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Investigation and optimization of each organic layer: A simple but effective approach towards achieving high-efficiency hybrid white organic light-emitting diodes

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ABSTRACT

A highly efficient hybrid white organic light-emitting diode based on a simple structure has been successfully fabricated and characterized. By systematically investigating the influence of the emissive layer thickness, electron transporting layer thickness, spacer and hole transporting layer, the forward-viewing current efficiency and power efficiency of the resulting device without any out-coupling schemes or n-doping strategies can be as high as 59.4 cd/A and 58.4 lm/W, respectively. Besides, a Commission International de l'Eclairage of (0.412, 0.393) and a color rendering index of 60 are obtained at the current density of 11 mA/cm^2. Through the optimization and investigation, the origin of this unique device is explored comprehensively. Undoubtedly, such presented results will be beneficial to the design of both material and device architecture for ultra high-performance white organic light-emitting diodes.

1. Introduction

White organic light-emitting diodes (WOLEDs) are now entering mainstream display markets and are also being explored for next-generation lighting applications owing to their high efficiency, light weight and low cost [1]. Generally, WOLEDs can be classified into three kinds according to the used emissive materials, including all-phosphorescent WOLEDs [2], all-fluorescent WOLEDs [3] and hybrid WOLEDs which are based on hybrid (fluorescent (F) and phosphorescent (P)) emissive materials technology [4]. Among them, the use of P emitters is desirable because P emitters can harvest both singlet and triplet excitons. Therefore, devices with internal quantum efficiencies up to 100% have been demonstrated, which corresponds to a fourfold increase in efficiency compared to that achievable in singlet-harvesting F emitters [5]. However, there is no perfect blue P material in terms of operational lifetime and color-stability up till now, limiting the development of all-phosphor-doped devices [6,4c]. Recently, several publications have been devoted to address the conflict by creating the so-called hybrid WOLEDs [4], which combine stable F blue emitters with efficient P green–red/orange emitters due to their merits, such as high efficiency, stable color and long lifetime. Sun et al. have taken the first step to bring the conceptually new hybrid WOLEDs into reality by utilizing the combination of stacked F–P–P–F emitters in an ambipolar host [4a]. The significant merit of the idea

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is that all singlet and triplet excitons can be effectively harvested along completely independent channels and hence nearly resonant energy transfer from the conductive host to dopants for both the singlet and triplet energy levels can be realized. Meanwhile, the 0.5–1.0 eV exchange energy losses resulting from intersystem crossing from the host singlet into a blue phosphor triplet state are eliminated by this exciton-confined architecture, allowing for increased power efficiency (PE) [4b]. However, this elegant device needs somewhat complicated fabrication processes and the efficiency is unsatisfactory (~22.1 lm/W), limiting its further practical applications. To improve these issues, an alternative approach is using the stacked P–F emitters to achieve highly efficient hybrid WOLEDs. In fact, tremendous efforts have been devoted to the pursuit of this simple way [7]. For example, Yan et al. described the construction of hybrid WOLEDs based on a P–F dual-emitting architecture by using a new family of electrophosphorescent platinum(II) emitters, achieving a PE of 18.4 lm/W [7a]. Wong et al. reported a two-element hybrid WOLED with the simple P–F structure by utilizing a novel heteroleptic orange-emitting P iridium (III) complex, obtaining an efficiency of 13.5 lm/W [7b]. Ma et al. realized the P–F concept by developing combinations of exciton-managed orange-phosphorescence and sky blue-fluorescence, achieving a PE of 24.9 lm/W [7c]. Zhao et al. also fabricated the P–F architecture to produce white light by selecting an appropriate spacer [7d]. Based on these facts, it can be concluded that the utilization of stacked P–F emitters is a very promising and simple way to construct efficient hybrid WOLEDs. However, in spite the efficiency of hybrid WOLEDs with P–F structures has experienced step-by-step increase over the past years and has already exceeded that of F–P–P–F structures, the efficiency (<30 lm/W) is still not high enough to satisfy the commercial requirements [7]. Therefore, there is still much room to further enhance the performance of hybrid WOLEDs by using this simple architecture. Moreover, although each organic layer of the hybrid WOLED has a great influence on enhancing the efficiency of P–F device, no systematic investigation has been documented to study their effects.

In this paper, we have successfully developed a highly efficient hybrid WOLED comprising an orange phosphor and a sky blue fluorophore. Particularly, we have systematically investigated the influence of the emissive layer (EML) thickness, electron transporting layer (ETL) thickness, spacer and hole transporting layer (HTL), from which a simple but effective way towards achieving highly efficient device has been vividly illustrated. Through optimization and investigation of the devices, the maximum forward-viewing current efficiency (CE) and PE of the simple P–F device are as high as 59.4 cd/A and 58.4 lm/W at low current density, respectively. Besides, a Commission International de l'Eclairage (CIE) of (0.412, 0.393) at the current density of 11 mA/cm^2 is also obtained, corresponding to a color rendering index (CRI) of 60. Encouraged by the ultra-high efficiencies, the origin of this unique device is explored comprehensively, which will undoubtedly be helpful for the rational design of both material and device structure for high-performance WOLEDs.

2. Experimental

Fig. 1 depicts the studied hybrid WOLEDs with the configuration of ITO/MeO–TPD: F4–TCNQ (100 nm, 4%)/HTL (20 nm)/CBP: $(MPPZ)_2Ir(acac)$ (*X* nm, 8%)/spacer (3 nm)/MADN: DSA-ph (20 nm, 1%)/TmPyPB (*Z* nm)/LiF (1 nm)/Al (200 nm), where ITO is indium tin oxide, MeO–TPD is N, N′,N′-tetrakis(4-methoxyphenyl)-benzidine, F4–TCNQ is tetrafluoro-tetracyanoqino dimethane, TmPyPB is 1,3,5-tri(*m*-pyrid-3-yl-phenyl)benzene, spacer is either N,N′-di(naphthalene-1-yl)-N,N′-diphenyl-benzidine (NPB) or 4,4-N,N-dicarbazolebiphenyl (CBP). F material p-bis (p-N,N-di-phenyl-aminostyryl) benzene (DSA-ph) as a blue guest, was doped into 2-methyl-9,10-di(2-naphthyl)anthracene (MADN), and P material iridium (III) diazine complexes $(MPPZ)_2Ir(acac)$, was doped into a CBP host. The cleaned ITO was loaded into an evaporator, and then 100 nm thick MeO–TPD: F4–TCNQ, 20 nm thick HTL, *X* nm thick CBP: $(MPPZ)_2Ir(acac)$ (*X* = 10, 25 or 40), 3 nm thick spacer, 20 nm thick MADN: DSA-ph, *Z* nm thick TmPyPB (*Z* = 20,40 or 60), 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. In the deposition of the doping layers, deposition rates of both host and guest were controlled with their correspondingly independent quartz crystal oscillators. The devices were encapsulated immediately after preparation under a nitrogen atmosphere using epoxy glue and glass lids. The electroluminescent (EL) spectra, CIE color coordinates and CRI 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 luminance–current density–voltage characteristics were recorded simultaneously, using a computer-controlled source meter (Keithley 2400) and multimeter (Keithley 2000) with a calibrated silicon photodiode. The detailed measurement external quantum efficiency (EQE) of devices followed well-established processes and as reported elsewhere [8]. All the measurements were carried out at room temperature under ambient conditions.

3. Results and discussions

3.1. The impact of orange EML-thickness

Since the thickness of EML is significant to WOLEDs, an inappropriate thickness of EML can result in rather poor performance [9]. Therefore, we first examine the influence of phosphor-layer thickness on the EL efficiency and spectrum of the device by fabricating three hybrid WOLEDs, that is, W_{11} with 10 nm thick orange EML, W_{12} with 25 nm thick orange EML, while 40 nm thick orange EML for W_{13}. The structure of these devices is ITO/MeO–TPD: F4–TCNQ(100 nm, 4%)/NPB(20 nm)/CBP: $(MPPZ)_2Ir(acac)$ (10, 25, 40 nm, 8%)/NPB (3 nm)/MADN: DSA-ph (20 nm, 1%)/TmPyPB (40 nm)/LiF (1 nm)/Al (200 nm). The spectra and efficiencies for devices with different thicknesses of orange EML are shown in Fig. 2 and Table 1 lists the performances of these devices.