hole-blocking layer, Vaeth *et al.* [14] recently reported a high η_{EQE} of 8.5% in a relatively more complex triple-layer device.

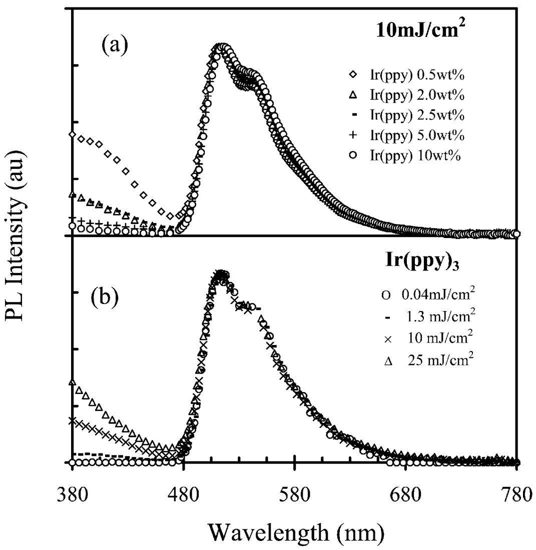

Fig. 1. (a) Normalized PL spectra of PVK doped with various concentrations of $\mathrm{Ir(ppy)_3}$ excited at an intensity of 10 mJ/cm^2. (b) Normalized PL spectra of PVK doped with 2 wt% $\mathrm{Ir(ppy)_3}$ excited at various intensity.

Since the performance of an OLED is sensitive to both the arrangement and the choice of the constituent organic layers, further improvement is quite possible. In this paper, the performance of simple double-layer diodes, consisting of poly(ethylene dioxythiophene):poly(styrene sulfonic acid) (PEDOT:PSS) as the hole-injection layer, $\mathrm{Ir(ppy)_3}$ doped PVK as emitting layers and aluminum(III) bis(2-methyl-8-quinolinato)4-phenylphenolate (BAlq) as electronic transport layers is reported. It was determined that BAlq behaved as an effective hole-blocking electron-transport layer. At an optimal $\mathrm{Ir(ppy)_3}$ concentration of 2 wt%, an η_{EQE} similar to the 10% photons/electron reported by Gong *et al.* and a high η_P of 7.3 lm/W have been obtained.

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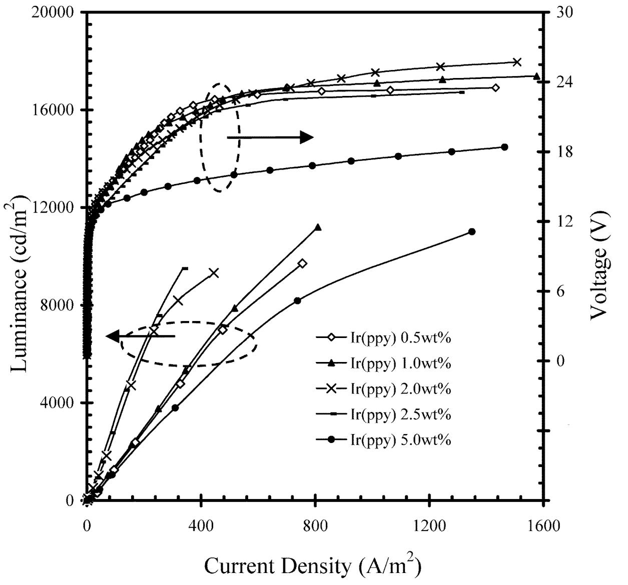

Fig. 2. EL intensity (L), current density (J), voltage (V) characteristics of Type B diodes with various $\mathrm{Ir(ppy)_3}$ concentrations.

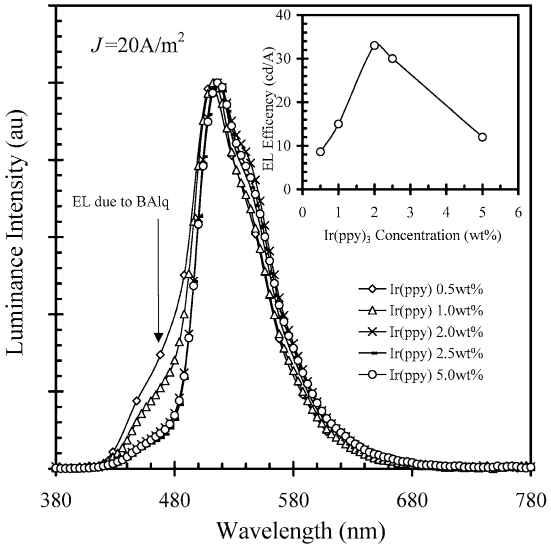

Fig. 3. Dependence of the EL spectra of Type B diodes on $\mathrm{Ir(ppy)_3}$ concentration. Inset is the corresponding dependence of the current efficiency on $\mathrm{Ir(ppy)_3}$ concentration.

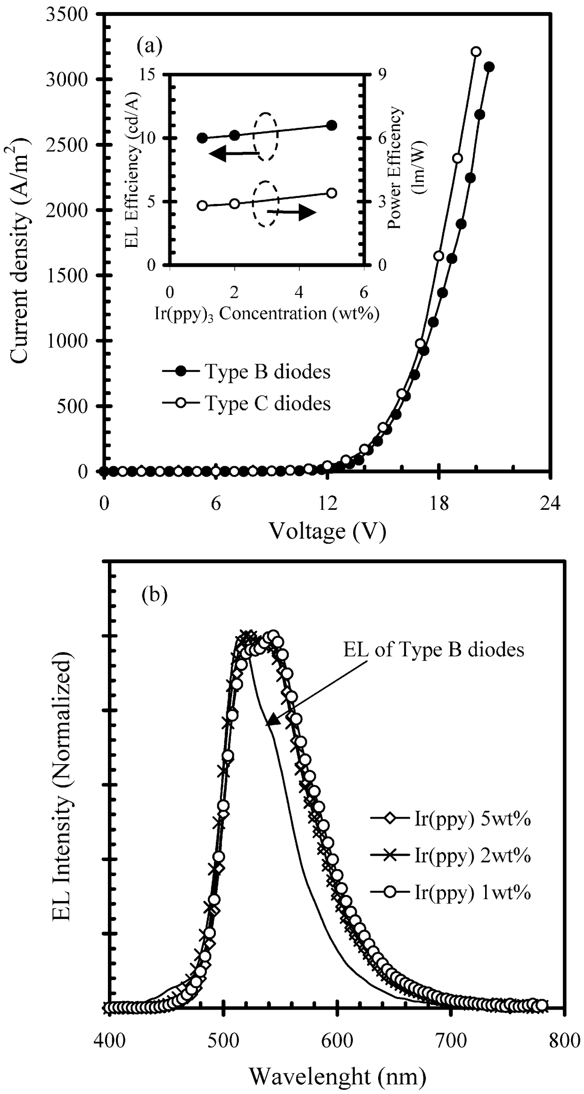

Fig. 4. (a) Current density (J), voltage (V) characteristics of Types B and C diodes with 5 wt% $\mathrm{Ir(ppy)_3}$ concentration. Inset is the dependence of the current efficiency and power efficiency of a Type C diode on $\mathrm{Ir(ppy)_3}$ concentration. (b) Corresponding normalized EL spectra of Type C diodes.

II. DIODE FABRICATION

Glass coated with 70-nm indium-tin oxide (ITO) with a sheet resistance of 30 Ω was used as the starting substrates. After a sequence of cleaning steps consisting of a 50 °C ultrasonic detergent soak for 30 min, deionized water spray for 10 min, ultrasonic deionized water soak for 30 min, 110 °C oven bake dry for 1–2 h and ultraviolet/ozone illumination for 10 min, PEDOT:PSS in water and $\mathrm{Ir(ppy)_3}$ doped PVK in trichloromethane (4 mg/mL) were sequentially spin coated. After each coating, the solvent was baked out in a vacuum oven at 54 °C for 2 h. The final thickness of PEDOT:PSS and $\mathrm{Ir(ppy)_3}$-doped PVK were 70 and 60 nm, respectively. Electron-transporting BAlq or *tris*-(8-hydroxyquinolinato) aluminum ($\mathrm{Alq_3}$) was then deposited in a vacuum thermal evaporation chamber with a base pressure of ~1 μ torr. The cathode, consisting of 1-nm lithium fluoride (LiF) and 150-nm aluminum (Al) [15], was subsequently evaporated through a shadow mask. The layer thickness during evaporation was measured *in situ* using a quartz crystal monitor. A Tencor P-10 surface profiler was used to determine the thickness of the spin-coated films.

Photo-luminance (PL) and electro-luminance (EL) spectra were measured using a PR650 photospectrometer. The PL was excited using the 337-nm line of a He-Cd laser, through a diffusing lens.

For comparison, two types of 4-mm-diameter OLEDs were fabricated using a set of shadow masks. These are as follows:

1) Type "B": ITO(75 nm)/PEDOT:PSS(70 m)/PVK: $\mathrm{Ir(ppy)_3}$(60 nm)/$\mathrm{BAlq_3}$ (50 nm)/LiF(1nm)/Al (150 nm).
2) Type "C": ITO/PEDOT:PSS/PVK: $\mathrm{Ir(ppy)_3/Alq_3}$ (50 nm)/LiF/Al.

TABLE I
COMPARISON OF PERFORMANCE OF TYPES B AND C OLEDS

OLED type	$\mathrm{Ir(ppy)_3}$ concentration (wt%)	Bias @ $10\mathrm{A/m^2}$ (V)	Current efficiency and brightness@ 16V	Maximum current efficiency (cd/A)	Maximum Power efficiency (lm/W)
B	0.5	9	14cd/A, $2376\mathrm{cd/m^2}$	15 (4.7%)	2.9
	1	10.5	14.5cd/A, $2284\mathrm{cd/m^2}$	15.4 (4.9%)	3.4
	2	11.2	33.1cd/A, $4521\mathrm{cd/m^2}$	34.1 (10.1%)	7.3
	2.5	10.9	30.5cd/A, $4718\mathrm{cd/m^2}$	30.5 (8.9%)	6.0
	5	9.8	12.3cd/A, $3795\mathrm{cd/m^2}$	12.3 (3.6%)	2.7
C	1	11	9.3cd/A, $4004\mathrm{cd/m^2}$	10 (2.9%)	2.8
	2	10.7	9.3cd/A, $5532\mathrm{cd/m^2}$	10.2 (3%)	2.9
	5	11	9.4cd/A, $4552\mathrm{cd/m^2}$	11 (3.2%)	2.8

Unless indicated otherwise, the thickness of the constituent layers in a Type C diode are the same as those in a Type B diode.

III. RESULTS AND DISCUSSION

Excited at an intensity of 10 $\mathrm{mJ/cm^2}$, the normalized PL spectra of PVK doped with various concentrations of $\mathrm{Ir(ppy)_3}$ are shown in Fig. 1(a). PL peaks are observed at ~400 nm for PVK fluorescence and at ~516 nm for $\mathrm{Ir(ppy)_3}$ phosphorescence. At low $\mathrm{Ir(ppy)_3}$ concentrations, the transfer of excitons from PVK to $\mathrm{Ir(ppy)_3}$ is less efficient because of the larger average separation between an excited site on the PVK host and an $\mathrm{Ir(ppy)_3}$ dopant. Therefore, emission near ~400 nm is observed, attributed to the PVK host. The average separation reduces and the exciton transfer efficiency improves with increasing $\mathrm{Ir(ppy)_3}$ concentration, leading to almost complete disappearance of the emission near 400 nm at concentration above 10 wt% at this excitation intensity. The PL spectra of PVK doped with 2 wt% $\mathrm{Ir(ppy)_3}$ and excited at various intensities is shown in Fig. 1(b). It can be seen that the PL peak at ~400 nm from the PVK host increases with excitation intensity. Below an excitation intensity of 1.3 $\mathrm{mJ/m^2}$, host fluorescence could not be observed. As the excitation intensity is increased and more excitons are generated on the host, a proportionally more severe incomplete transfer of these excitions to the phosphorescent dopants results in increased host florescence and reduced PL efficiency [16].

The EL intensity (L), current density (J), and voltage (V) characteristics of Type B diodes doped with various $\mathrm{Ir(ppy)_3}$ concentrations are shown in Fig. 2. For the L-J characteristics, only the current range up to the peak L of each diode is shown. The driving voltage at $J = 10$ $\mathrm{A/m^2}$ for each diode is shown in Table I. It can be seen that the driving voltage initially increases with $\mathrm{Ir(ppy)_3}$ concentration, peaks at 2 wt%, then decreases with further increase in concentration. The dependence of the effective current efficiency, defined as the slope of the L-J characteristics, on $\mathrm{Ir(ppy)_3}$ concentration is extracted and shown in the inset of Fig. 3. It is clear that a maximum is also obtained at 2 wt%.

The EL spectra of Type B diodes doped with various concentrations of $\mathrm{Ir(ppy)_3}$ and driven at $J = 20$ $\mathrm{A/m^2}$ are shown in Fig. 3. The most distinguishing feature differentiating the EL from the PL spectra [Fig. 1(a)] is the absence of any PVK-induced emission near ~400 nm, irrespective of the $\mathrm{Ir(ppy)_3}$ concentration. All EL spectra are quite similar, being dominated by $\mathrm{Ir(ppy)_3}$-induced peaks near ~ 516 nm. A further contrast with the PL spectra is the appearance of a blue emission shoulder at ~460 nm in diodes with $\mathrm{Ir(ppy)_3}$ concentration below 2 wt%. It is clear this emission originates from BAlq.

With BAlq replaced by $\mathrm{Alq_3}$ as the electron-transport layers in Type C diodes, similar J-V characteristics [Fig. 4(a)] but distinct EL spectra [Fig. 4(b)] with $\mathrm{Alq_3}$-induced emission at ~540 nm are obtained even in diodes with a high 5 wt% $\mathrm{Ir(ppy)_3}$ concentration. The effective current and power efficiencies of Type C diodes with various $\mathrm{Ir(ppy)_3}$ concentration are shown in the inset of Fig. 4(a). The effective current efficiency changes little with the $[\mathrm{Ir(ppy)}]_3$ concentration. The driving voltage at $J = 10$ $\mathrm{A/m^2}$ for each diode is also shown in Table I.

These observations can be explained with the following transport/emission mechanism [17]. Holes and electrons drift preferably and respectively through the $\mathrm{Ir(ppy)_3}$-doped PVK and BAlq (or $\mathrm{Alq_3}$). Emission is initiated in regions where the two populations overlap. At low $\mathrm{Ir(ppy)_3}$ concentration, both the fraction of holes trapped and the reduction in effective hole mobility are low. Consequently, holes are transported mainly through the PVK host. Blocked by neither BAlq nor $\mathrm{Alq_3}$, a finite amount of holes "leaks" into and initiates emission in BAlq [Fig. 5(a)] or $\mathrm{Alq_3}$. As the $\mathrm{Ir(ppy)_3}$ concentration is increased and the average separation among the $\mathrm{Ir(ppy)_3}$ molecules is reduced, direct $\mathrm{Ir(ppy)_3}$-to-$\mathrm{Ir(ppy)_3}$ hole hopping comes into play. Eventually it becomes the dominant transport mechanism, the effective hole mobility increases, and thus results in a reduction in the driving voltage (Table I). Similar concentration-induced switching from host- to dopant-based transport has been invoked to explain the behavior of OLEDs doped with fluorescent dopants [18]. At $\mathrm{Ir(ppy)_3}$ concentrations above 2 wt%, BAlq-induced blue EL emission is no longer observed [Fig. 5(b)]. Therefore, BAlq is an effective hole block for $\mathrm{Ir(ppy)_3}$-based hole transport. Even at a high concentration of 5 wt% $\mathrm{Ir(ppy)_3}$, holes transported on $\mathrm{Ir(ppy)_3}$ are injected upon reaching a nonblocking $\mathrm{Alq_3}$ interface, thus initiating emission in $\mathrm{Alq_3}$. These observations

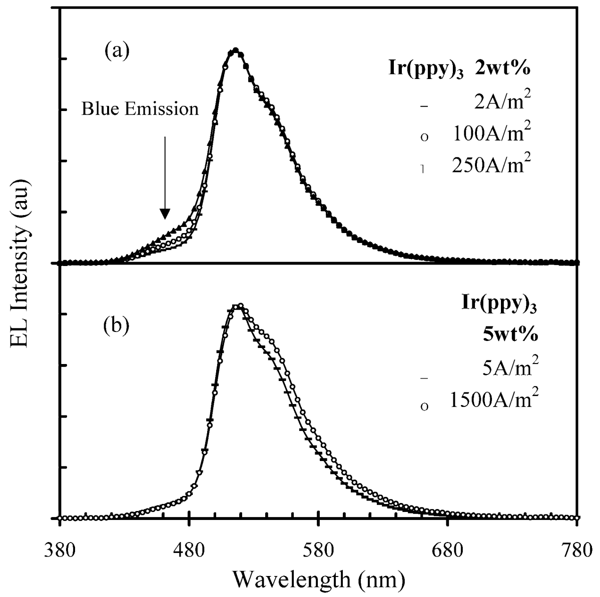

Fig. 5. (a) EL spectra of Type B diodes with (a) 2 wt% and (b) 5 wt% $\mathrm{Ir(ppy)_3}$ concentration at different current density.

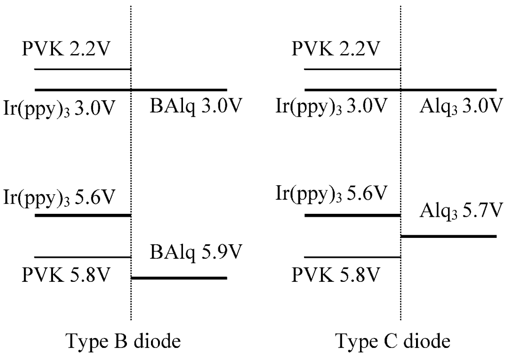

Fig. 6. Relative alignment of the characteristic energy levels of PVK, $\mathrm{Ir(ppy)_3}$, BAlq, and $\mathrm{Alq_3}$.

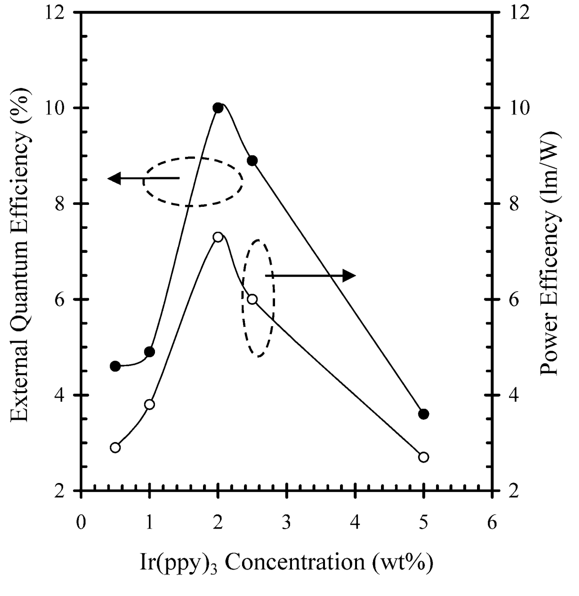

Fig. 7. Dependence of the effective external quantum and luminance power efficiencies on $\mathrm{Ir(ppy)_3}$ concentration of Type B diode.

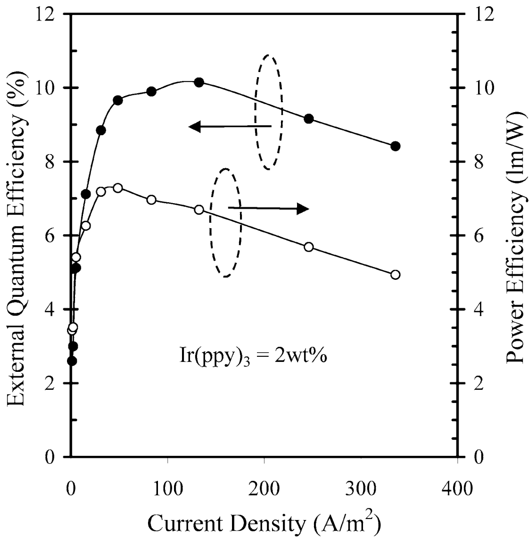

Fig. 8. Dependence of the effective external quantum and luminance power efficiencies on current density of a Type B diode with 2 wt% $[\mathrm{Ir(ppy)}]_3$ concentration.

are compatible with the alignment of the energy levels of the highest occupied molecular orbitals (HOMO) shown in Fig. 6 [19]. The dependence of the EL spectra of the Type B diodes on current density further verifies this mechanism.

The dependence of both peak η_{EQE} and η_P on $\mathrm{Ir(ppy)_3}$ concentration is shown in Fig. 7. Both efficiencies first increase, peak at $\sim$2 wt%, then gradually decrease with increasing $\mathrm{Ir(ppy)_3}$ concentration. The reduction in the efficiencies beyond the optimal concentration has been attributed to aggregate quenching [12].

The dependence of η_{EQE} and η_P on the current density of a diode with an optimal 2 wt% $\mathrm{Ir(ppy)_3}$ concentration is shown in Fig. 8. The peak η_{EQE} is 10% photons/electron (or $L = 2770$ cd/m^2 at $J = 83$ A/m^2). The peak η_P is 7.3 lm/W (or $L = 1577$ cd/m^2 at $J = 49$ A/m^2 and a driving voltage of 14 V). The turn-on voltage of 6 V (Fig. 2) is significantly lower than those of the single layer device [12] or diodes with 2,9-dimethyl-4,7-dipheny-1–10-phenanthroline (BCP) hole-blocking layers [19], [20]. The peak luminance is 9501 cd/m^2 at 18 V. The corresponding J is 336 A/m^2.

IV. CONCLUSION

The performance of OLEDs constructed with $\mathrm{Ir(ppy)_3}$ doped PVK and BAlq as the electron-transport layers has been characterized. The BAlq functions additionally as a hole-blocking layer for holes transported on $\mathrm{Ir(ppy)_3}$ but not for those transported on PVK. The peak efficiency of the diode was sensitive to the concentration of $\mathrm{Ir(ppy)_3}$. At 2 wt% concentration, a low turn-on voltage of 6 V, an effective peak external quantum efficiency of 10% photons/electron and a peak power efficiency of 7.3 lm/W have been measured.

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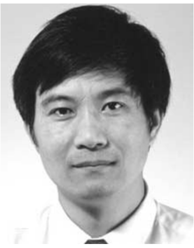

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