



Efficient blue organic light-emitting diodes based on triphenylimidazole substituted anthracene derivatives



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ABSTRACT

A series of new blue emissive materials based on the conjugates of highly fluorescent diaryl anthracene and electron-transporting triphenylimidazole moieties: 2-(4-(anthracen-9-yl)phenyl)-1,4,5-triphenyl-1H-imidazole (**ACBI**), 2-(4-(10-(naphthalen-1-yl)anthracen-9-yl)phenyl)-1,4,5-triphenyl-1H-imidazole (**1-NaCBI**), 2-(4-(10-(naphthalen-2-yl)anthracen-9-yl)phenyl)-1,4,5-triphenyl-1H-imidazole (**2-NaCBI**) were designed and synthesized successfully. These materials exhibit good film-forming properties and excellent thermal stabilities. Meanwhile, the decreased π -conjugation in these compounds compared with phenanthroimidazole derivatives leads to obvious hypsochromic shift. To explore the electroluminescence properties of these materials, typical three-layer organic light-emitting devices were fabricated. With respect to the three layer device **2** using **1-NaCBI** as the emitting layer, its maximum current efficiency reaches 3.06 cd A^{-1} with Commission Internationale de l'Eclairage (CIE) coordinates of (0.149, 0.092). More interestingly, sky blue doped device **5** based on **1-NaCBI** achieved a maximum current efficiency of 15.53 cd A^{-1} and a maximum external quantum efficiency of 8.15%, high EQE has been proved to be induced by the up-conversion of a triplet excited state.

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1. Introduction

In the past decade, organic light-emitting diodes (OLEDs) have attracted much scientific and commercial interest because of their potential application in full-color displays and large-area, flexible, light-weight light sources [1]. Full-color displays require red, green, and blue emissions of relatively equal stability, efficiency, and color purity. Specifically, highly efficient blue electroluminescent devices with Commission Internationale de l'Eclairage (CIE) y-coordinate value <0.10 , can not only effectively reduce the power consumption of a full-color OLED but

be utilized to generate light of other colors by energy cascade to lower energy fluorescent to realize white light. Red and green phosphorescent electroluminescent devices with high efficiencies, long lifetimes, and proper CIE coordinates have been well developed. However, blue phosphorescent devices are still the bottleneck for the high CIE coordinates (y-coordinate value >0.30), high cost and short device lifetime [2]. To address the problem, a continuous effort has been contributed to develop highly efficient blue-light emitters with good color purity and high efficiency [3].

Anthracene derivatives have been studied extensively as blue-light-emitting materials in OLEDs because of their excellent photoluminescence (PL) and electroluminescence (EL) properties [4]. For example, Li' group [5] have synthesized some fluorene derivatives with naphthylanthracene

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endcaps and achieved device efficiency of 2.10 lm W^{-1} , color coordinates (0.16, 0.09). Shu's group [6] have developed 2-tert-butyl-9,10-bis[4'-(diphenylphosphoryl)phenyl]anthracene (POAn) with a maximum luminance efficiency of 2.9 cd A^{-1} , CIE coordinates of (0.15, 0.07). On the other hand, phenanthroimidazole derivatives have attracted great attention as electroluminescent materials because of their high thermal stability and efficient electron transporting ability. Huang's group [7] exploited bis(phenanthroimidazolyl)biphenyl derivatives as excellent non-doped blue emitting materials with optimized device efficiency of 7.3 lm W^{-1} , CIE coordinates of (0.15, 0.14). Using phenanthroimidazole as a building block for luminescent materials, Ma's group [8] fabricated pure blue devices with luminance efficiency of 6.87 cd A^{-1} , CIE coordinates of (0.15, 0.21) and a turn on voltage of 2.8 V. The impressive device performances of 2.63 cd A^{-1} CIE (0.15, 0.09) and 5.66 cd A^{-1} , (0.15, 0.11) based on donor-linker-acceptor structural deep-blue emitting phenanthroimidazole derivatives have been reported by Tong and Ma's group, respectively [9]. Very recently, an promising device performance with EQE of 7.8% and current efficiency of 10.4 cd A^{-1} has been reported based on an excellent blue dopant PPIE containing n-type imidazole moiety [10]. In the subsequent research, the high EQE is proved to be induced by the triplet-triplet annihilation (TTA). In spite of their successful performance in a given aspect, satisfactory blue luminescent materials are still rare.

In our previous work, some hybrid bipolar phosphorescent hosts for green and orange OLEDs have been synthesized by conjugating carbazole moiety to the rigid skeleton of 1,2-diphenyl-1H-phenanthro[9,10-d]imidazole [11]. We also developed some asymmetric phenanthroimidazole C2-phenyl or N1-phenyl position substituted anthracene derivatives for efficient blue OLEDs [12]. The introduction of anthracene moieties can effectively increase the electron injection and transport ability and also finely adjust the ionization potentials (Ip) of the compounds, resulting in reduced hole injection barrier and balanced recombination ability. In this paper, we introduced the bulky non-planar triphenylimidazole instead of phenanthroimidazole moiety to obtain deeper blue fluorescent materials. As expected, these new materials show excellent thermal stabilities, proper HOMO levels and excellent EL performances with low onset voltages. Three layer device **2** using **1-NaCBI** as the emitting layer, achieved a maximum current efficiency of 3.06 cd A^{-1} with Commission Internationale de l'Eclairage (CIE) coordinates of (0.149, 0.092). In addition, device **5** based on doping BUBD-1 in **1-NaCBI**, the maximum current efficiency reaches 15.53 cd A^{-1} with maximum external quantum efficiency of 8.3%. The high EQE has been proved to be induced by the up-conversion of a triplet excited state.

2. Experimental section

2.1. Material and methods

All reagents and solvents were used as purchased from Aldrich without further purification. Most of experiment methods were according to our published results [11,12].

^1H NMR spectra were recorded on a Bruker-AF301 at 400 MHz. Mass spectra were carried out on an Agilent MALDI-TOF. Elemental analyzes of carbon, hydrogen, and nitrogen were performed on an Elementar (Vario Micro cube) analyzer. Fluorescence spectra and transient fluorescence decay lifetimes were obtained on Edinburgh instruments (FLSP920 spectrometers) and UV-Vis absorption spectra were measured using a Shimadzu UV-Vis-NIR Spectrophotometer (UV-3600). The differential scanning calorimetry (DSC) analyzes were performed under a nitrogen atmosphere at a heating rate of 10°C/min using a PE Instruments DSC 2920. Thermogravimetric analyzes (TGA) were undertaken using a PerkinElmer Instruments (Pyris1 TGA) under nitrogen atmosphere at a heating rate of 10°C/min . Atomic force microscopies (AFMs) were measured using Shimadzu (SPM9700). To measure the photoluminescence (PL) quantum yields (Φ_F), degassed solutions of the compounds in CH_2Cl_2 were prepared. The concentration was adjusted so that the absorbance of the solution would be between 0.05 and 0.1. The excitation was performed at 330 nm and 9,10-diphenylanthracene (DPA) in cyclohexane ($\Phi = 0.9$ in cyclohexane) was used as a standard [13]. Cyclic voltammetry (CV) measurements were carried out in a conventional three electrode cell using a Pt button working electrode of 2 mm in diameter, a platinum wire counter electrode, and a Ag/AgNO_3 (0.1 M) reference electrode on a computer-controlled EG&G Potentiostat/Galvanostat model 283 at room temperature. Oxidations of all compounds were performed in dichloromethane containing 0.1 M tetrabutylammonium hexafluorophosphate (Bu_4NPF_6) as supporting electrolyte. The onset potential was determined from the intersection of two tangents drawn at the rising and background current of the cyclic voltammogram and calibrated to the ferrocene/ferrocenium (Fc/Fc^+) redox couple. DFT calculations were performed to characterize the 3D geometries and the frontier molecular orbital energy levels of **ACBI**, **1-NaCBI** and **2-NaCBI** at the B3LYP/6-31G* level by using the ADF2009.01 program.

2.2. Device fabrication and measurement

The EL devices were fabricated by vacuum thermal evaporation technology according to the methods modified from our previous approach [11,12]. Before the deposition of an organic layer, the clear ITO substrates were treated with oxygen plasma for 5 min. The deposition rate of organic compounds was $0.9\text{--}1.1 \text{ \AA s}^{-1}$. Finally, a cathode composed of cesium pivalate (2 nm) and aluminum (100 nm) was sequentially deposited onto the substrate in the vacuum of 10^{-5} Torr. The $L\text{--}V\text{--}J$ curves of the devices were measured with a Keithley 2400 Source meter and PR655. All measurements were carried out at room temperature under ambient conditions.

3. Results and discussion

3.1. Synthesis

The structures and synthetic routes of the three well-defined compounds are shown in Scheme 1. Anthracen-9-yl-

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