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Hybrid white organic light emitting diodes with low efficiency roll-off, stable color and extreme brightness

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ABSTRACT

Highly efficient and bright hybrid white organic light emitting diodes (WOLEDs) based on simple architectures have been successfully fabricated and characterized. The optimized device can reach a maximum forward-viewing power efficiency (PE) of 20.2 lm/W, a peak forward-viewing current efficiency (CE) of 30.7 cd/A, an extremely high brightness of 95,683 cd/m^2, and a Commission International de l'E clairage chromaticity coordinates of (0. 436, 0.425) at 12 V. Even at the illumination-relevant brightness of 1000 cd/m^2, a forward-viewing PE of 17.0 lm/W and CE of 30.7 cd/A are obtained. Moreover, it is found that the device not only suffers slight efficiency roll-off but also exhibits a stable color during a large range of brightness, indicating that the device can satisfy the future commercial requirements. Undoubtedly, the results will be beneficial to the design of both material and device architecture for high-performance WOLEDs and next-generation solid-state lighting sources.

1. Introduction

White organic light emitting diodes (WOLEDs) are of particular significance in our life owing to their various potential applications in solid-state lighting and flat panel displays. They have advantages of excellent electroluminescence (EL) performances, good flexibility, low cost, and batch fabrication [1]. Currently, three kinds of WOLEDs have been reported, including all-phosphorescent WOLEDs [2], all-flourescent WOLEDs [3] and hybrid WOLEDs (HWOLEDs) which are based on hybrid (flourescent and phosphorescent) emissive materials technology [4]. Among these white devices, the utilization of phosphorescent emitters is desirable because phosphorescent emitters enable an internal efficiency as high as 100% converting both singlet and triplet excitons into photons and make a fourfold increase in efficiency compared to that achievable in singlet-harvesting flourescent emitters [1,5]. Unfortunately, there is no satisfactory phosphorescent blue (B) material in terms of operational lifetime and color-stability up till now, limiting the development of all-phosphor-doped devices [6]. In recent years, researchers have diverted their attention to solve this issue by creating the so-called HWOLEDs [4,7], which combine stable flourescent B emitters with phosphorescent green (G)-red (R)/orange (O) emitters because of their advantages, such as high efficiency and long lifetime. Sun et al. took the first step to bring the conceptually highly efficient HWOLEDs into reality (a maximum forward-viewing power efficiency (PE) is 22.1 lm/W), which is the milestone in HWOLEDs [4]. However, their devices not only showed a relatively complex structure (using B/R+G/B emitters) but also exhibited a rather serious efficiency roll-off (the forward-viewing PE is 14 lm/W at 500 cd/m^2). To simplify the structures, an alternative approach is to replace the G+R emitters with R emitters or O emitters. In fact, a large number of reports have been proposed. For example, Seo et al. utilized the B/R/B architecture, following the same principle as Sun's work, to obtain a high efficiency HWOLED (33.2 lm/W, 38.0 cd/A), however, their device suffered a quite serious efficiency roll-off [8]. Bhansali et al. used the B/O/B structure to achieve a simplified HWOLED, however, the device efficiencies are very low (5.7 lm/W, 13.0 cd/A) [9]. Wang et al. also tried to use the B/O/B structure to obtain a white color, however, they only achieved an unsatisfactory result (~7 cd/A) [10]. Based on these facts, there is still much room to further enhance the performance of HWOLEDs with simple structure to satisfy future commercial requirements.

Herein, to solve issues as mentioned above, we demonstrate a simple HWOLED architecture, using 'B/O/B' emitters, namely B emitter/spacer/O emitter/spacer/ B emitter. The optimized device

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shows a peak brigthness of 95,683 cd/m^2 and a maximum forward-viewing PE of 20.2 lm/W. Moreover, the maximum forward-viewing current efficiency (CE) of 30.7 cd/A at 700 cd/m^2 is obtained, which suffers no roll-off to a rather high brightness of 1100 cd/m^2, and even at an ultra high brightness of 5000 cd/m^2, the CE rolls off slightly to 28.6 cd/A. Additionally, a Commission International de l'Eclairage (CIE) chromaticity coordinates of (0. 436, 0.425) is observed at 12 V, and the device only suffer a negligible color change during a large range of brightness (500–6000 cd/m^2). To the best of our knowledge, both PE and CE of the optimized device are the highest values in HWOLEDs based on B/O/B structures. Moreover, the optimized device exhibits the highest efficiencies (30.7 cd/A, 17.0 lm/W) in HWOLEDs based on B/R+G/B, B/R/B or B/O/B architectures at the practical brightness of 1000 cd/m^2 in the literature so far.

2. Experimental

As vividly shown in Fig. 1, the configuration of HWOLEDs is ITO/MeO-TPD: F4-TCNQ(100 nm, 4%)/NPB(20 nm)/ MADN: DSA-ph(3, 6, 10, 15 nm, 7%)/ CBP (2 nm)/CBP: (MPPZ)$_2$Ir(acac) (20 nm, 8%)/CBP (2 nm)/MADN: DSA-ph(30 nm, 7%)/Bebq$_2$(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, NPB is *N*,*N*'-di(naphthalene-1-yl)-*N*,*N*'-diphenyl-benzidine, Bebq$_2$ is bis(10-hydroxybenzo[*h*] quinolinato) beryllium complex, flourescent material *p*-bis(*p*-*N*,*N*-di-phenyl-aminostyryl) benzene (DSA-ph) as a B guest, doped into a 2-methyl-9,10-di(2-naphthyl)anthracene (MADN) host, and phosphorescent material iridium (III) diazine complexes (MPPZ)$_2$Ir(acac) which can yield very high-efficiency O emission as an O guest [11], doped into a 4,4-*N*,*N*-dicarbazolebiphenyl (CBP) host, to achieve balanced white light. The fabricated devices were grown on precleaned ITO glass substrates, and then 100 nm thick MeO-TPD: F4-TCNQ (as a hole injection layer), 20 nm thick NPB (as a hole transporting layer), *x* nm thick MADN: DSA-ph (as a B-light-emission layer), the undoped 2 nm thick CBP (as a spacer), 20 nm thick CBP: (MPPZ)$_2$Ir(acac) (as an O-light-emission layer), the undoped 2 nm thick CBP (as a spacer), 30 nm thick MADN: DSA-ph (as a B-light-emission layer), 25 nm thick Bebq$_2$ (as an electron transporting layer), and finally 1 nm thick LiF and 200 nm thick Al cathode were evaporated in sequence. All material layers were thermally deposited without breaking the vacuum at a base pressure of 2×10^{-7} Torr. All the organics were evaporated at a rate in the range of 0.007–0.1 nm/s, and the metallic

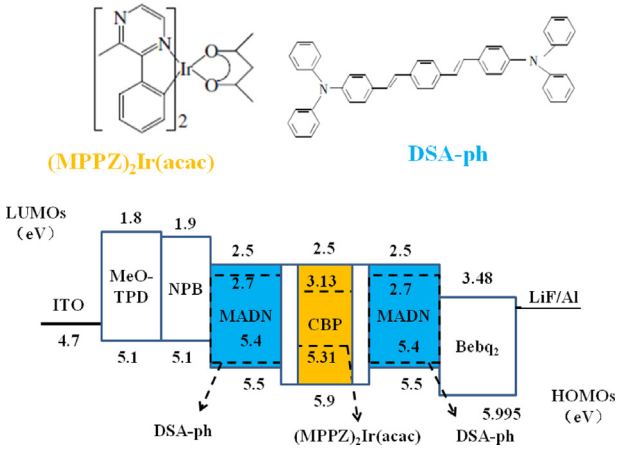

Fig. 1. Top: The chemical structure of emissive dopants. Bottom: Proposed energy-level diagram of HWOLEDs, showing the highest occupied and lowest unoccupied molecular orbital energies relative to the vacuum level. The energy values are taken from the representative works [11–13,15].

cathode was evaporated at a rate of 0.07–0.7 nm/s. The evaporation rates were monitored by frequency counter and calibrated by Ambios XP2. The devices were encapsulated immediately after preparation under a nitrogen atmosphere using epoxy glue and glass lids. The EL spectra and CIE color coordinates of packaged devices were obtained by a Konica Minolta CS2000 spectra system. The emission area of the devices is 3×3 mm^2 as defined by the overlapping area of the anode and cathode. The brightness–current density–voltage 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 discussion

Since improvement of both efficiency and stability in WOLEDs can be expected by rational design of the structure, several design strategies have been employed to achieve high efficiency, brightness together with stable EL spectra for our HWOLED with B/O/B structure. First, to lower manufacturing cost, device architectures should be simple. Thus, we utilized two complementary colors to furnish white emission, surprisingly, performances of the simple device are very impressive.

Second, to maximize exciton formation and emission in WOLEDs, it is necessary to optimize the concentration in the host-guest doped system. Therefore, monochromatic devices with various concentrations of dopant were tested to determine the best concentrations. As the results showed in our lab, it was found that the optimized concentrations for blue guest DSA-ph and orange dopant (MPPZ)$_2$Ir-(acac) were fixed at 7% and 8%, respectively.

Next, since the highest occupied molecular orbital (HOMO) of NPB is 5.1 eV [12] and the HOMO of CBP is 5.9 eV [13], a large energy barrier of 0.8 eV between NPB and CBP is formed, as shown in Fig. 1. As a result, holes may be prevented to hopping from NPB to CBP due to the fact that the current flow is limited by the injection of carriers when the injection energy barrier is higher than 0.3–0.4 eV [14]. Given this fact, we selected two different hosts instead of one host for B and O dopants, respectively. The HOMO of B host MADN is 5.5 eV [15] which locates between that of NPB and CBP, forming a step barrier which may benefit the holes to hop from NPB to CBP [16]. The step barrier can also reduce the accumulation of holes at the interfaces of NPB and CBP, resulting in low efficiency roll-off, as demonstrated by the achieved results (will be shown below). Moreover, it has been showed that MADN is a more suitable host for the DSA-ph guest [17]. It is noted that this strategy has not been used for previous HWOLEDs [4,9,18].

On the other hand, since additional reductions in efficiency could possibly occur when the thickness of the electron transporting layer is <25 nm due to exciton quenching at the metal cathode [19], we fix the Bebq$_2$ thickness at 25 nm.

In addition, by placing 2 nm undoped CBP between flourescent and phosphorescent emitters as the spacer, both singlet and triplet excitons can be effectively harvested to produce white light [4], guaranting the high-performance of the device. It is noted that this strategy has not been applied to the previous reports [10,18].

Last but not the least, MADN exhibits bipolar transport property, however, the electron mobility is relatively higher than hole mobility [20]. Consequently, it is essential to optimize the thickness of B emissive layer nearby the anode so that the exciton-recombination probability can be enhanced effectively. Note that this strategy has not been employed to the previous device [8].

Based on these considerations, we put forward a simple HWOLED, simultaneously achiving high efficiency, extreme brightness and stable color.