



An ideal host-guest system to accomplish high-performance greenish yellow and hybrid white organic light-emitting diodes



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ABSTRACT

Greenish yellow organic light-emitting diodes (GYOLEDs) have steadily attracted researcher's attention since they are important to our life. However, their performance significantly lags behind compared with the three primary colors based OLEDs. Herein, for the first time, an ideal host-guest system has been demonstrated to accomplish high-performance phosphorescent GYOLEDs, where the guest concentration is as low as 2%. The GYOLED exhibits a forward-viewing power efficiency of 57.0 lm/W at 1000 cd/m², which is the highest among GYOLEDs. Besides, extremely low efficiency roll-off and voltages are achieved. The origin of the high performance is unveiled and it is found that the combined mechanisms of host-guest energy transfer and direct exciton formation on the guest are effective to furnish the greenish yellow emission. Then, by dint of this ideal host-guest system, a simplified but high-performance hybrid white OLED (WOLED) has been developed. The WOLED can exhibit an ultrahigh color rendering index (CRI) of 92, a maximum total efficiency of 27.5 lm/W and a low turn-on voltage of 2.5 V (1 cd/m²), unlocking a novel avenue to simultaneously achieve simplified structure, ultrahigh CRI (>90), high efficiency and low voltage.

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1. Introduction

As one of the most promising candidates for the energy-saving light sources, organic light-emitting diodes (OLEDs) arouse both academic and industrial interest due to their extraordinary characteristics, such as high efficiency, light weight and flexibility [1–3]. Greenish yellow OLEDs (GYOLEDs) have the potential in numerous applications, including stage lights, signal lights and high quality RGBY-TV [4]. However, compared with red, green and blue OLEDs, only negligible attention for GYOLEDs has been paid, resulting in the fact that their development significantly lags behind [5–14]. On one hand, the efficiency of GYOLEDs is not high enough, since devices with forward-viewing power efficiency (PE) >55 lm/W at 100 cd/m² are rarely reported. For example, although Lu et al. introduced an effective interzone exciton transfer based structure to enhance the efficiency, their maximum efficiency is ~55 lm/W

[5]. On the other hand, the efficiency roll-off is afflicting the development of GYOLEDs. For instance, despite Hung et al. reported a GYOLED with a maximum PE of 53.2 lm/W by using a fluorene-based asymmetric bipolar universal host, the efficiency at 1000 cd/m² is only ~25 lm/W [6]. Moreover, the driving voltages are usually high, since no GYOLED can satisfy the demand of <3, 3.5 and 5 V at 100, 1000 and 10,000 cd/m², respectively.

In addition to the high efficiency, low efficiency roll-off and low voltage, ideal host-guest systems should also possess low cost, which can be accomplished by reducing the guest concentration [15–17]. This is because expensive noble metals (i.e., iridium and platinum) are needed to synthesize phosphors. Additionally, to establish high-performance OLEDs, the phosphorescent guest concentration is usually quite high (6%–25%) [4], further increasing the cost and limiting their long-term mass production. Therefore, reducing the guest concentration is one of the most effective ways to lower the cost, from the point of view of real commercialization. Towards this end, Kwon et al. used an ideal host-guest system to realize an efficient red phosphorescent OLED with 1% guest concentration [15]. Lee et al. studied the optimum guest concentration

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of red, green and blue OLEDs, suggesting that triplet hosts can show <5% concentrations by managing the energy levels of the hole and electron transport materials [16]. Qiu et al. utilized an ideal host-guest system to build a green phosphorescent OLED with 3% guest concentration, obtaining a maximum PE of 77 lm/W [17]. However, despite that greenish yellow emission is important, no ideal host-guest system has been documented to realize high-performance GYOLEDs.

Since there is no proper blue phosphor in terms of lifetime and color-stability, the utilization of hybrid WOLEDs, which refer to devices combining blue fluorophors with green–red/orange phosphors, is considered to be an alternative way due to their merits, like high efficiency, high color rendering index (CRI) and long lifetime [18–24]. Over the last few years, a large number of hybrid WOLEDs have been reported, such as managing both singlet and triplet excitons by inserting bipolar spacers [18,25], harvesting triplet excitons from fluorescent blue emitters with high triplet energies [24,26], using hole-type interlayers to ensure the blue emissions [27,28], designing single-emitting-layer (single-EML) structures to harvest excitons [29,30], and adopting bipolar mixed-hosts to suppress the mutual quenching between the fluorophor and phosphors [31,32]. In our previous works, we have also developed a series of hybrid WOLEDs by improving the device engineering [33–35]. However, despite a plenty of hybrid WOLEDs have been prepared, hybrid WOLEDs with ultrahigh CRIs (>90) are rarely reported. Particularly, in the case of three-color hybrid WOLEDs, only one device with an ultrahigh CRI of 91.2 has been realized [36]. However, the efficiency is very low, since the recorded PE is only 5.5 lm/W. Besides, the structure is complicated, since three EMLs with additional interlayer (IL) are necessary. Therefore, an effort is urgently needed to enhance the performance of three-color hybrid WOLEDs with ultrahigh CRIs.

In this paper, for the first time, an ideal host-guest system has been demonstrated to achieve high-performance GYOLEDs, where the guest concentration is as low as 2%. The device exhibits a forward-viewing PE of 57.0 lm/W at the illumination-relevant luminance of 1000 cd/m², which is the highest among GYOLEDs, to the best of our knowledge. Even at 10,000 cd/m², the PE can be as high as 30.0 lm/W, indicating an extremely low efficiency roll-off. Besides, the device exhibits low voltages. At 100, 1000 and 10,000 cd/m², the voltages are 2.95, 3.45 and 4.85 V, respectively. Then, the origin of the high performance is unveiled and it is found that the combined mechanisms of host-guest energy transfer and direct exciton formation on the guest are effective to furnish the greenish yellow emission. To further exploit this ideal host-guest system, we have incorporated it into a three-color hybrid WOLED by using a simplified structure (i.e., only two EMLs). For this WOLED, an ultrahigh CRI of 92 and a maximum total efficiency of 27.5 lm/W are achieved. Besides, the WOLED exhibits low voltages (i.e., 2.5 V for 1 cd/m²). Apparently, compared with the previous three-color hybrid WOLED with ultrahigh CRI [36], our device shows a remarkable step. In fact, the CRI of 92 is the highest among three-color WOLEDs, while 27.5 lm/W is one of the highest efficiencies among WOLEDs with ultrahigh CRIs (>90). Such superior properties would unlock a novel avenue to simultaneously achieve simplified structure, low voltage, high efficiency and ultrahigh CRI (>90).

2. Experimental

As displayed in Fig. 1, the structure of the GYOLED is ITO/HAT-CN (100 nm)/NPB (15 nm)/TCTA (5 nm)/Bepp₂: Ir(dmppy)₂(dpp) (35 nm, 2%)/Bepp₂ (30 nm)/LiF (1 nm)/Al (200 nm), where ITO is indium tin oxide, HAT-CN is 1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile [as a hole injection layer], NPB is N,N'-

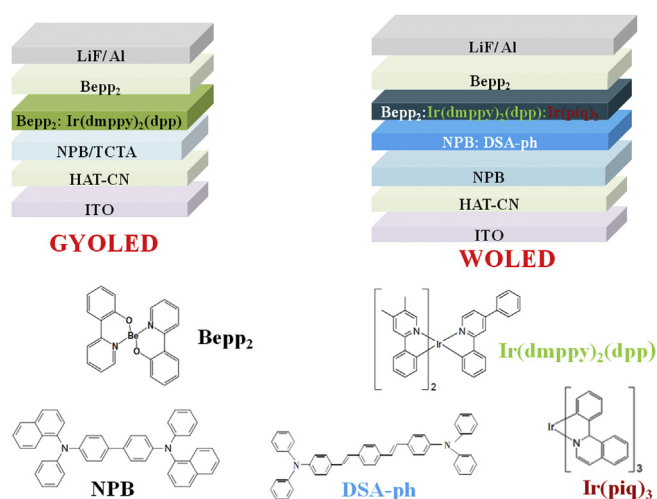


Fig. 1. Top: the schematic layer structure of processed GYOLED and WOLED. Bottom: the chemical structure of hosts and guests materials.

di(naphthalene-1-yl)-N,N'-diphenyl-benzidine, TCTA is 4,4',4''-tri(9-carbazoyl) triphenylamine, Ir(dmppy)₂(dpp) is bis(2-phenyl-4,5-dimethylpyridinato)[2-(biphenyl-3-yl)pyridinato] iridium(III) (as an greenish yellow emitter), Bepp₂ is bis[2-(2-hydroxyphenyl)pyridine] beryllium. All material layers were thermally deposited without breaking the vacuum at a base pressure of 2×10^{-7} Torr. In the deposition of the doping layers, deposition rates of both host and guest were controlled by their correspondingly independent quartz crystal oscillators. The devices were encapsulated immediately after preparation under a nitrogen atmosphere using epoxy glue and glass lids. The emission area of the devices is 3×3 mm² as defined by the overlapping area of the anode and cathode. The electroluminescent (EL) spectra, Commission International de l'Eclairage (CIE) color coordinates and CRI of packaged devices were obtained by a Konica Minolta CS2000 spectra system. The current density (J)–voltage (V)–luminance (L) characteristics were recorded simultaneously, using a computer-controlled source meter (Keithley 2400) and multimeter (Keithley 2000) with a calibrated silicon photodiode. All the measurements were carried out at room temperature under ambient conditions.

3. Results and discussions

3.1. The development of GYOLEDs

3.1.1. Devices design strategies

To achieve high-performance GYOLEDs, several design strategies are employed. First, one of the most crucial things is the selection of an appropriate host to give off greenish yellow emission. We used Bepp₂ as the host, since the triplet energy (T_1) of Bepp₂ is 2.6 eV [25], high enough to satisfy the demand of the guest Ir(dmppy)₂(dpp) ($T_1 < 2.25$ eV) [37], which can prevent the reverse energy transfer from the guest to the host and confine triplets in the whole EML, effectively consuming the electrically generated triplets contributing to emission [22].

Then, a double hole transport layer (HTL) NPB (15 nm)/TCTA (5 nm) is used to give an effective hole transport due to their high hole mobilities [3]. Besides, since the lowest unoccupied molecule orbital (LUMO) of TCTA is 2.3 eV [25], a large barrier (0.3 eV) exists between the HTL and EML, which can prevent the electron leakage. Additionally, triplets in the EML can be well confined owing to the high T_1 of TCTA (2.8 eV) [25], ensuring the high performance [35].

Next, Bepp₂ is used as an excellent electron transport layer

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