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Solution processed single-emissive-layer white organic light-emitting diodes based on fluorene host: Balanced consideration for color quality and electroluminescent efficiency

Shang-hui Ye [a, *], Tian-qing Hu [a], Zhou Zhou [a], Min Yang [a], Qun-bo Mei [a], Yun-qi Liu [b, **], Li-ping Liu [a], Bang-cheng Zhai [a], Zhen-hong Jia [a], Wei Huang [a, c]

[a] Key Laboratory for Organic Electronics and Information Displays & Institute of Advanced Materials (IAM), Jiangsu National Synergetic Innovation Center for Advanced Materials (SICAM), Nanjing University of Posts & Telecommunications, 9 Wenyuan Road, Nanjing 210023, China
[b] Beijing National Laboratory for Molecular Sciences (BNLMS), Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China
[c] Key Laboratory of Flexible Electronics (KLOFE) & Institute of Advanced Materials (IAM), Jiangsu National Synergetic Innovation Center for Advanced Materials (SICAM), Nanjing Tech University (NanjingTech), 30 South Puzhu Road, Nanjing 211816, China

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Cost-effective fabrication of white organic light-emitting diodes (WOLED) is meaningful toward commercial application of environment-friendly solid-state lighting sources. Electroluminescent efficiency and color quality are two opposite performance characteristics facing solution processed WOLEDs requiring balanced consideration. Herein, a recently synthesized molecule of 4,4'-(9,9'-(1,3-phenylene) bis(9*H*-fluorene-9,9-diyl))bis(*N*,*N*-diphenylaniline) (DTPAFB) is introduced as a host material for solution processed all-phosphor WOLEDs, embracing four well-known molecules which are blue iridium (III) bis(2-(4,6-difluorophenyl)pyridinato-N,C^2)(picolinate) (FIrpic), green iridium (III) bis[2-(2-pyridinyl-*N*) phenyl-C](2,4-pentanedionato-O^2,O^4) [$Ir(ppy)_2(acac)$], and orange iridium (III) bis(2-phenyl-benzothiazole-C^2,N)(acetylacetonate) [$Ir(bt)_2(acac)$] plus a home-made red phosphor of iridium (III) tris(1-(2,6-dimethylphenoxy)-4-(4-chlorophenyl)phthalazine) [$Ir(MPCPPZ)_3$]. Illumination quality white light with high brightness, high efficiency, suitable correlated color temperature (CCT), high color-rendering index (CRI), and stable electroluminescent (EL) emission is obtained. A stable white emission with a CRI over 70, Commission Internationale de L'Eclairage (CIE) of (0.37, 0.42), and high EL efficiency of 19.6 lm W^{-1} at high luminance of 2000 cd m^{-2} for blue/orange complementary color WOLEDs is demonstrated. The optimized red/green/blue three primary color WOLEDs show improved CRI up to 81, moderate high efficiency of 25.8 cd A^{-1}, 14.4 lm W^{-1}, and EQE of 13.9%. Furthermore, the red/green/blue/orange four primary color WOLEDs show the optional balance between color quality and EL efficiency with high CRI of around 81–83 and medium CCT of 3755–3929 K which is warm and soft to human eyes. At an illumination relevant luminance of 1000 cd m^{-2}, the total power efficiency reaches 33.6 lm W^{-1}, and still remains 30.2 lm W^{-1} at 3000 cd m^{-2}, approaching the efficiency of state-of-the-art fluorescent-tube (40–70 lm W^{-1}), potentially suitable as an environment-friendly solid-state lighting source. This work indicates that developing high performance host materials and highly efficient phosphors and carefully combining them with common phosphors is an effective way toward high performance WOLEDs.

1. Introduction

White organic light-emitting diodes (WOLEDs) featured by high-quality color, energy-saving properties, ease of large-area fabrication, bio-friendly diffusive warm light, and freedom from radiation damage have witnessed the emerging and dramatic development for commercial lighting panel models. It has been generally recognized as the most promising candidate as a next generation lighting technique [1].

As for a solid-state lighting source, two primary parameters must be taken into account: one is the electroluminescent (EL)

* Corresponding author.
** Corresponding author.
E-mail addresses: iamshye@njupt.edu.cn (S.-h. Ye), liuyq@iccas.ac.cn (Y.-q. Liu).

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efficiency, and the other is color quality.

To obtain high efficiency, phosphors are always introduced into the emissive layer due to the spin-coupling effect, which gives them the ability to harvest both singlet and triplet excitons leading to nearly 100% internal quantum efficiency [2–4]. To achieve white emission, more than one luminophore is utilized. The dominating approach is to combine the electroluminescence from blue, green, and red luminophores which leaves some freedom in how the luminophores are combined, i.e., whether they are blended in a single layer, separated into different layers of the same OLEDs, or contained in several formally independent devices each of which emits different colors [5]. In principle, tandem WOLEDs by red, green, and blue (RGB) emitters can improve the operation stability, but the spectral instability due to high energy exciton of the blue emitter makes it problematic [6–8]. In contrast, single-emissive-layer from RGB blend is more advantageous for stable white electroluminescence emissions change as the operating voltage increases, free from problematic recombination zone-shifting [9]. This leads to brightness-dependent color-shifts [10]. In regards to the components of the emissive layer, blue and orange (BO) or yellow (BY) two complementary color WOLEDs are generally more efficient than the RGB three primary color types. In this system the phosphors are commonly dispersed into a host material to avoid concentration annihilation and triplet-triplet quenching, and thus a suitable combination of highly efficient phosphors with a proper host material is equally important [11].

Apart from electroluminescent (EL) efficiency, color quality is another important factor to be considered, which involves EL spectrum, color-rendering index (CRI), Commission Internationale de L'Eclairage chromaticity coordinates ($CIE_{x,y}$), and correlated color temperature (CCT). The EL spectrum should cover the entire visible region (400–700 nm), CRI around 70 for general lighting source, $CIE_{x,y}$ in the white color region, and CCT between 2500 K and 6000 K [12]. In general, WOLEDs based on BO complementary emitters always exhibit moderate color quality with intermediate CRI in the range of 52–65, compared with RGB three primary color devices. Obviously, the more primary components the emitting layer contains, the better the EL color quality will become. Therefore, red, green, blue, and orange (GRBO) four primary color WOLEDs possess even better color quality to their RGB counterpart.

As a result, a balanced consideration should be addressed between color quality and EL efficiency. For common phosphors and host materials, optimizing one physical parameter often frustrates other aspects, leaving a limited space for optimization. However, combining newly developed materials with common small host materials [13,14] brings a bright hope to this field.

In 2008, Cao's group utilizing poly(9-vinyl carbazole) (PVK) host coupled with blue and red phosphors fabricated a RB WOLED with a CRI of 52 and EL efficiency of 16.1 cd A^{-1}, and an RGB WOLED with a CRI of 77 and an EL efficiency of 24.3 cd A^{-1} [15]. In 2009, this group recorded an efficiency of 42.9 cd A^{-1} by doping the PVK host with a home-made yellow phosphor and the commonly used sky-blue Iridium complex, FIrpic, iridium (III) bis(2-(4,6-difluorophenyl) pyridinato-N,C^2)(picolinate). Yet the resulting moderate color quality with CRI falling in the narrow range of 52–55 is not impressive [16]. In 2012, Wang et al. reported extremely high-performance solution-processed BO WOLEDs, establishing a new world record efficiency of 70.6 cd A^{-1}, 47.6 lm W^{-1}, and 26.0% at 100 cd m^{-2} with a CRI of 62 by incorporating a new dendritic carbazole host material with a home-made orange-emitting iridium complex plus a low-conductivity PEDOT:PSS [17]. Yet the color quality still needs enhancement. In 2013, Xie's group employed a bipolar transport host of diphenylphosphoryl fluorene derivative doped with the common RGB emitters and fabricated warm WOLEDs with a power efficiency of 12.95 lm W^{-1} and a CRI of 82 at high luminance of 1594 cd m^{-2} [18].

WOLEDs possessing both good color quality and high efficiency, especially at high brightness is a demanding challenge facing the solution processing method.

Recently, Xu and his coworkers reported single-emissive-layer BO WOLEDs and obtained high efficiency of 28.3 cd A^{-1} at high luminance of 5000 cd m^{-2} by employing a newly synthesized orange phosphor [19]. In 2013, Xie and colleagues synthesized a new deep red phosphorescent dye $Ir(pmiq)_2(acac)$ and fabricated high quality WOLEDs with ultra-high CRI up to 95, power efficiency of 7.86 lm W^{-1} and high luminance of 2529 cd m^{-2} at 5 V [20]. Recently, this team synthesized a new green-yellow (GY) phosphorescent Iridium complex and fabricated three-component WOLEDs with very high CRI above 85 and improved power efficiency of 10.4 lm W^{-1} [21]. Yang's group proposed tetraarylsilane host materials and fabricated highly efficient WOLEDs with a peak power efficiency of 47.2 lm W^{-1} and a CRI of 58–60 [22]. Inverted device architecture from common emitters toward stable WOLEDs was developed by Colsmann and his coworkers with stable EL efficiency of 18 cd A^{-1} and a CRI of 75 at a large luminance regime between 1500 and 7500 cd m^{-2} [23]. This tremendous progress makes us believe that the cost-effective WOLEDs will become accessible for general applications in the near future.

In this article, we introduce a home-made host material of 4,4'-(9,9'-(1,3-phenylene)bis(9*H*-fluorene-9,9-diyl)) bis(*N*,*N*-diphenylaniline) (DTPAFB) and fabricated single-emissive-layer WOLEDs through the simple solution processing method by embracing four common molecules which are blue FIrpic, green iridium (III) bis[2-(2-pyridinyl-*N*)phenyl-C](2,4-pentanedionato-O^2,O^4) [$Ir(ppy)_2(acac)$], and orange iridium (III) bis(2-phenyl-benzothiazole-C^2,N)(acetylacetonate) [$Ir(bt)_2(acac)$], plus a home-made red phosphor of iridium (III) tris(1-(2,6-dimethylphenoxy)-4-(4-chlorophenyl)phthalazine) [$Ir(MPCPPZ)_3$]. DTAPFB host possesses more suitable energy level alignment (for details, see Fig. 1) compared to a PVK host material. The binary BO complementary color WOLEDs show stable EL emission with a high CRI over 70 and relatively high luminance efficiency of 26.3 cd A^{-1} and power efficiency of 19.6 lm W^{-1}. The optimized RGB three-component WOLEDs show improved CRI up to 81 with the maximum luminous efficiency of 25.8 cd A^{-1}, 14.4 lm W^{-1}, and external quantum efficiency (EQE) of 13.9%. Furthermore, we have studied RGBO four primary color WOLEDs and obtained good results with high EL efficiency of 28.9 cd A^{-1}, 17.4 lm W^{-1}, and EQE of 13.1% which is comparable to some thermally deposited multiply doped WOLEDs. At the illumination-relevant luminance of 1000 cd m^{-2}, the total power efficiency reaches 33.6 lm W^{-1}, and still remains 30.2 lm W^{-1} at 3000 cd m^{-2}, approaching the efficiency of state-of-the-art fluorescent-tube (40–70 lm W^{-1}), potentially suitable for environment-friendly solid-state lighting sources.

2. Experimental

2.1. Materials

TPBI and PVK were purchased from Aldrich, while OXD-7, FIrpic, $Ir(ppy)_2(acac)$, and $Ir(bt)_2(acac)$ were purchased from Luminescence Technology Corp, and the red phosphors of $Ir(MPCPPZ)_3$ and DTPAFB were synthesized by our group. Polyethylenedioxythiophene:polystyrene sulfonate (Baytron Clevios AI4083, Bayer AG), PEDOT:PSS, was commercially available and used as received.

2.2. Characteristics

UV–vis absorption and fluorescence spectra were collected with Hitachi U3010 and Hitachi F-4500 spectrophotometers,