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Regulating charges and excitons in simplified hybrid white organic light-emitting diodes: The key role of concentration in single dopant host–guest systems

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ABSTRACT

A series of high-performance hybrid white organic light-emitting diodes (WOLEDs) with simplified structures are developed by systematically investigating the influence of dopant concentrations. An ultrahigh color rendering index of 88 and a peak forward-viewing power efficiency of 44.9 lm/W are achieved in the single-emitting-layer WOLED with a low concentration (0.1%) and a moderate concentration (1%), respectively. To investigate the effect of high concentration (4%), a dual-emitting-layer device is realized, achieving an efficiency of 15.8 lm/W (1000 cd/m^2). Besides, the devices show extremely stable color with a Commission Internationale de L'Eclairage (CIE*x*, *y*) variation of $\Delta(x, y) \leqslant (0.008, 0.008)$ during a large range of luminance. Furthermore, the origin of the color-stability and working mechanism of the devices are discussed, which can be attributed to the multifunctional dopant which reduces charge mobilities together with a suitable hole transport material which exhibits strong electron- and exciton-blocking ability.

1. Introduction

White organic light-emitting diodes (WOLEDs) are now entering mainstream display markets and are also being intensively explored for the next-generation lighting applications owing to their high efficiency, flexibility and low cost [1,2]. Generally, three approaches have been attempted to realize WOLEDs over the last few years according to the used emissive materials, including all-phosphorescent WOLEDs [3], all-flourescent WOLEDs [4] and the so-called hybrid WOLEDs which are based on hybrid (phosphorescent (P) and flourescent (F)) emitters [5–7]. Since P emitters can allow for a conversion of up to 100% of injected charges into emitted photons (both singlet and triplet excitons can be harvested), resulting in a theoretical internal quantum efficiency of unity [8], highly efficient P materials are usually imperative to boost the efficiency of WOLEDs. Unfortunately, although most of the highly efficient WOLEDs can be realized based on all-phosphors, there is still no proper blue P material in terms of operational lifetime and color-stability until now, limiting the development of all-phosphor devices [7]. Recently, to solve the above conflicts, the hybrid WOLEDs, which combine stable blue F emitters with efficient P green–red/orange emitters, are considered to be an effective and favourable way due to their merits, such as high efficiency, stable color and long lifetime [5–7]. In the case of hybrid WOLEDs, Sun et al. took the first step to develop a smart

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device by employing the combination of stacked F-spacer-P-P-spacer-F emitters in an ambipolar host material 4,4′-bis(N-carbazolyl)-1,1′-biphenyl [5], achieving a maximum total power efficiency (PE) of 37.6 lm/W. However, it is noted that this sophisticated device needs somewhat complicated fabrication processes and the efficiency is unsatisfactory. To address these issues, elaborate device structures and elegant materials have to be developed. In terms of device structures, an alternative way is the use of a single dopant host–guest strategy, which needs only two complementary colors (blue–orange or blue–red) to produce white light and eliminates the spacer located between F and P emitters. Consequently, this strategy is expected to simultaneously simplify device configuration (reducing the number of dopants and structural heterogeneities inherent in the multiple-color band architectures, for example), lower the cost as well as achieve high efficiency.

As a matter of fact, some endeavors have been made by using this very simple way. On the one hand, this simple method can be realized by developing dual-emitting-layer (dual-EML) devices, avoiding using the additional spacer which often cause high driving voltages, complicated fabrication processes and the undesirable emissions since a substantial amount of generated excitons are consumed by the spacer to be emitted inefficiently [6,9]. For example, Anzenbacher et al. reported a simple hybrid WOLED with two EMLs: an undoped host layer (blue EML) coupled with the host region doped with an orange dopant (orange EML), achieving a low turn-on voltage of 3 V without p–i–n structures [10]. Zhang et al. employed this strategy to realize an efficient WOLED based on an ambipolar and electric-field independent F emitter, achieving a PE of 14.1 lm/W at 1000 cd/m^2 [11]. More impressively, the single dopant host–guest strategy can be attained by constructing single-EML hybrid WOLEDs, which can not only avert the utilization of spacers but also achieve high efficiency. For example, Leo et al. took the first step to realize a two-color single-EML hybrid WOLED with small molecules, yielding a PE of 10.5 lm/W at 1000 cd/m^2 [12]. Tao et al. developed a two-color (blue flourescence and red phosphorescence) single-EML WOLED, achieving a maximum PE of 4.7 lm/W [13]. Hung et al. fabricated a deep-blue electrofluorescence/yellow-green electrophosphorescence singly doped device, obtaining a PE of 12.8 lm/W [14]. Peng et al. realized a deep-blue flourescence/orange phosphorescence single-EML device with a peak PE of 48.8 lm/W [15]. Most recently, Ye et al. proposed a single-EML hybrid WOLED by using a bipolar F material as the host of orange dopant and the sky-blue emitter, achieving a maximum total PE of 67.2 lm/W at a low luminance ($\sim$6 cd/m^2) and a total PE of 33.5 lm/W at 1000 cd/m^2 [16]. Based on these facts, it can be concluded that the use of single dopant host–guest strategy is a rather simple but remarkably promising approach to develop highly efficient hybrid WOLEDs. However, although WOLEDs using this strategy can be realized, the color-stability of these devices has been usually overlooked so far and there is still much room to further enhance the device efficiency. Moreover, despite the dopant concentration plays a crucial role in the performance of this type of hybrid WOLEDs, no detailed investigation has been documented to study its effect and hence the working mechanism of these devices is still unclear.

It is deserved to point out that the motivation of the single dopant host–guest hybrid WOLEDs is the introduction of appropriate blue F emitters. In the case of the blue fluorophor with low triplet energy, triplet excitons can transfer from phosphor to fluorophor, resulting in the non-radiative decay. That is, triplets in the phosphor can be quenched by the fluorophor, leading to no white emission. Therefore, a key feature of designing the single dopant host–guest hybrid WOLEDs is using blue F emitters with relatively high triplet energies, which can not only avoid the utilization of spacers but also ensure the white emission since triplets in the blue fluorophor can be effectively harvested by the complementary color phosphor. In this paper, we use $Bepp_2$ as the blue fluorophor, because: (i) the triplet energy of $Bepp_2$ is 2.6 eV [7], high enough to satisfy the demand of green, orange and red P emitters, such as $Ir(ppy)_3$ (2.4 eV) [7], $(bzq)_2Ir(dipba)$ (2.2 eV) [15], $(fbi)_2Ir(acac)$ (2.2 eV) [2] and $Ir(MDQ)_2(acac)$ (2.0 eV) [7]. (ii) $Bepp_2$ can exhibit a deep blue color with a peak emission of 445 nm and 3.43 lm/W [17]. Taking these factors into account, we have comprehensively investigated the effect of dopant concentration in the single dopant host–guest systems and achieved a series of high-performance WOLEDs by varying the dopant concentration from 0.1% to 4%. At a low concentration of 0.1%, a two-color single-EML WOLED with an ultrahigh color rendering index (CRI) of 88 is achieved. At a moderate concentration of 1%, the maximum forward-viewing current efficiency (CE) and PE of the single-EML WOLED are as high as 37.6 cd/A and 44.9 lm/W, respectively. At a high concentration of 4%, the dual-EML device with the forward-viewing CE and PE of 18.6 cd/A and 15.8 lm/W are obtained at 1000 cd/m^2, respectively. Besides, it is found that these devices show extremely stable color with a Commission Internationale de L'Eclairage (CIEx, y) variation of $\Delta(x, y) \leqslant (0.008, 0.008)$ during a large range of luminance (100–5000 cd/m^2). Moreover, the origin of the color-stability and working mechanism of the high-performance devices are discussed, which can be attributed to the multifunctional dopant which effectively reduces charge mobilities (both holes and electrons) combined with a suitable hole transport material which exhibits strong electron- and exciton-blocking ability. Such superior results not only indicate that the dopant concentration plays a key role in the device properties based on single dopant host–guest systems, but also demonstrate a significant step towards extremely high-performance WOLEDs with extremely simplified structures.

2. Experimental

As depicted in Fig. 1, the configuration of single-EML hybrid WOLEDs is ITO/MeO-TPD: F4-TCNQ (100 nm, 4%)/TAPC (20 nm)/$Bepp_2$: $(fbi)_2Ir(acac)$ (25 nm, X%)/$Bepp_2$ (25 nm)/LiF (1 nm)/Al (200 nm), where ITO is indium tin oxide, MeO-TPD is N,N,N′,N′-tetrakis(4-methoxyphenyl)-benzidine, F4-TCNQ is tetrafluoro-tetracyanoqino dimethane, TAPC is 1-bis[4-[N,N-di(4-tolyl)amino]phenyl]-cyclohexane, $(fbi)_2Ir(acac)$ is bis(2-(9,9-diethyl-9H-fluo-