Very high-efficiency organic light-emitting diodes based on cyclometallated rhenium (I) complex

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The heavy metal complexes based phosphorescent (Ph) emitting materials have been extensively used to fabricate highly efficient electroluminescent (EL) devices since the pioneering work of Baldo et al. in Ph organic light-emitting diodes (PhOLEDs).\textsuperscript{1} The PhOLEDs based on Ph materials can significantly improve the EL performances because of the use of both singlet and triplet excitons which is attributed to strong spin-orbital coupling by heavy metal atoms and, therefore, have the potential of achieving an internal quantum efficiency of 100%.\textsuperscript{2} In recent years, heavy metal complexes of Os\textsuperscript{II},\textsuperscript{3–5} Ir\textsuperscript{III},\textsuperscript{6–9} Pt\textsuperscript{II},\textsuperscript{10–12} etc., based PhOLEDs have been extensively studied. In addition, Re\textsuperscript{I} complexes can also serve as a class of phosphor materials of the PhOLEDs,\textsuperscript{13–16} because they behave high phosphorescence quantum yield at room temperature (RT) and relatively short excited state lifetime as well as the excellent thermal and photochemical stabilities.\textsuperscript{17} Although red, green, and blue emitters of high color purity are required for full color display applications, there is still a great demand for the emitters that emit bright color such as orange emission could be used for multiple color display purposes or a component of the white OLEDs.\textsuperscript{18,19} Therefore, we demonstrate an electrically neutral Re\textsuperscript{I} bispyridyl complex, [(2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline)Re(CO)\textsubscript{3}]Br (Re-DDPA), that was used as an orange emitting dopant to fabricate PhOLEDs using 4,4′,N,N′-dicarbazole-biphenyl (CBP) as the host. The devices exhibit a maximum efficiency of 21.8 cd/A and a peak luminance of 8315 cd/m\textsuperscript{2}, respectively. Such high EL performances may be attributed to the synergistic effects of two repulsive phenyl groups and incorporation of two methyl groups.

Re-DDPA was synthesized by reacting 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline with pentacarbonyl bromorhenium in refluxing toluene solutions. The precipitates were easily sublimed to obtain a pure electrically neutral carbonyl diamine Re\textsuperscript{I} complex, (2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline)Re(CO)\textsubscript{3}Br. The Re complex was used as an orange emitting dopant in 4,4′-N,N′-dicarbazole-biphenyl host to fabricate Ph organic light-emitting diodes (PhOLEDs). The maximum electroluminescence (EL) efficiency and luminance of 21.8 cd/A and 8315 cd/cm\textsuperscript{2} were harvested, respectively, which were the highest EL results among the PhOLEDs based on Re\textsuperscript{I} complexes. The improvements of the EL performances could be ascribed to the synergistic effects of together incorporation of two reciprocally repulsive phenyl and methyl groups on the backbone of 1,10-phenanthroline molecule of the ligand. © 2008 American Institute of Physics. [DOI: 10.1063/1.2888767]
sorption bands in the range 350–460 nm are tentatively ascribed to the metal-to-ligand charge transfer (MLCT) transition as well as an intense 566 nm emission at RT under excitation of 360 nm is determined. Obviously, there is a sufficient overlap between PL spectrum of CBP film and MLCT absorption band of the Re-DDPA in chloroform solutions. This indicates that, if Re-DDPA is doped into CBP host, efficient Förster energy transfer from the CBP host to Re-DDPA dopant in the device would occur.

Figure 2 presents the characteristics of current efficiency as a function of current density in various Re-DDPA contents based devices, and a maximum current efficiency of 21.8 cd/A for the 7 wt% Re-DDPA doped device is observed. The value is higher than the highest efficiency of the PhOLED device based on Re complex reported in Ref. 23 by about 1.2 folds. Moreover, it is also interestingly noted that at a current density of 100 mA/cm² the 30 wt% Re-DDPA doped device still exhibits a maximum current efficiency of 5.1 cd/A, which shows smaller concentration quenching although the Re complex has a short excited state lifetime of 0.24 μs. The reason may be attributed to the special molecular structure and high electron mobility of the ligand (i.e., DDPA) (Ref. 24) of the Re complex.

Figure 3(a) depicts luminance-current-voltage (L-I-V) curves of 30 wt% Re-DDPA doped CBP device. We can see that the device presents a peak luminance of 8315 cd/cm² at 17.5 V, which is larger than the highest one of the Re complex based the device previously reported23 by about 1.3 folds, and the turn-on voltages are lower than 5 V. The inset of Fig. 3(a) indicates EL spectra of the devices with 30 wt% Re-DDPA at a variety of bias voltages from 8 to 18 V. All EL spectra show only identical orange emission bands and their spectral figures are the same as the PL spectrum in Fig. 1. Thinking about the relationship between PL and EL emissions as well as the independence of EL emission spectra on the bias voltage, we conclude that, under electrical excitation, there could be an efficient energy transfer from CBP host to the Re complex emitter. However, comparing the LUMO and HOMO levels of the Re complex with those of CBP host molecule shown in Fig. 4. We note that both the LUMO and HOMO levels of the Re-DDPA molecule locate within those of CBP host molecule. This implies that carrier
trapping at the Re-DDPA molecule in the host also probably occurs. The detailed luminescent mechanism will be published elsewhere.

On basis of the excellent EL properties of the phosphor Re-DDPA, we compare its maximum EL efficiency and luminescence with those of the previous three structurally similar Re complexes, (1,10-phenanthroline)Re(C)3Cl (Re-phen), (2,9-dimethyl-1,10-phenanthroline)Re(CO)3Cl (Re-dmphen), and (bathophenanthroline)Re(CO)3Cl (Re-bath), as listed in Table I. Compared with the complex without substituents on the 1,10-phenanthroline ligand (i.e., Re-phen), incorporation of two methyl groups on 2,9 positions of 1,10-phenanthroline can sterically prevent structural relaxation resulting in nonradiative decay due to the narrowed energy gap between 1MLCT and 3MLCT. In addition, introduction of two phenyl groups on 4,7 positions of 1,10-phenanthroline backbone does not greatly affect the energy level is used as a unit of eV. The maximum current efficiency at 7 wt % doped CBP based device.

The 7 wt % Re-DDPA doped device showed a maximum current efficiency of 21.8 cd/A and 30 wt % Re-DDPA doped device offered a peak luminance of 8315 cd/m², and under even a current density of 100 mA/cm² the latter device still maintained a current efficiency of 5.1 cd/A. Such excellent performances may be attributed to special molecule structure of Re-DDPA and high electron mobility of the ligand in the Re complex, although the detailed EL mechanisms need to be further understood. We can expect that the Re-DDPA complex could be a potential candidate in construction of simpler white PhOLEDs devices by combining a blue emitter and also become a luminescent sensitizer of the red or infrared emissive EL materials.

\[\text{TAB. I. Combination of key parameters between Re-phen, Re-dmphen, Re-bath, and Re-DDPA based PhOLEDs.}\]

<table>
<thead>
<tr>
<th>Triplet emitters</th>
<th>(\eta (\text{cd}/\text{A})^a)</th>
<th>(L_{\text{max}} (\text{cd}/\text{m}^2)^a)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Re-phen</td>
<td>6.67</td>
<td>2769</td>
<td>15</td>
</tr>
<tr>
<td>Re-dmphen</td>
<td>7.15</td>
<td>3686</td>
<td>15</td>
</tr>
<tr>
<td>Re-bath</td>
<td>13.8</td>
<td>4347</td>
<td>29</td>
</tr>
<tr>
<td>Re-DDPA</td>
<td>21.8(^c)</td>
<td>8315(^d)</td>
<td>This work</td>
</tr>
</tbody>
</table>

\(^a\)The peak current efficiency.
\(^b\)The maximum luminescence.
\(^c\)The maximum current efficiency at 7 wt % doped CBP based device.
\(^d\)The maximum luminescence at 30 wt % Re-DDPA doped CBP based device.