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Organic light-emitting devices using ruthenium (II) (4,7-diphenyl-1,10-phenanthroline)$_3$ as dopant

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Abstract

The light emitting devices using ruthenium(II)(4,7-diphenyl-1,10-phenanthroline)$_3$ ($[Ru(dphphen)_3]^{2+}$) complex as dopant and emitter in poly(*N*-vinylcarbazole) (PVK)-based blend matrix have been studied in different structures. In comparison with the indium-tin-oxide glass (ITO)/PVK:2-(4-biphenyl)-5-(4-ter-butylphenyl)-1,3,4-oxadiazole (PBD):$[Ru(dphphen)_3]^{2+}$/tris-(8-hydroxyquinoline) aluminum (Alq$_3$)/Al, ITO/PVK:PBD:$[Ru(dphphen)_3]^{2+}$/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP)/Alq$_3$/LiF/Al, and ITO/PVK:*N,N'*-diphenyl-*N,N'*-bis(3-methylphenyl)-[1,1'-biphenyl]-4,4'-diamine (TPD):PBD:$[Ru(dphphen)_3]^{2+}$/BCP/Alq$_3$/LiF/Al devices, the ITO/PVK:PBD:$[Ru(dphphen)_3]^{2+}$/Alq$_3$/BCP/Alq$_3$/LiF/Al device shows efficient improvement in device brightness and efficiency. Furthermore, the chromacity-tunable electroluminescence is obtained by the hybrid emission from $[Ru(dphphen)_3]^{2+}$ and Alq$_3$ species in such a device structure.

Keywords: Electroluminescence; Conducting polymer; Heterojunctions

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1. Introduction

Organic light-emitting devices (OLEDs) have been prompted greatly in both basic science and device technology for large-area and ultra-thin color display systems, due to the low cost fabrication process and variable emission color in the entire visible region. In such devices, the light is generated from the radiative recombination of hole (injected from a high-work function transparent anode) and electron (injected from a low-work function metal cathode) at an electrically excited emitter. Besides controlling the optical bandgap of the emitting species, the emission color can be tuned by doping dyes with different color in single-layer device, polymer blends or by controlling the exciton ratio between the emitters/carrier recombination zones through the bias or device structure adjustment [1-5].

Ruthenium (Ru) complexes are receiving attention as a new class of emitting materials [6-16]. With the advantage of chemical stability, this kind of material can give adjustable emission color in the orange-red region and redox properties by varying ligand structure. In a indium-tin-oxide glass (ITO)/Ru complex/cathode structure, the light emitting process is via a electrochemical bimolecular reaction [10]:

$$[Ru^{III}(L)_3]^{3+} + [Ru^{II}(L^{\cdot -})(L)_2]^{+} \rightarrow [Ru^{III}(L^{\cdot -})(L)_2]^{2+*} + [Ru^{II}(L)_3]^{2+} \rightarrow 2[Ru^{II}(L)_3]^{2+} + h\nu \quad (1)$$

In such a device structure, the turn on voltage is low (close to the metal-to-ligand charge transfer bandgap of typically 2-3 eV), but the response time is slow due to the redistribution process of the counterions in $[Ru(L)3]^{2+}$ emissive layer [8-11].

We have reported the instantaneous emission from ruthenium (II) (4,7-diphenyl-1,10-phenanthroline)3 ([Ru(dphphen)3]2+) complex, by using it as a dopant and emitter in poly(N-vinylcarbazole) (PVK)-based blend matrix [18]. Here, we investigate the electroluminescent properties of LEDs in two new device structures with $[Ru(dphphen)3]^{2+}$ complex as dopant. The devices were fabricated in ITO/ PVK:N,N'-diphenyl-N,N'-bis(3-methylphenyl)-[1,1'-biphe-nyl]-4,4'-diamine (TPD):2-(4-biphenyl)-5-(4-ter-butylphe-nyl)-1,3,4-xadiazole (PBD):$[Ru(dphphen)3]^{2+}$/2,9-dimethyl-4,7-diphenyl-1,10-phenanthrol-ine (BCP)/tris-(8-hydroxyq-uinoline) aluminum (Alq3)/LiF/Al and ITO/PVK:PBD:[Ru (dphphen)$_3]^{2+}$/Alq$_3$/BCP/Alq$_3$/LiF/Al. The former structure was expected to improve $[Ru(dphphen)_3]^{2+}$ emission and the latter for tunable emission chromacity.

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2. Experimental

PVK (average Mw 1,100,000), Alq_3 and BCP were purchased from Aldrich Chem. Co. and used as received. $[Ru(dphphen)_3](ClO_4)_2$ was prepared as described elsewhere [19]. The $[Ru(dphphen)_3]^{2+}$ doped PVK solution was prepared from a 1,1,-2 trichloroethane solution of 100: 25:40:0.75 PVK:TPD:PBD:$[Ru(dphphen)_3]^{2+}$ or 100:40: 0.75 PVK:PBD blend by weight. The ITO glass (Rs: 5-15 Ω/□) was etched into 2 × 3 mm^2 emitting areas, and ultrasonically cleaned in ethanol, acetone, dichloromethane for 10 min each. Before film spinning, the ITO glass was irradiated using a UV lamp (250 W) for 15 min.

The $[Ru(dphphen)_3]^{2+}$ doped PVK blend layers were spin coated onto the ITO glass from the above solutions. The obtained blend films have a thickness range of 50-60 nm measured by a Taylor Hobson Talystep. The films were heat-treated at 90 °C for 1 h in a vacuum oven. A BCP layer (10 nm) was used for blocking hole from entering the cathode, followed by an Alq_3 buffer layer (15 nm). Finally a bilayer cathode of LiF (1 nm)/Al (100 nm) was deposited. All deposition was under a vacuum pressure of ~ 10^{-5} Torr. For the tunable chromacity, the device was constructed in a ITO/PVK:TPD:PBD:$[Ru(dphphen)_3]^{2+}$/Alq_3/BCP/Alq_3/Al structure by inserting an Alq_3 layer (15 nm) as the co-emitter, between the BCP and $[Ru(dphphen)_3]^{2+}$ doped PVK blend layers.

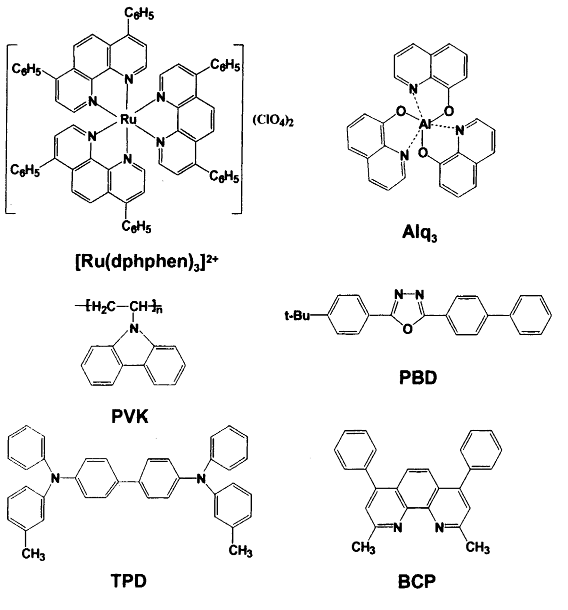

Fig. 1. Molecular structures of the $[Ru(dphphen)_3]^{2+}$ and Alq_3, PVK, TPD, PBD and BCP, with $[Ru(dphphen)_3]^{2+}$ and Alq_3 as the emitters, PVK, TPD as the hole transporters, PBD as the electron transporter and BCP as the hole blocker.

3. Results and Discussion

Fig. 1 shows the molecular structures of the used materials $[Ru(dphphen)_3]^{2+}$, Alq_3, PVK, TPD, PBD and BCP. In a ITO/$[Ru(dphphen)_3]^{2+}$/Ag device $[Ru(dphphen)_3]^{2+}$ showed emission characteristic of a electrochemical mechanism (emitting at 630 nm), with a response time longer than 15 minutes [16]. The electronic/instantaneous $[Ru(dphphen)_3]^{2+}$ emission was obtained and was dominant in both device structures of ITO/PVK:PBD:$[Ru(dphphen)_3]^{2+}$/Alq_3/Al and ITO/PVK: PBD:$[Ru(dphphen)_3]^{2+}$/BCP/Alq_3/Al [18], in which $[Ru(dphphen)_3]^{2+}$ was used as the dopant. Fig. 2 shows the electroluminescence spectra of those devices. In such devices the $[Ru(dphphen)_3]^{2+}$ molecules could be electronically isolated from each other by the PVK:PBD host matrix, as indicated by the blue-shifted emission wavelength which remains at 602 nm when the doping concentration is decreased further. The instantaneous $[Ru(dphphen)_3]^{2+}$ emission has been attributed to the direct exciton formation at $[Ru(dphphen)_3]^{2+}$ molecule by charge trapping from PVK and PBD carrier transporters. A ITO/PVK:PBD:$[Ru(dphphen)_3]^{2+}$/BCP/Alq_3/LiF/Al device was measured to give a brightness of 1100 cd/m^2 (0.73 cd/A) at 26 V bias.

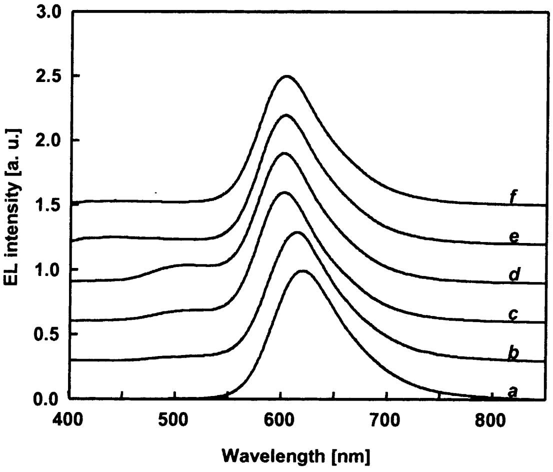

Fig. 2. Electroluminescence spectra of the ITO/PVK:PBD:$[Ru(dphphen)_3]^{2+}$/Alq_3/Al devices with $[Ru(dphphen)_3]^{2+}$ doping concentrations of 26% (*a*), 10% (*b*), 0.7% (*c*), and 0.5 % (*d*) at 16 V bias and a ITO/PVK:PBD:$[Ru (dphphen)_3]^{2+}$/BCP/Alq_3/Al device with 0.5 % doping concentration at 16 V (*e*) and 24 V (*f*) biases.

The ITO/PVK:TPD:PBD:$[Ru(dphphen)_3]^{2+}$/BCP/Alq_3/LiF/Al device shows the same electroluminescent mechanism and spectral property with the ITO/PVK:PBD :$[Ru(dphphen)_3]^{2+}$/BCP/Alq_3/Al device. Fig. 3 *a* shows the electroluminescence spectrum of the ITO/PVK:TPD:PBD: $[Ru(dphphen)_3]^{2+}$/BCP/Alq_3/LiF/Al device. Also peaked at about 602 nm, the emission is dominated by the $[Ru(dphphen)_3]^{2+}$ emitter by trapping charge carriers from the PVK:TPD:PBD blend matrix. The device has a turn on voltage of 9-10 V, and a maximum brightness of 820 cd/m^2 (0.68 cd/A) at 26 V.

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