Thin Solid Films 548 (2013) 389–397

Contents lists available at ScienceDirect

Thin Solid Films

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

Aligned energy-level design for decreasing operation voltage of tandem white organic light-emitting diodes

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ARTICLE INFO

Article history:
Received 9 March 2013
Received in revised form 22 August 2013
Accepted 23 August 2013
Available online 31 August 2013

Keywords:
Organic light-emitting devices
Tandem
Operation voltage
Bipolar host materials
Energy level alignment

ABSTRACT

In general, organic light-emitting devices (OLEDs) need to operate at higher current density levels to ensure an ample light flux. However, stressed operation will result in poor performance and limited device lifetime. Recently, a tandem structure has been proposed as a pivotal technique to meet the stringent lighting requirements for OLED commercialization, with a research focus on decreasing the concomitant higher operation voltage. Driving two connected emission units (EMUs) in a tandem structure often requires more than twice the driving voltage for a single EMU. This study investigates bipolar host materials and their effective employment in fabricating tandem white phosphorescent OLEDs (PhOLEDs). In addition, the design of a mechanism to align the energy level between the hole transport layer/emitting layer is shown to effectively mitigate operational voltages. In sharp contrast to devices using a unipolar host material, we demonstrate that the turn-on voltage of blue PhOLEDs could be decreased from 3.8 V to 2.7 V through utilizing a bipolar host. Furthermore, applying the proposed techniques to tandem white PhOLEDs produces a luminance of 10^3 cd/m^2 by a 10.1 V driving voltage.

1. Introduction

Organic light-emitting devices (OLEDs) were developed in 1987 [1], using a double-layer structure with hole-transport and electron-transport materials to reduce the operation voltage. Since then researchers have focused their efforts on identifying the distinct functionalities of various materials to improve OLED performance. Recently, phosphorescent materials have been the subject of extensive investigation because their internal quantum efficiencies can reach the ideal level of 100% [2], which is far superior to the 25% upper limit imposed by the formation of singlet excitons in fluorescent materials. The electrophosphorescence can efficiently utilize both the singlet and triplet excited states, thus OLEDs incorporating phosphorescent emitters can potentially meet the requirements for practical display and lighting applications. Therefore, it is reasonable to envision that phosphorescent OLEDs will replace fluorescent OLEDs in the near future.

A tandem structure is an indispensible method to obtain a qualified lighting source with an ample flux, employing charge generation layers (CGL) to connect the individual emission units (EMUs). The efficiency, luminance and operation voltage are multiplied by the number of connected units. However, the operation voltages of tandem OLEDs connecting two units are usually much greater than twice the voltage used in a single unit [3,4]. Hence, the operation voltage mitigation provided by connecting multiple EMUs has become a compelling area of research interest. Several design rules should be considered in the initial developing status of a single EMU. A host with wider triplet energy gaps should be used in phosphorescent OLEDs to ensure the effective confinement of the triplet exciton, particularly in blue and white OLEDs [5]. Once the host and emissive dopant are determined, the appropriate carrier-transport materials with their corresponding triplet energy gaps are required to prevent exciton migration and diffusion. In addition, a step-wise injection is frequently used to smooth the carrier injection, thus reducing the likelihood of the formation of a localized electrical field. In fact, the ideal method is to use carrier transport and emitting host materials with identical energy levels.

Currently, the most advanced architectures include the transparent anode, hole-injection layer, hole-transport layer (HTL), emitting layer (EML), electron-transport layer (ETL), electron-injection layer, and reflective cathode. Undoubtedly, the host materials play a key role since they determine the effective exciton formation and confinement. The carrier-transport characteristics of the host materials can be roughly categorized as hole-transport, electron-transport and bipolar. Herein, three wide triplet-energy-gap host materials, 9-(4-*tert*-butylphenyl)-3,6-bis(triphenylsilyl)-9*H*-carbazole (CzSi) [6], 3-bis(9-carbazolyl) benzene (mCP) [7] and 3,6-di(9-carbazolyl)-9-(2-ethylhexyl) carbazole (TCz1) [8], were selected as the test host materials because of their useful application in blue phosphorescent OLEDs. The triplet energy gaps (E_T) of CzSi, mCP, and TCz1 are 3.02 eV, 2.9 eV and 2.92 eV, respectively. Each

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0040-6090/$ – see front matter
http://dx.doi.org/10.1016/j.tsf.2013.08.084

has a diverse ionization potential and carrier transport capability, providing an opportunity to compare their corresponding operation voltages. In sharp contrast to the operation voltage of 6.9 V at 10^3 cd/m^2 in CzSi-based blue phosphorescent OLEDs (PhOLEDs), the operation voltage of TCz1-based PhOLEDs was as low as 4.2 V, demonstrating the significance of bipolar transport and energy-gap alignment. In addition, the location of the exciton formation zone provides a glimpse into the carrier transport. Moreover, the TCz1 host was employed in tandem white OLEDs, with maximum efficiencies of up to 20.1%, 25.2 cd/A and 14.2 lm/W.

2. Experimental details

2.1. OLED fabrication and characterizations

An indium–tin–oxide (ITO)-coated substrate (0.7 mm) with a resistance of ≤15 Ω was purchased from AimCore. The commercial materials used were purchased from Nichem and purified by temperature-gradient sublimation under high vacuum before use. Substrates were cleaned by 5 min exposure to an UV-ozone atmosphere. OLED devices were fabricated using ITO substrates with multiple organic layers, sandwiched between the transparent bottom ITO anode and the top metal cathode. The organic and metal layers were deposited by thermal evaporation in a vacuum chamber with a base pressure of $<10^{-4}$ Pa without breaking the vacuum. The deposition rate of the organic layers was kept at around 0.1 nm/s. The active area of the device was 2×2 mm^2, as defined by the shadow mask used for cathode deposition. Current–voltage–luminance (J–V–L) characterization was performed using an Agilent4156C semiconductor parameter analyzer equipped with a calibrated Si-photodiode. Electroluminescence spectra of the devices were recorded by using an Ocean Optics spectrometer.

3. Results and discussions

3.1. The designs of energy level alignment

The sky-blue phosphorescent emitter, iridium(III) bis[(4,6-difluorophenyl)-pyridinato-N,$C^{2'}$] picolinate (FIrpic), has been shown to possess a photoluminescence quantum yield that could reach nearly 100% in wide-gap hosts. Consequently, the FIrpic was used to examine the role of the hosts in OLEDs. The di-[4-(N,N-ditolylamino)-phenyl] cyclohexane (TAPC) [9] and 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) [10] were respectively selected as the hole-transport layer and electron-transport layer due to their excellent carrier transport capabilities. The hole and electron mobilities of TAPC and TmPyPB are about 10^{-2} and 10^{-3} cm^2/Vs, respectively [7,8]. Furthermore, both possess wide triplet gaps (E_T) of about 2.87 and 2.78 eV, respectively, facilitating high energy exciton confinement [9,10].

The complexity of white PhOLEDs with tandem structures requires the development of a simplified architecture for the intrinsic OLEDs without compromising performance [11]. Based on our previous experience, we constructed the device using an ITO (110 nm)/TAPC (40 nm)/host doped with 8.0 wt.% FIrpic (21 nm)/TmPyPB (50 nm)/LiF (0.8 nm)/Al (150 nm), with aluminum used as the cathode. The devices using CzSi, mCP and TCz1 as emitting host materials were named Devices A1, B1 and C1, respectively. A structural drawing of the materials is shown in Fig. 1, while the schematic structures of the tested OLEDs are shown in Fig. 2.

The EL characteristics of Devices A1, B1 and C1 are shown in Fig. 3 (Table 1). The pure FIrpic emission shown in the EL spectra indicates that the host materials provide an effective carrier transport as well as sufficient exciton formation in the emitting layer. In addition, the carrier

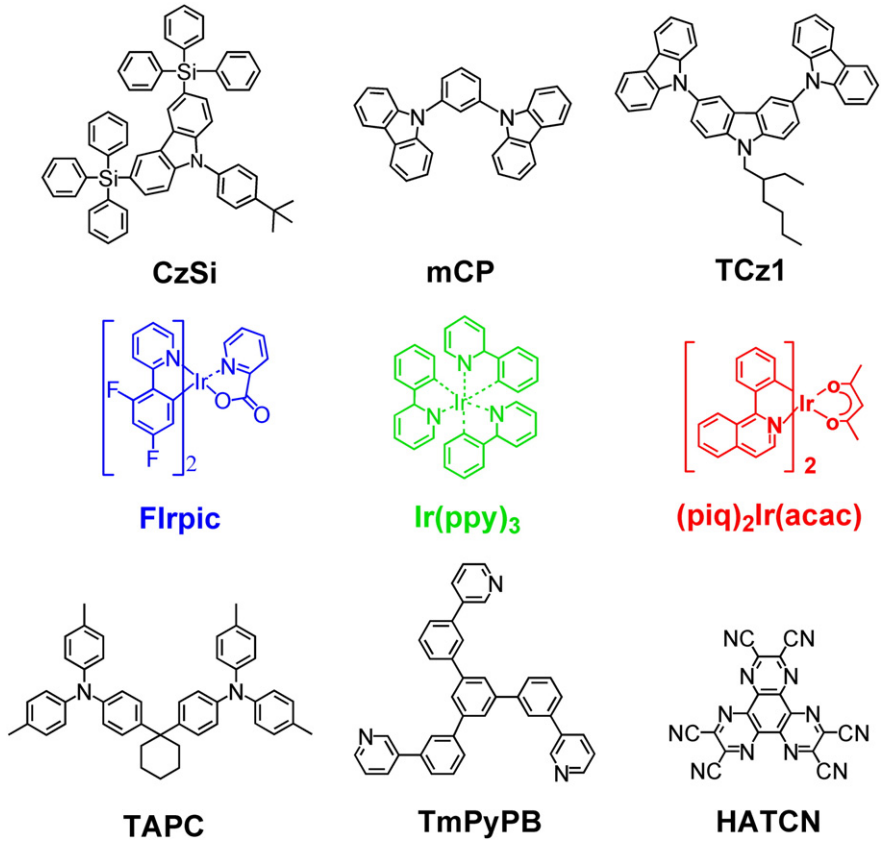

Fig. 1. Structural drawing of employed materials.