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Efficient red organic electroluminescent devices based on trivalent europium complex obtained by designing the device structure with stepwise energy levels

Liang Zhou, Yunlong Jiang, Rongzhen Cui, Yanan Li, Xuesen Zhao, Ruiping Deng, Hongjie Zhang*

State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Renmin Street 5625, Changchun 130022, People's Republic of China

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ABSTRACT

In this study, we aim to further enhance the electroluminescence (EL) performances of trivalent europium complex Eu(TTA)₃phen (TTA=thenoyltrifluoroacetone and phen=1,10-phenanthroline) by designing the device structure with stepwise energy levels. The widely used bipolar material 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy) was chosen as host material, while the doping concentration of Eu(TTA)₃phen was optimized to be 4%. To facilitate the injection and transport of holes, MoO_3 anode modification layer and 4,4',4''-Tris(carbazole-9-yl)triphenylamine (TcTa) hole transport layer were inserted in sequence. Efficient pure red emission with suppressed efficiency roll-off was obtained attributed to the reduction of accumulation holes, the broadening of recombination zone, and the improved balance of holes and electrons on Eu(TTA)₃phen molecules. Finally, the device with 3 nm MoO_3 and 5 nm TcTa obtained the highest brightness of 3278 cd/m², current efficiency of 12.45 cd/A, power efficiency of 11.50 lm/W, and external quantum efficiency of 6.60%. Such a device design strategy helps to improve the EL performances of emitters with low-lying energy levels and provides a chance to simplify device fabrication processes.

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1. Introduction

Organic light-emitting devices (OLEDs) have attracted great interest throughout the world owing to their potential application in solid-state lighting and in full-color flat panel displays [1–3]. For commercial application, three primary colors of blue, green and red are basically required. However, pure red electroluminescence (EL) is still a challenge although many organic compounds showing red emission have been investigated [4-6]. Complexes of rare earth metals with organic ligands present an astounding class of emissive materials for OLEDs due to peculiarities of their luminescence properties [7-9]. Among these materials, trivalent europium (Eu³⁺) complexes are most interesting because they can emit highly monochromatic red light at around 612 nm corresponding to the ${}^{5}D_{0} \rightarrow {}^{7}F_{2}$ transition of Eu³⁺ ions [10,11]. Furthermore, theoretically they can offer 100% internal quantum efficiency because both singlet and triplet excitons are involved in the emission process [12,13]. Several groups have devoted to the synthesis of novel europium

http://dx.doi.org/10.1016/j.jlumin.2015.02.053 0022-2313/© 2015 Elsevier B.V. All rights reserved. complexes and the optimization of device structures [14–20]. Although significant improvements have been realized, EL performances of the devices based on europium complexes are still not satisfactory compared with the expected values.

Early, the main obstacles were the poor charge-carrier transporting ability and poor film-forming ability of most europium complexes [17,21], which brought difficulties for the design and fabrication of EL devices. In recent years, much important effort has been paid to doping europium complexes into host materials because doping method can prevent concentration quenching of the dyes' emission, avoid crystallization of the dye molecules as well as improve the injections of electrons and holes [18,19,22-25]. By codoping Alq₃ (tris(8-hydroxyquinoline) aluminum) and europium complex into light-emitting layer (EML) [14], EL devices with the brightness up to 2394.6 cd/m^2 were obtained. In a previous paper [19], we have realized the significant improvements of both current efficiency and power efficiency by controlling the injection of electrons. By designing the co-doped stacked device structure [20], Ma et al. have obtained the EL devices with the current efficiency up to 14.5 cd/A. However, the low-lying highest occupied molecular orbital (HOMO) levels of europium complexes make it difficult to choose proper host materials for device fabrication [14,21]. For







^{*} Corresponding author. fax: +86 43185685653. *E-mail address:* hongjie@ciac.ac.cn (H. Zhang).

example, the lowest unoccupied molecular orbital (LUMO) and HOMO levels of the well-known europium complex Eu(TTA)₃phen (TTA=thenoyltrifluoroacetone and phen=1,10-phenanthroline) are -3.27 and -6.3 eV, respectively [26]. We have experimentally demonstrated that Eu(TTA)₃phen molecules trap only electrons in Eu(TTA)₃phen doped CBP (4,4'-N,N'-dicarbazole-biphenyl) system. Consequently, electrons and holes are the majority carriers on Eu(TTA)₃phen and CBP molecules, respectively. Finally, the complicated carriers' distribution in EML strongly influences the EL processes, thus influencing the EL efficiency [15,19]. In addition, most europium complexes used in EL devices absorb only ultraviolet light, which usually leads to the incomplete Förster energy transfer from host materials to europium complexes [13,14,27].

On the other hand, the mechanisms of light generation in the doped devices are not fully understood, which partially restricts the optimization of doped devices based on europium complexes. In the past years, several groups paid attention to the detailed carrier trapping process in the doped devices, and certain progress was achieved [28–30]. For example, Nüesch and co-workers reported that hole or electron trapping ability of dye molecules depends on HOMO or LUMO levels of dye and host materials [31]. Previously, Wu et al. demonstrated this conclusion by fabricating a doped device with only electron trapping [32]. In a previous paper, we have discovered that the dominant EL mechanism of the devices based on the Eu(TTA)₃phen doped CBP system changes gradually from carrier trapping to Förster energy transfer with increasing current density due to the saturation of electron trapping [26]. In addition, the long excited state lifetime of europium complexes causes often the severe triplet-triplet annihilation, thus the rapid roll-off of EL efficiency [33].

In this work, a series of devices with stepwise energy levels were designed and fabricated to further improve the EL performances of Eu(TTA)₃phen. The bipolar host material 2.6-bis(3-(9H-carbazol-9-vl)) phenyl)pyridine (26DCzPPy) was chosen to broaden the recombination zone, thus to suppress the annihilation of triplet excitons. By inserting MoO₃ anode modification layer and 4,4',4''-Tris(carbazole-9-yl)triphenylamine (TcTa) hole transport layer, injection and transport of holes were significantly improved, which helps to facilitate the balance of holes and electrons on Eu(TTA)₃phen molecules. However, the device with Eu(TTA)₃phen doped TcTa system as supplementary EML displayed inferior EL performances because many electrons were trapped by Eu(TTA)₃phen molecules within TcTa layer, which destroys the balance of carriers within recombination zone. Ultimately, the device with 3 nm MoO₃ and 5 nm TcTa obtained the highest EL efficiencies and brightness. Our experimental results demonstrated that designing the device structure with stepwise energy levels is helpful in facilitating the injection and transport of holes, thus in enhancing the recombination probability of holes and electrons on the molecules of emitters with low-lying energy levels.

2. Experimental

Most of the organic materials used in this study were obtained commercially and used as received without further purification, while Eu(TTA)₃phen was synthesized and purified in our laboratory. Indium-tin-oxide (ITO) coated glass with a sheet resistance of 10 Ω /sq was used as the anode substrate. Prior to film deposition, patterned ITO substrates were cleaned with detergent, rinsed in de-ionized water, dried in an oven, and finally treated with oxygen plasma for 10 min at a pressure of 10 Pa to enhance the surface work function of ITO anode (from 4.7 to 5.1 eV) [34]. All organic layers were deposited with the rate of 0.1 nm/s under high vacuum ($\leq 3 \times 10^{-5}$ Pa). The EMLs were prepared by co-evaporating Eu(TTA)₃phen and host material from two individual sources, and the doping concentration

was modulated by controlling the evaporation rate of Eu(TTA)₃phen. MoO₃, LiF and Al were deposited in another vacuum chamber $(\le 8.0 \times 10^{-5} \text{ Pa})$ with the rates of 0.01, 0.01, and 1 nm/s, respectively, without being exposed to the atmosphere. The thicknesses of these deposited layers and the evaporation rate of individual materials were monitored in vacuum with quartz crystal monitors. A shadow mask was used to define the cathode and to make ten 9 mm² devices on each substrate. Current density-voltage-brightness (I-V-B) characteristics were measured by using a programmable Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a silicon photodiode. The EL and photoluminescence (PL) spectra were measured with a calibrated Hitachi F-7000 fluorescence spectrophotometer. The external quantum efficiency of EL device was calculated based on the photo energy measured by the photodiode, the wavelength of emission maximum (612 nm) of the EL spectrum, and the current pass through the device [35].

3. Results and discussion

The device structure and HOMO/LUMO levels diagram of the designed OLEDs are depicted in Fig. 1. Di-[4-(N,N-ditolyl-amino)phenyl]cyclohexane (TAPC) were used as hole transport and electron block layers (HTL/EBL) due to its high hole mobility $(1 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$ and high-lying LUMO level (-1.8 eV) [36]. 1,3,5-Tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) was used as hole block/electron transport layer (HBL/ETL) due to its low-lying HOMO level (-6.7 eV) and high electron mobility $(1 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$ [37]. A widely used europium complex Eu(TTA)₃phen was selected as an emitter [26,33,38], and 26DCzPPy was selected as a host material due to its wide energy gap and bipolar characteristic [39]. The molecule structures of Eu(TTA)₃phen and 26DCzPPy are also shown in Fig. 1. Within EML, most electrons would be well trapped by Eu(TTA)₃phen molecules due to its lower LUMO level (-3.27 eV)than that of 26DCzPPy (-2.6 eV); however, holes would preferentially situate on 26DCzPPy molecules due to its higher HOMO level (-6.1 eV) than that of Eu(TTA)₃phen (-6.3 eV). Based on previous results, appropriate accumulation of holes is helpful in balancing the distribution holes and electrons on Eu(TTA)₃phen molecules [19]. Therefore, how to facilitate the injection and



Fig. 1. Proposed energy level diagram of the devices used in this work and the molecule structures of Eu(TTA)₃phen and 26DCzPPy.

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