

Efficient Single Active Layer Electrophosphorescent White Polymer Light-Emitting Diodes**

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White organic light-emitting devices (WOLEDs) have drawn intense attention in both scientific and industrial communities due to their potential applications in full-color flat-panel displays, back-lighting sources for liquid-crystal displays and solid-state lighting sources.^[1,2] To achieve white emission, mixtures of the three red, green and blue (RGB) primary colors or two complementary colors, are typically required. Various approaches towards realizing WOLEDs have been reported, including multilayer structures capable of sequential energy transfer,^[3–5] multiple component emissive layers containing an appropriate ratio of RGB phosphorescent or fluorescent dopants,^[6–14] polymer blends containing RGB emitting species,^[8–16] charge transfer exciplexes or excimers broad emission^[4,17] and single component layers that utilize a polymer with broad emission.^[18–21]

Polymer light-emitting devices (PLEDs)^[8–15] offer the advantages of solution processing, including screen printing and ink-jet deposition, large area coverage, and low power consumption. However, white emission PLEDs are less efficient with respect to power efficiency (PE) and luminous efficiency (LE), when compared with devices fabricated using vacuum-deposition technologies. For example, vacuum deposited WOLEDs have efficiencies that exceed that of the incandescent light bulb (12–17 lm W^{–1}).^[1] In contrast, to the best of our knowledge, the most efficient white emission PLEDs are below 10 lm W^{–1},^[12–14] still far away from practical applications for solid-state lighting.

In this communication, we report single layer white PLEDs with an emission layer containing a blend of two or three phosphorescent iridium complexes^[22] within a poly(*N*-vinylcarbazole) (PVK)/1,3-bis[(4-*tert*-butylphenyl)-1,3,4-oxadiazolyl] phenylene (OXD-7) host matrix. Despite their simple architectures and straightforward fabrication procedures, these devices show excellent color purity and considerably improved power efficiencies and luminous efficiencies, when compared to other polymer-based white emission devices.

Ideal white light emission occurs near the equi-energy white point with Commission Internationale de L'Eclairage (CIE) coordinate of (0.333, 0.333). In a blend, the contributions from independent emitters need to be carefully balanced so that their collective integrated emission approaches this coordinate. We relied on the previously reported iridium bis(2-(4,6-difluorophenyl)-pyridinato-*N,C*^{2'}) picolinate (FIrpic, blue emission), iridium tris(2-(4-tolyl)pyridinato-*N,C*^{2'}) (Ir(mppy)₃, green emission), and iridium bis(1-phenylisoquinoline) (acetylacetonate) (Ir(piq), red emission); see Figure 1 for molecular structures. PVK was chosen as a host material due to its higher-lying triplet states (3.0 eV), excellent film-forming properties, high glass transition temperature (*T*_g) (~ 160 °C) and hole transport characteristics.^[23] OXD-7 was included into the host matrix to facilitate electron transport. To take advantage of ease of solution processing, for the RGB monochromatic and white emission devices, all have identical single emission layer configurations: ITO/poly (ethylenedioxythiophene): poly(styrene sulfonic acid) (i.e., PEDOT) (40 nm)/emission layer (70–80nm)/Ba (4 nm)/Al (120 nm).

PLEDs containing the individual phosphorescent species showed excellent efficiencies. For example, the peak LEs were 3.0 cd A^{–1} for red (Ir(piq)), 48.5 cd A^{–1} for green (Ir(mppy)₃), and 15.6 cd A^{–1} for blue (FIrpic), corresponding to external quantum efficiencies (EQEs) of 6.9 %, 14.5 %, and 7.7 %, respectively. The maximal luminances were 5760 cd m^{–2} (R), 24060 cd m^{–2} (G), and 26070 cd m^{–2} (B). Figure 2 shows where the electroluminescence (EL) spectrum of each RGB device falls on the CIE coordinates. The line joining the CIE coordinates of FIrpic (0.17, 0.36) and Ir(piq) (0.67, 0.32) pass through the white emission region, while Ir(mppy)₃ (0.30, 0.62) will shift the CIE coordinates toward the yellow emission region by increasing the green component.

A series of devices with various dopant concentrations and ratios were fabricated and evaluated, and the results are summarized in Table 1. The devices can be divided into two classes. One class contains a matrix of PVK/OXD-7 doped

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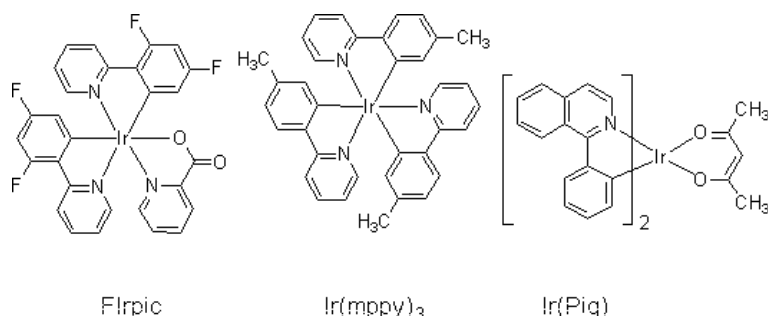


Figure 1. Chemical structure of iridium complexes.

with FIrpic (Devices A–D in Table 1), which is fixed at 10 wt % of the EL layer, and different Ir(piq) concentration. Depending on the content of Ir(piq), devices are referred to as Device A (0.25 wt %), B (0.33 wt %), C (0.50 wt %), and D (1.0 wt %). Figure 2a shows the EL spectra at 12 mA cm⁻² of these doubly-doped devices. Two intense peaks at 470 nm and 620 nm, with a small shoulder peak at 500 nm were observed, which are assigned to the emission from FIrpic and Ir(piq). There is a monotonous increase of the red emission as the doping concentration of Ir(piq) relative to FIrpic increases from 1:40 to 1:10. As shown in Table 1, the CIE coordinates at 12 mA cm⁻² fluctuate from (0.255, 0.357) to (0.465, 0.339); all are within or close to the white emission region. Of which, the EL colors of Devices B (0.319, 0.352), and C (0.329, 0.362) are close to the ideal CIE coordinate for pure white color.

The composition and performance of a second type of device, which contains the three phosphorescent dopants, are also included in Table 1. For these, the FIrpic is also maintained at 10 % of the total blend composition. The EL spectra of the devices with FIrpic: Ir(mppy)₃: Ir(piq) ratios from 50:1:1, 20:1:1, 30:0.1:1 to 50:0.1:1, i.e., Devices E to H in Table 1, are dominated by the narrow band gap green/red emitter despite their low concentrations (Fig. 2b). The CIE

coordinates of all devices, together with equi-energy white point (0.333, 0.333), are shown in Figure 3c for comparison.

Typical current density–luminance–voltage (*J*–*L*–*V*) characteristics and luminous efficiency–power efficiency–current density (LE–PE–*J*) characteristics of Device F in Table 1 are presented in Figure 3. The turn-on voltage (defined as the voltage where 1 cd m⁻² is measured) of 3.9 V is quite low among PVK-based white PLEDs, where usually high drive voltages are required because of low hole transport mobility (~ 10⁻⁵ m² V⁻¹ s⁻¹), unmatched charge carrier injection barrier heights and charge trapping effects. The low operation volt-

ages of the devices are important for reducing power consumption. The lowering of the turn-on voltage of the white emission devices can be attributed to enhanced hole transport properties due to incorporation of FIrpic which have been shown to exhibit bipolar transport properties.^[24] Device C is closest to white emission color and is characterized by a maximum forward viewing LE of 16.1 cd A⁻¹ at 15.3 mA cm⁻² and 8.4 V, which corresponds to an EQE of 10.0 %. Furthermore, the maximum PE of Device C is 6.3 lm W⁻¹ at 7.0 mA cm⁻² and 7.5 V. Device C reaches a maximum luminance of 14200 cd m⁻² at 12.0 V and 193.0 mA cm⁻².

Table 1 shows that Device E and F have the highest efficiencies, although their color is not exactly within the white emission region. For Device F, the corresponding efficiencies are 24.3 cd A⁻¹ at 20.8 mA cm⁻² and 9.3 V, which correspond to EQE of 14.4 %, and 9.5 lm W⁻¹ at 3.3 mA cm⁻², and 7.2 V, respectively. Device F reaches a maximum luminance of 25800 cd m⁻² at 13.2 V and 226.0 mA cm⁻². At a luminance for practical displays application, for example, 1000 cd m⁻², good forward viewing LE and PE values are maintained: 15.1 cd A⁻¹, 6.3 lm W⁻¹ at 7.0 mA cm⁻² and 7.5 V for Device C and 22.0 cd A⁻¹, 9.2 lm W⁻¹ at 4.4 mA cm⁻² and 7.5 V for Device F.

From Table 1, one observes that the triply-doped devices show better performance than the doubly-doped counterparts. This can be understood since Ir(mppy)₃ is a very efficient green emitter with intense emission at 512 nm, where the photopic response of human eyes is 344 lm W⁻¹, much stronger than for emission peaks of FIrpic (470 nm, 62 lm W⁻¹) or Ir(piq) (620 nm, 260 lm W⁻¹).

Color quality of the devices must be considered if used as lighting source.^[1] As shown in Table 1, Device C, E, F, G with appropriate color temperature (as measured as Correlated Color Temperature (CCT) within 2500–6500 K and relative higher Color Rendering Index (CRI) can find application for solid-state lighting. For solid-state lighting all the photons should be taken into account for illumination since they can be redirected to the forward viewing direction by engineering the lighting fixtures. That is, to get the total LE and PE, a fac-

Table 1. Device performance of RGB iridium complexes doped white emitting PLEDs.

Device	Composition	Device Performance					
		CIE [e]		Max LE [f] [cd A ⁻¹]	Max EQE [g] [%]	Max PE [h] [lm W ⁻¹]	CCT [K]
A [a]	40:1 [c]	0.255	0.357	16.0	12.9	7.6	9083
B [a]	30:1 [c]	0.319	0.352	14.6	11.4	5.6	6131
C [a]	20:1 [c]	0.329	0.362	16.1	10.0	6.3	5896
D [a]	10:1 [c]	0.465	0.339	4.9	5.0	2.2	2061
E [b]	50:1:1 [d]	0.301	0.467	21.9	13.8	10.0	6297
F [b]	20:1:1 [d]	0.343	0.468	24.3	14.4	9.5	5010
G [b]	30:0.1:1 [d]	0.382	0.469	18.9	12.9	8.2	3891
H [b]	50:0.1:1 [d]	0.257	0.395	16.3	12.1	5.4	8267

[a] Doubly-doped devices. [b] Triply-doped devices. [c] FIrpic:Ir(piq) ratio; FIrpic is fixed at 10 wt % of the active layer blend. [d] FIrpic:Ir(mppy)₃:Ir(piq) ratio; FIrpic is fixed at 10 wt % of the active layer blend. [e] Observer: 2 Degrees; obtained at 12 mA cm⁻² [f] Maximal front viewing luminous efficiency in cd A⁻¹ [g] Maximal external quantum efficiency in %. [h] Maximal front viewing power efficiency in lm W⁻¹.

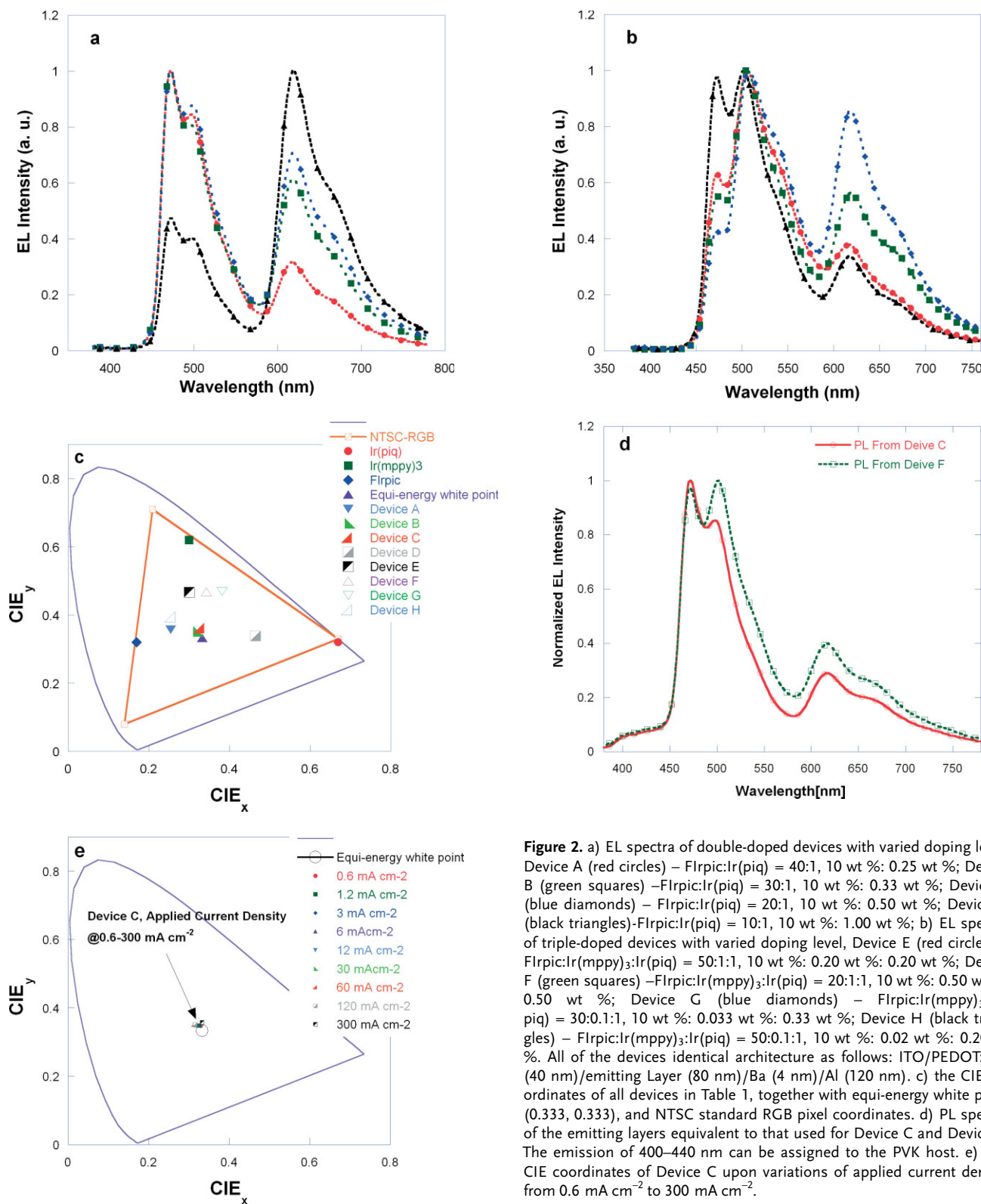


Figure 2. a) EL spectra of double-doped devices with varied doping level, Device A (red circles) – Flrpic:Ir(piq) = 40:1, 10 wt %: 0.25 wt %; Device B (green squares) – Flrpic:Ir(piq) = 30:1, 10 wt %: 0.33 wt %; Device C (blue diamonds) – Flrpic:Ir(piq) = 20:1, 10 wt %: 0.50 wt %; Device D (black triangles) – Flrpic:Ir(piq) = 10:1, 10 wt %: 1.00 wt %; b) EL spectra of triple-doped devices with varied doping level, Device E (red circles) – Flrpic:Ir(mppy)₃:Ir(piq) = 50:1:1, 10 wt %: 0.20 wt %: 0.20 wt %; Device F (green squares) – Flrpic:Ir(mppy)₃:Ir(piq) = 20:1:1, 10 wt %: 0.50 wt %: 0.50 wt %; Device G (blue diamonds) – Flrpic:Ir(mppy)₃:Ir(piq) = 30:0.1:1, 10 wt %: 0.033 wt %: 0.33 wt %; Device H (black triangles) – Flrpic:Ir(mppy)₃:Ir(piq) = 50:0.1:1, 10 wt %: 0.02 wt %: 0.20 wt %. All of the devices identical architecture as follows: ITO/PEDOT:PSS (40 nm)/emitting Layer (80 nm)/Ba (4 nm)/Al (120 nm). c) the CIE coordinates of all devices in Table 1, together with equi-energy white point (0.333, 0.333), and NTSC standard RGB pixel coordinates. d) PL spectra of the emitting layers equivalent to that used for Device C and Device F. The emission of 400–440 nm can be assigned to the PVK host. e) The CIE coordinates of Device C upon variations of applied current density from 0.6 mA cm⁻² to 300 mA cm⁻².

tor of 1.7–2.3 should be applied to the forward viewing efficiencies.^[5,7,12] Under these conditions, a maximum total LE of around 32 cd A⁻¹, a maximum total PE of around 12 lm W⁻¹

for Device C and 48 cd A⁻¹, 19 lm W⁻¹ for Device F are obtained. The overall performance of these devices is higher than those reported for multiply doped white PLEDs,^[8–14] and

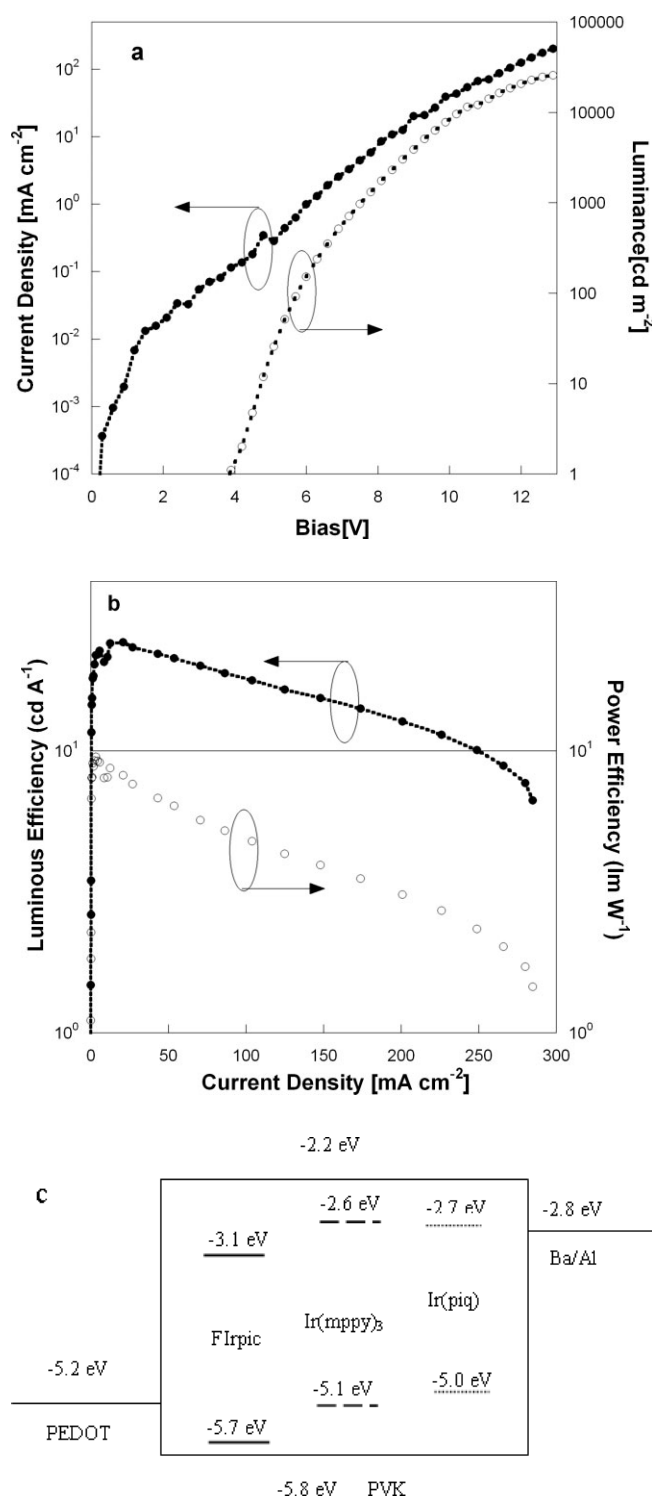


Figure 3. a) Current density-bias (J - V) characteristics. b) Luminous efficiency-power efficiency-current density (LE-PE- J) characteristics of Device F and c) Energy diagram of the devices.

is comparable with the performance parameters of vacuum-deposited devices.^[5,7]

It is interesting to consider the origin of the efficient white emission from devices in Table 1. Two independent excitation

mechanisms may occur; energy transfer from the fluorescent host to the phosphorescent guest materials and charge trapping at the emitter sites.^[1,7-11] As pointed out by many authors,^[25-28] significant deviation of EL spectra with PL emissions of devices from blend systems can be assigned to dominance of trapping mechanism in the EL process. Comparison of PL spectra (Fig. 2d) from the emitting layers with the EL spectra (Fig. 2a and b) of the devices reveals that the EL spectra have a much higher contribution from the narrow band gap red/green emitters, indicating that charge trapping is the dominant process. Moreover, the fact that the emission from PVK host (400–440 nm) is observed in PL but not in EL also supports such charge trapping mechanism. That is, when charge carriers are injected, simultaneous excitation of the three Ir complexes via direct charge trapping occurs; this is energetically favored and can be inferred from the energy level shown in Figure 3c. However, at the same time, energy transfer from wide band gap FIrpic to narrow band gap Ir(mppy)₃ or Ir(piq) via Dexter type short-range electron exchange cannot be ruled out. At this stage, the exact contribution to the excitation of narrow band gap emitters by energy transfer from FIrpic and by charge trapping is not understood.

The devices in Table 1 have similarities with those in ref. [7], where blue emitting bis(4',6'-difluorophenylpyridinato)tetraakis(1-pyrazolyl)borate (FIr6), green emitting *fac*-tris(2-phenylpyridine) iridium (Ir(ppy)₃), and red emitting iridium(III) bis(2-phenylquinolyl-*N,C*^{2'}) acetylacetonate (PQIr) were simultaneously doped into the wide gap host *p*-bis(triphenylsilyl)benzene (UGH2) to avoid exchange energy loss due to energy transfer from the host to guest. One of important features of these devices is that excitons are mainly formed on FIr6 sites by charge trapping directly due to the bipolar transport characteristics of FIr6 and are subsequently transferred to the narrow band gap emitter, where FIr6 sets the upper limit of the device efficiency. The devices in Table 1 are different in some important aspects, in which PVK/FIrpic and OXD-7 facilitate bipolar transport, allowing for simultaneous excitation for the three complexes rather than only for FIrpic via charge trapping (Fig. 3c). That is, in the devices in Table 1, Ir(piq) or Ir(mppy)₃ can be simultaneously excited via charge trapping and by energy transfer from FIrpic while FIrpic sites are mainly excited via direct resonant injection and charge trapping without the need of PVK excitation.^[7,29] The simultaneous excitation process is further supported by the fact that EQEs of the white emission devices in Table 1 is much higher than that of the monochromatic blue emitting devices (>10 % versus 7.7 %). Beside this, as stated above, comparison of EL spectra and PL spectra also reveals that charge trapping on the triplet emitters is the dominant process in the EL excitation. Although charge trapping is the dominant mechanism and Dexter energy transfer from FIrpic could also play an role in the excitation of low band gap emitters, conventional exothermic energy transfer from PVK to Ir(mppy)₃ and Ir(piq) may take place, which we could not rule out at this stage. In contrast, FIrpic sites are mainly excited via direct resonant injection and charge trap-

ping without need of PVK excitation due to bipolar transport properties and its behaving as traps. The low turn-on voltage of 3.3–3.6 V for Devices A–D (see Fig. 4) is also consistent with the charge trapping mechanism and direct exciton formation on the triplet emitters.^[7,29] Furthermore, much higher optimized doping level to achieve optimum device performance

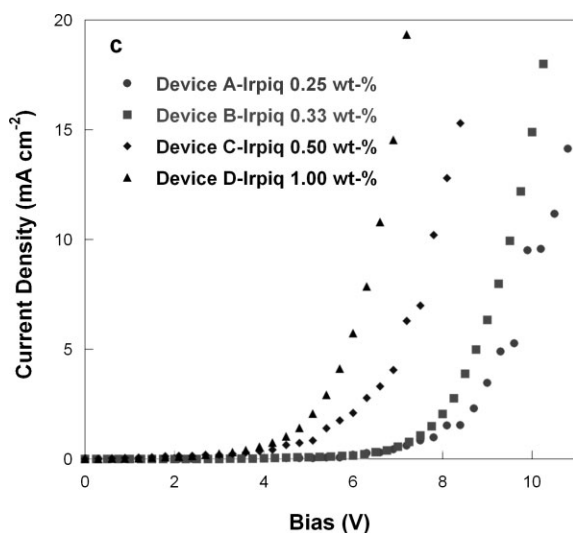


Figure 4. *J*–*V* characteristics for Device A–D, each emitting layer are uniformed as 80 nm.

for FIrpic monochromatic emission device (~10 wt %) imply excitation of FIrpic is not mainly through PVK host, and relative higher triplet energy (FIrpic (2.65 eV), PVK (3.0 eV), OXD (2.7 eV)) of FIrpic also suggests that energy transfer from PVK to FIrpic sites are much weaker than that of Ir(mppy)₃ and Ir(piq).

Figure 4 shows the *J*–*V* characteristics of the devices A–D in Table 1 as a function of Ir(piq) concentration. The drive voltage decreases with increasing Ir(piq) concentration, that is, as the doping concentration of Ir(piq) increases from 0.25 wt % to 1.00 wt %, the drive voltage corresponding to a current density of 10 mA cm^{−2} drops from 10.3 V to 6.5 V. At first glance one would expect that the increase in the Ir(piq) content would introduce more hole traps due to its high HOMO level, leading to lower hole mobilities and an increase in driving voltage. That the opposite is observed can be understood by considering that incorporating Ir(piq) into PVK:OXD-7:FIrpic not only introduces deep hole traps and shallow electron traps, but also facilitates hole injection. Actually, hole traps with depths of 0.6 eV and densities of 10¹⁸ cm^{−3} can retard hole transport thus improving charge balance. Moreover, the formation of Ohmic contacts (de-

fined as an injection barrier height < 0.3 eV) in the presence of Ir(piq) reduces the hole injection barrier of 0.5 eV (see Fig. 3c) and account for the drop in drive voltage upon increasing the Ir(piq) doping level of (Fig. 4). Therefore, the incorporation of Ir(piq) has a dual influence on the efficiency and *J*–*V* characteristics by decreasing hole mobility while forming Ohmic contact for hole injection. These arguments are consistent with theoretical analysis by Konezny and Smith et al.,^[30] in which the influence of traps on the efficiency of host-guest PLEDs are modeled. Recently, Campbell et al.^[31] have shown how shallow traps influence carrier mobility and relative EL intensity in doped conjugated polymer devices using conventional semiconductor statistics, and demonstrated that shallow traps (< 0.2 eV) at low concentration (~ 10¹⁸ cm^{−3}) do not significantly reduce charge carrier mobility. On the basis of these studies, we speculate that incorporating Ir(piq) (shallow traps with depth of 0.1 eV and densities of 10¹⁸ cm^{−3} for electron in this case) does not significantly change the transport properties of electrons.

Significantly, the emission color remains stable upon variations of applied current, which is important for illumination. For instance, for Device C, as the current density increases from 1.2 mA cm^{−2} to 120 mA cm^{−2}, the CIE coordinate shift from (0.336, 0.364) at 1.2 mA cm^{−2}, to (0.329, 0.362) at 12 mA cm^{−2} and (0.315, 0.352) at 120 mA cm^{−2}. Increasing the current density to 300 mA cm^{−2} produces a minor color shift leading to a CIE coordinate of (0.333, 0.357) (Fig. 2e). It is interesting to point out that similar color shifts can be observed for all devices in Table 1, that is, the relative intensity of the blue component of the emission color increases initially in the low current density region (0.6 mA cm^{−2} to 120 mA cm^{−2}). In the high current density region (≥120 mA cm^{−2}), the relative intensities of the red component start to increase slightly.

It is important to point out that thermal annealing post spin coating is important for achieving good device performance. Table 2 presents the influence of baking temperatures (from room temperature to 160 °C) on the device performance for Device C. After baking at 100 to 160 °C, the device performance improved considerably over the control devices (which is 6.4 cd A^{−1} for Device C). Despite these observations the role of thermal annealing remains uncertain at this point. The improvement may be attributed to improvements in charge injection and transport property. Enhanced electron injection can be supported by the change on surface morphology of the emissive layers upon annealing and variation of *J*–*L*–*V* char-

Table 2. Dependence of device performance on annealing temperature (Device C).

Device	Performance	Temperature [°C]					
		RT	80	100	120	140	160
Device C	Maximal LE [cd A ^{−1}]	6.4	8.3	15.3	16.1	15.6	15.1
	CIE [a]	(0.318,	(0.316,	(0.312,	(0.329,	(0.301,	(0.326,
	Coordinate	0.347)	0.355)	0.352)	0.362)	0.353)	0.357)

[a] Observer: 2 Degrees; obtained at 12 mA cm^{−2}.

acteristics of the devices on annealing temperature. As determined by AFM analysis (Fig. 5), for the control film, a root-mean-square (RMS) roughness of (0.42 ± 0.10) nm is obtained, corresponding to a smooth and homogenous surface. After thermal treatment, the film roughness increases to (2.12 ± 0.31) nm. Improved electron injection would be expected since rougher polymer-metal electrode contact can produce a more effective contact area for the cathode. On the other hand, morphology change upon annealing can also play an important role in determining the charge transport properties, in terms of release of buildup of space charge to a less extent^[32] and increase charge transport mobility. Indeed, a strong dependence of J - L - V characteristics (not shown) on annealing temperature clearly supports such improved charge transport and enhanced injection picture.

In summary, we report efficient single emission layer white PLEDs containing an active layer with two or three iridium

complexes doped into a PVK:OXD-7 matrix. We attribute the high efficiency to a combination of efficient RGB triplet emitters, the incorporation of electron transport materials with high triplet energy level (OXD-7) and bipolar carrier transport Iridium emitter (FIrpic) which ensures bipolar transport across the device and efficient exciton confinement on the Iridium sites. In addition, the narrow band gap red/green emitters are capable of balancing charge carrier ratio and forming Ohmic contact for hole injection, thereby improving power efficiency by reducing power consumption. Furthermore, post-annealing is necessary to achieve optimal performance. The simplicity of the device structure and the fabrication method, together with the high efficiencies observed, makes these white emitting PLEDs promising candidates for lighting applications.

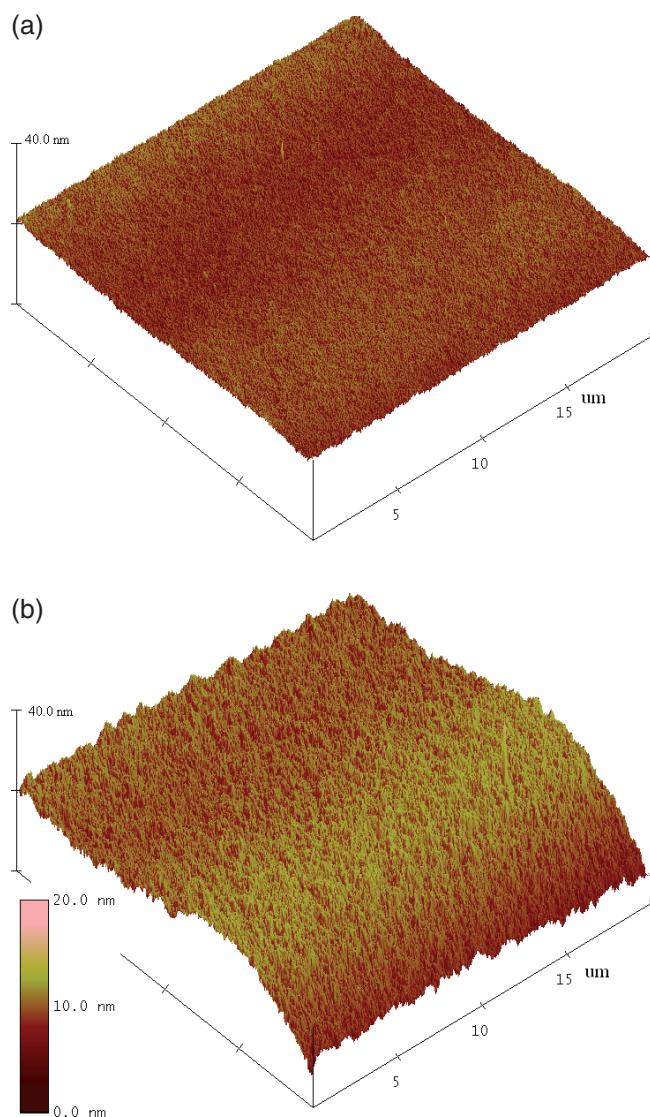
Experimental

PVK was purchased from Aldrich, while Ir(piq) and Ir(mppy)₃ were obtained from America Dyes Sources, respectively, and used as received. FIrpic and OXD-7 were synthesized in our laboratory. The PEDOT used was Baytron P 4083 (Bayer AG, H. C. Starck, Inc). Device fabrication process followed well-established processes that have been described elsewhere [33]. The J - L - V characteristics were obtained using a Keithley 236 source-measurement unit and a silicon photodiode calibrated by a spectroradiometer (PR-705 SpectraScan, Photo Research). The external QE of EL was collected by measuring the total light output in all directions in an integrating sphere with calibrated photodiodes (IS-080, Labsphere). The PL and EL spectra was recorded by a single-grating monochromator (Instaspec IV, Oriel Co.) equipped with a CCD detector or a PR-705 photometer.

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Figures 5. Typical topography images of films of a) before heat treatment and b) after heat treatment at 120 °C.

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