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We report highly efficient red-light-emitting devices based on a fluorene-triphenylamine tris[4-(9-phenylfluoren-9-yl)phenyl]amine (TFTPA) host doped with an osmium phosphor. The sterically hindered fluorene peripheries provided a compatible environment for the osmium dopant and alleviated concentration quenching of the phosphor at high doping levels. Increasing the doping concentration from 7 to 21 wt % dramatically decreased the driving voltage of the TFTPA-based devices, leading to improved power efficiency. The 21 wt % Os-doped device exhibited maximum luminous and powder efficiencies of 29.9 cd/A and 25.2 lm/W, combined with high efficiencies at high brightness. At 1000 cd/m^2 the efficiencies remained 29.2 cd/A and 22.2 lm/W. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2943155]

Organic light-emitting diodes (OLEDs) are attracting much attention because of their potential applications in full-color flat-panel displays.[1] Among these devices, electrophosphorescent OLEDs can harvest up to 100% of the excitons generated upon electrical injection, corresponding to a four-fold improvement in efficiency relative to that achievable in single-harvesting fluorescent OLEDs.[2–4] Over the past few years, many blue- and green-emitting phosphorescent OLEDs have been fabricated with highly luminous power efficiencies; for example, electroluminescence (EL) efficiencies as high as 26 and 60 lm/W have been reported for blue- and green-light-emitting devices, respectively.[3,5,6] In stark contrast, there are far fewer reports describing highly efficient red-emitting phosphorescent devices. For future device applications, it is essential for OLEDs to also exhibit energy-efficient, saturated red emissions. Although third-row metal complexes, such as those of Pt(II) and Ir(III), have been extensively applied as phosphorescent dopants in red-emitting OLEDs,[7–10] the device efficiencies remain far inferior to those of blue—or green-emitting phosphorescent devices. Recently, Os(II) complexes have been reported for use in red-emitting phosphorescent devices exhibiting high external quantum efficiencies; the relatively low oxidation potentials of these red-emitting osmium complexes, however, result in extremely dominant carrier direct-trapping effects in these

devices when using 4,4′-bis(9-carbazolyl)-2,2′-biphenyl (CBP) as the host material, in addition to higher operation potentials and unsatisfactory power efficiencies.[11]

In this letter, we describe a highly efficient red-emitting device incorporating a sterically hindered host material [the fluorene/triphenylamine hybrid tris[4-(9-phenylfluoren-9-yl)phenyl]amine (TFTPA)] and a phosphorescent guest Os(fptz)$_2$(PPh$_2$Me)$_2$ (fptz=3-trifluoromethyl-5-pyridyl-1,2,4-triazole), which possesses a relatively short phosphorescence lifetime (τ=96 ns).[11] Because triplet excitons tend to relax more slowly, their emissions inevitably reach saturation through a quenching mechanism involving triplet-triplet annihilation. As a result, we expected the shorter triplet-state exciton lifetime of Os(fptz)$_2$(PPh$_2$Me)$_2$ to minimize this problem. On the other hand, TFTPA is a good candidate for application as the host material for phosphorescent OLEDs because of its high thermal stability and excellent compatibility with organometallic phosphors.[12] The presence of the sterically hindered fluorene peripheries of TFTPA could alleviate the degree of concentration quenching arising from strong bimolecular interactions of the phosphors at high doping levels, leading to highly efficient phosphorescent devices.

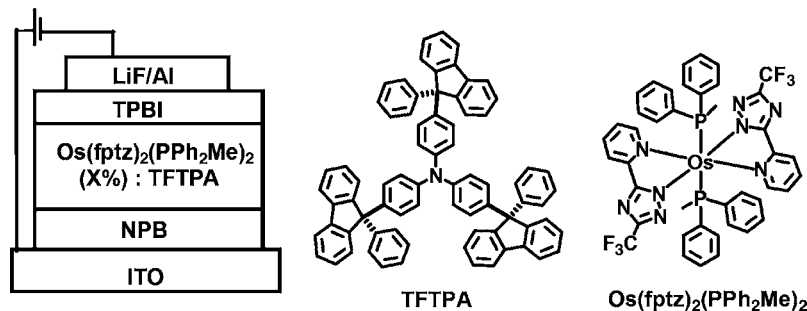Figure 1 illustrates the device configuration and molecular structures of the dopant and host used in

FIG. 1. Chemical structures of TFTPA and Os(fptz)$_2$(PPh$_2$Me)$_2$, and a schematic illustration of the device configuration used in this study.

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TABLE I. EL properties of TFTPA-based devices.

Dopant concentration (wt %)	V_{on}[a]	External quantum efficiency[b] (%)	Luminance efficiency[b] (cd/A)	Power efficiency[b] (lm/W)	Maximum power efficiency (lm/W)	CIE corrdinates[c] (x, y)
(7%)	3.0	11.9	19.4	11.0	11.6	(0.64,0.36)
(14%)	2.6	13.6	20.7	12.8	12.9	(0.64,0.36)
(21%)	2.5	18.0	29.2	22.2	25.2	(0.64,0.36)

[a]Recorded at 1 cd/m^2.
[b]Recorded at 1000 cd/m^2.
[c]Recorded at 7 V.

this study; Table I lists the crucial device performance characteristics. The typical multilayer architecture consisted of indium tin oxide (ITO)/4,4′-bis[*N*-(1-naphthyl)-*N*-phenylamino]biphenyl (NPB) (30 nm)/TFTPA: Os(fptz)$_2$(PPh$_2$Me)$_2$ (7–21 wt %, 40 nm)/1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBI), (40 nm)/LiF (1 nm)/Al (100 nm), where NPB and TPBI were employed as the hole-transporting layer and the electron-transporting layer, respectively. The fabrication of these EL devices— through sequential vapor deposition of the materials onto ITO glass under vacuum (3×10^{-6} torr)—and their characterization were performed using techniques similar to those we have previously reported.[13] Figure 2 displays the EL spectrum of the 21 wt % Os(fptz)$_2$(PPh$_2$Me)$_2$-doped TFTPA device. The EL spectrum exhibits a maximum intensity at 622 nm, with CIE coordinates (0.64, 0.36) very close to the standard red emission recommended by the National Television Standards Committee.

Figure 3 displays the current density-voltage-luminance (*I*-*V*-*L*) characteristics of red-emitting devices containing various Os(fptz)$_2$(PPh$_2$Me)$_2$ concentrations. The driving voltage of these devices remarkably decreased when the doping concentration was increased from 7 to 21 wt %; indeed, the 21 wt % Os(fptz)$_2$(PPh$_2$Me)$_2$-doped device exhibited an extremely low turn-on voltage (~2.5 V). This phenomenon suggests that the charges are directly injected to the dopant molecules at high doping concentrations; consequently, these moieties serve as an extra channel for the transport of carriers through their hopping between dopant molecules, thereby reducing the driving voltage.[3,14–17] Increasing the doping concentration from 7 to 21 wt % also improved the brightness of these devices; the 21 wt % Os(fptz)$_2$(PPh$_2$Me)$_2$-doped red-emitting device exhibited a

maximum luminance of 74 095 cd/m^2 at 11.5 V (797 mA/cm^2).

Furthermore, according to the plots of both the external quantum efficiency and the power efficiency with respect to the current density (Fig. 4), the device efficiency significantly improved when the dopant concentration was increased from 7 to 21 wt %. The maximum external quantum efficiency and maximum power efficiency of the 21 wt % Os(fptz)$_2$(PPh$_2$Me)$_2$-doped red-emitting device were 18.4% (29.9 cd/A, 6.07 mA/cm^2) and 25.2 lm/W, respectively. Relative to reported Os(fptz)$_2$(PPh$_2$Me)$_2$-doped CBP devices,[11] our TFTPA-based red-emitting device exhibits superior performance, particularly in terms of its power efficiency. We note that the power efficiency of the TFTPA-based device (17.4 lm/W at 20 mA/cm^2) was approximately three times higher than that of the optimized CBP-based device (6.3 lm/W);[11] we attribute this large improvement in device performance to the lower driving voltage of the TFTPA-based device. The inset of Fig. 2 reveals that the highest occupied molecular orbital energy level of TFTPA is −5.6 eV, which is higher than that of CBP (−6.1 eV). Os(fptz)$_2$(PPh$_2$Me)$_2$ serves as a less effective trap site for holes because of the shallow trap (depth: 0.7 eV) constructed by Os(fptz)$_2$(PPh$_2$Me)$_2$ in the TFTPA host; consequently, the minor degree of charge trapping results in the lowered operating voltage of the TFTPA-based device. More importantly, at high doping levels, TFTPA may act as an inert host matrix such that the charges are directly injected to the dopant molecules, which provide an additional channel for the transport of charges through hopping between dopant sites. Figure 5 displays topographic atomic force microscopy (AFM) images of the TFTPA and CBP films doped with 21 wt % Os(fptz)$_2$(PPh$_2$Me)$_2$. Apparently, the rigid cardo structure of
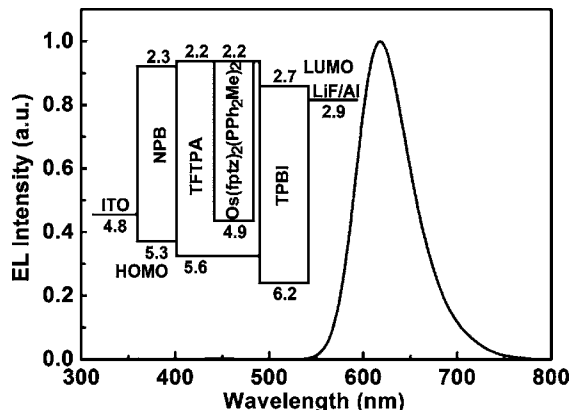

FIG. 2. EL spectrum of the of 21 wt % Os(fptz)$_2$(PPh$_2$Me)$_2$-doped TFTPA device. Inset: Energy level diagram of the device.
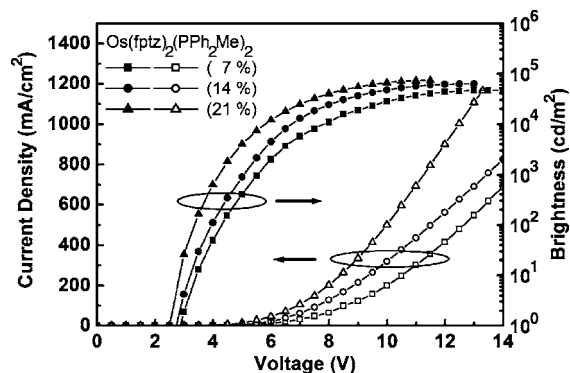

FIG. 3. Plots of current density (open symbols) and brightness (soild symbols) with respect to voltage for TFTPA-based devices at various doping concentrations.
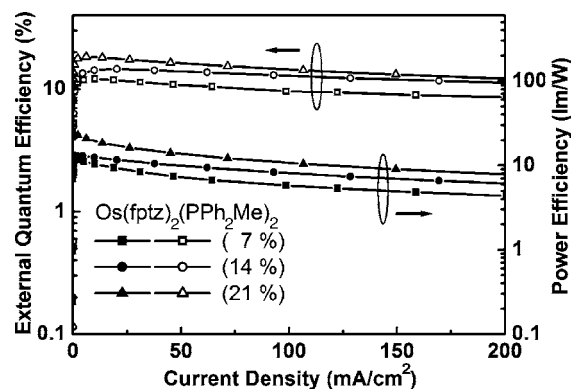
FIG. 4. Plots of external quantum efficiency and power efficiency with respect to current density for the TFTPA-based devices at various doping concentrations.

FIG. 5. (Color online) AFM topographic images (tapping mode) of the 21 wt % $Os(fptz)_2(PPh_2Me)_2$-doped (a) CBP and (b) TFTPA films.

TFTPA provides an ideal packing environment for dispersing and isolating the Os emitters. The doped TFTPA film was smooth, featureless, and homogeneous—its root-mean-square (rms) surface roughness was 0.41 nm—with no signs of phase separation or aggregation. In contrast, the doped CBP film possessed a rather rough morphology with a rms surface roughness of 2.10 nm, five times higher than that of the TFTPA blend. In addition to the excellent device performance, our TFTPA-based red-emitting device exhibited a less pronounced efficiency roll off at high brightness. For example, the efficiencies of the 21 wt % $Os(fptz)_2(PPh_2Me)_2$-doped TFTPA device at a practical brightness of 1000 cd/m^2 remained above 18.0% (29.2 cd/A) and 22.2 lm/W. Even when we increased the brightness to 10 000 cd/m^2, the corresponding power efficiency remained greater than 15 lm/W (27.4 cd/A). A high-current decline in efficiency is typical in phosphorescent OLEDs because of triplet-triplet annihilation.[5] The AFM images reveal that the Os emitters can be dispersed homogeneously in the TFTPA host, giving rise to improved efficiencies as a result of reductions in the degrees of both triplet-triplet annihilation and self-quenching of the red dopants.

In summary, we have realized a highly efficient red-emitting device through the incorporation of an osmium complex into the sterically hindered host material TFTPA. The peak power efficiency of our red-emitting device reached as high as 25.2 lm/W; this value is among the highest ever reported for a red electrophosphorescent device.[18] In addition to achieving a highly efficient red phosphorescence, our TFTPA-based devices exhibit a less-pronounced efficiency roll off at high cu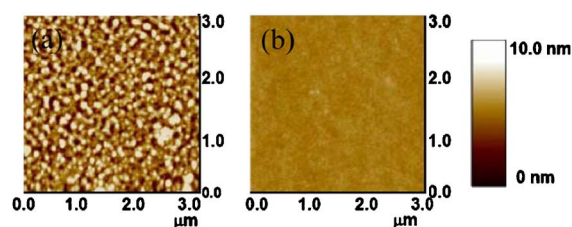rrent densities. Moreover, the excel-lent performances of our TFTPA-based devices were obtained using a simple device architecture, making them very attractive for commercial applications.

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