Journal of Luminescence 142 (2013) 231–235

Contents lists available at SciVerse ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

White organic light-emitting devices based on blue fluorescent dye combined with dual sub-monolayer

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ARTICLE INFO

Article history:
Received 2 December 2012
Received in revised form
31 March 2013
Accepted 9 April 2013
Available online 19 April 2013

Keywords:
White organic light-emitting devices
Dual sub-monolayer
Luminance
Efficiency
The Commission Internationale de 1'Eclairage (CIE) coordinates
Color rendering index

ABSTRACT

White organic light-emitting devices have been realized by using highly blue fluorescent dye 4,4′-Bis (2,2-diphenyl-ethen-1-yl)-4,4′-di-(tert-butyl)phenyl(p-TDPVBi) and [2-methyl-6-[2-(2, 3,6,7-tetrahydro-1H, red fluorescent dye 5H-benzo[ij] quinolizin-9-yl) ethenyl]-4H-pyran-4-ylidene] propane-dinitrile (DCM2), together with well known green fluorescent dye quinacridone (QAD). The fabrication of multilayer WOLEDs did not involve the hard-to-control doping process. The structure of the device is ITO/m-MTDATA (45 nm)/NPB(8 nm)/p-TDPVBi(15 nm)/DCM2(x nm)/Alq_3 (5 nm)/QAD(y nm)/Alq_3(55 nm)/LiF(1 nm)/Al, where 4,4′,4″-tris{*N*,-(3-methylphenyl)-*N*-phenylamine}triphenylamine (m-MTDATA) acts as a hole injection layer, *N*,*N*′-bis-(1-naphthyl)-*N*, *N*′-diphenyl-1, 1′-biph-enyl-4, 4′-diamine (NPB) acts as a hole transport layer, p-TDPVBi acts as a blue emitting layer, DCM2 acts as a red emitting layer, QAD acts as a green emitting layer, tris-(8-hydroxyquinoline) aluminum (Alq_3) acts as an electron transport layer, and WOLEDs of devices A, B, C and D are different in layer thickness of DCM2 and QAD, respectively. To change the thickness of dual sub-monolayer DCM2 and QAD, the WOLEDs were obtained. When x, $y=0.05$, 0.1, the Commission Internationale de 1'Eclairage (CIE) coordinates of the device change from (0.4458, 0.4589) at 3 V to (0.3137, 0.3455) at 12 V that are well in the white region, and the color temperature and color rendering index were 5348 K and 85 at 8 V, respectively. Its maximum luminance was 35260 cd/m^2 at 12 V, and maximum current efficiency and maximum power efficiency were 13.54 cd/A at 12 V and 6.68 lm/W at 5 V, respectively. Moreover, the current efficiency is largely insensitive to the applied voltage. The electroluminescence intensity of white EL devices varied only little at deferent dual sub-monolayer. Device D exhibited relatively high color rendering index (CRI) in the range of 88–90, which was essentially voltage-independent.

1. Introduction

White organic light-emitting devices (WOLEDs) are attracting significant attention with respect to their potential applications in backlight, full color applications, as well as in lighting purposes [1]. Also the efficiencies and lifetimes of WOLEDs have steadily improved over the past several years [2,3]. However, lighting applications are largely dictated by the need for the WOLED color to be at a particular color point on the black body locus as well as the requirement for provision of an adequate color rendering capability for objects viewed in the white light [1]. In order to generate the desired broadband white light typically requires two or three types of emitters, which add to the complications involved in material selection and device design [4,5]. WOLEDs with various configurations have been proposed, such as using multiple emission layers in which each layer emits a different color light to generate white-light emission [6–13], using single emission layer with multiple dyes [14], using exciplex and excimer emitting WOLEDs with high color rendering index (CRI) [15–17], using microcavity structure [18] and so on. In the application of WOLED, an important issue is achieving a high CRI and maintaining a stable electroluminescent (EL) spectrum. In general, OLEDs with three types of emitters are preferred for high CRI applications [19]. Although it is possible to adjust the emission color by tuning the driving voltage [20], it is often preferable to achieve a white OLED emission independent of the driving conditions. To fulfill this requirement, one may also utilize a very thin emitting layer to ensure that their combination zone is fixed and carrier confinement is automatically achieved [21], thereby stabilizing the EL emission [22]. WOLEDs are always constructed on a multilayer device structure with two to three light-emitting components that can be fluorescent materials and/or phosphorescent materials. Among these color components of WOLEDs, many blue, most of yellow, and nearly all orange to red fluorescent dye suffer from a common problem, namely the concentration quenching of fluorescence in solid state. Consequently, one of the key developments in the advancement of organic light-emitting device (OLED) technology for flat panel display applications can be attributed to the discovery of the guest–host doped emitter system, and the dopant light-emitter of guest–host system becomes a universal method for solving the quenching problem [23]. However, in practical OLED

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0022-2313/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jlumin.2013.04.010

manufacture, the doping process is not a trivial task to handle, considering the producibility of the optimum doping level, which is normally low and less than 1–2% and only that should be carefully controlled in a narrow effective range of 0.5% for a consistent performance of the devices. In order to overcome this problem, another concept of delta-doping technique to fabrication of WOLEDs has been reported by Tsuji et al. and Xie et al. [24–26]. The simple device structures and excellent reproducibility make this well suited to low-cost lighting applications and beneficial to industrialization. In addition, from the lighting perspective, light quality refers primarily to the color and the CRI of the light. Therefore, obtaining the high CRI is another issue for WOLEDs [27]. For high-quality white-light illumination, WOLEDs with CIEx,y coordinates similar to those of the black body radiation (with a correlated color temperature (CCT) between 2500 and 6500 K), and a CRI above 80 are required [1]. Intuitively, the CRI can be enhanced by increasing the number of dopants to extend the spectral range of electroluminescence (EL). For instance, in 2002, D'Andrade et al. demonstrated that the CRI of phosphorescent WOLEDs could be greatly improved from 50 to 83 by using three (red, green and bluish-green) phosphorescent dopants instead of two complementary (red and bluish-green) dopants [8]. As another example, in 2006 the same group combined blue fluorophors and red/green phosphors to generate white EL, in which deeper blue emission was obtained from the singlet emission of the fluorophors (to relax the requirement for the more challenging true-blue phosphors with high emission efficiencies) while red/green emission was obtained from the triplet emission of the phosphors [28]. In such WOLEDs, the CRI was improved to 85. In 2007, Schwartz et al. also successfully demonstrated high CRI WOLEDs (CRI=86) using a similar hybrid fluorophor/phosphor approach [29]. However, the adoption of a larger number of emissive dopants not only complicates device fabrication, but also renders the color control in WOLEDs fraught with difficulties. In addition, most reported phosphorescent WOLEDs yielded problems remain in terms of a significant efficiency roll-off at high brightness required for practical light sources [2], or a low CRI [3], the fast drop of current efficiency results from triplet–triplet annihilation, which would limit their applications in lighting. There are a few reports on the CRI of the sub-monolayer fluorescent WOLEDs [30,31], since white light is made up of nearly equal intensities of light from all regions of the visible spectrum. Combining three different wavelengths of light can produce white light, provided they are widespread in the visible region. Any such sets of the three different wavelengths that on additive mixing can produce white light are called 'primary colors'. The most common set of primary color is red, green and blue. Another approach is 'complementary colors' where the combination of only two colors can produce white light. The CRI of the earlier reported sub-monolayer fluorescent WOLEDs [25–27] should be low because it utilizes two complementary colors to obtain white emission. From the point of view of color rendering, all the three primary colors (red, green, and blue) have to be produced simultaneously. A high-CRI non-doped-type white organic light-emitting device with a RGB-stacked multilayer structure has been reported. The color temperature and color rendering index were 6080 K and 97, respectively. But its maximum power efficiency was only 1.31 lm W^{-1} [30]. In this work, four devices A, B, C and D were fabricated to test the idea of bright and efficient all non-doped WOLEDs using p-TDPVBi, DCM2, and QAD for blue, red, and green emitters, respectively. We adjust the thickness of DCM2 and QAD to match the control on exciton recombination zone. Its maximum current efficiency and maximum power efficiency were 13.54 cd/A at 12 V and 6.68 lm/W at 5 V, respectively. Moreover, the current efficiency is largely insensitive to the applied voltage. The electroluminescence intensity of white EL of devices varied only little at deferent dual sub-monolayer. Device D exhibited relatively high color rendering index (CRI) in the range of 88–90, which was essentially voltage-independent.

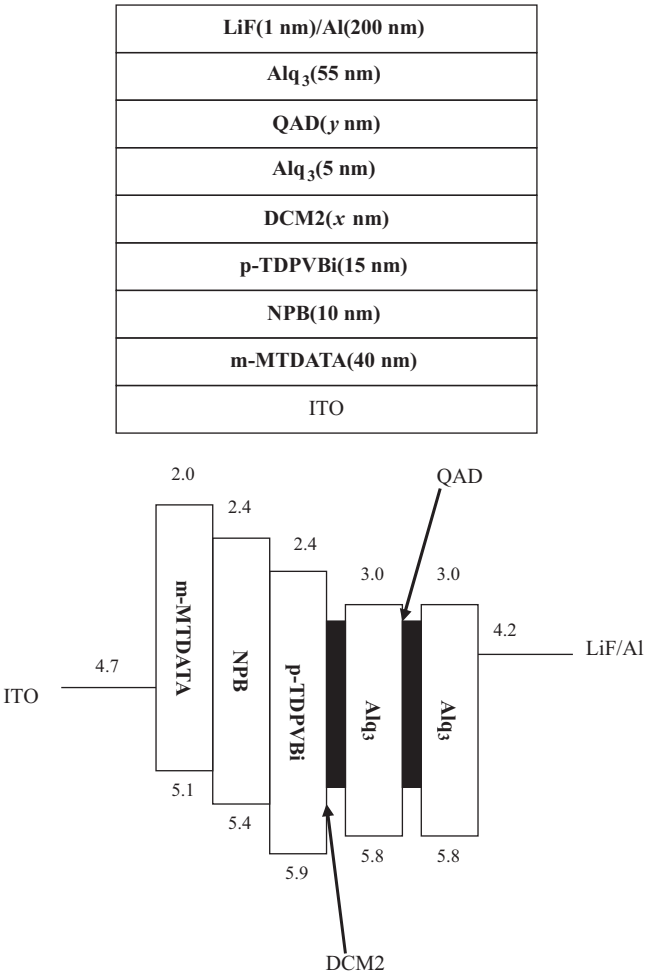

Fig. 1. The device structure of the OLEDs and schematic energy band diagram of devices.

2. Experimental details

Fig. 1 shows the structure of the device and the energy level diagram of the device. The structure of the device is ITO/m-MTDATA (45 nm)/NPB (8 nm)/p-TDPVBi(15 nm)/DCM2(x nm)/Alq_3 (5nm)/QAD (y nm)/Alq_3 (55 nm)/LiF (1 nm)/Al, where x, y=0.05, 0.05; 0.05, 0.1; 0.1, 0.05; 0.1, 0.1 (The thickness of the ultrathin layer in our device is an average value. The ultrathin layer is not a continuous layer but has some dispersive islands and an average thickness of 0.05 or 0.1 nm.) and the corresponding devices are named A, B, C and D, respectively.

Organic layers were deposited by high-vacuum (10^{-6} Torr) thermal evaporation onto a cleaned indium tin oxide (ITO) coated glass substrate. The layer thickness of the deposited material was monitored using an oscillating quartz thickness monitor. EL spectra and CIE coordination of the devices were measured by PR655 spectra scan spectrometer and the current–voltage–brightness characteristics were simultaneously measured by a Keithley 2400 programmable voltage–current source. All measurements were carried out at room temperature under ambient conditions.

3. Results and discussion

In general, the EL spectra and the CIE coordinates of the white light-emitting device are strongly influenced by the thickness of each emissive layer and applied voltage. Fig. 2(a) shows the EL