Displays 34 (2013) 320–325

Contents lists available at SciVerse ScienceDirect

Displays

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

Small molecules with pyridine backbone modified with carbazole, fluorine and bromine for white light-emitting diode applications

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ARTICLE INFO

Article history:
Available online 25 June 2013

Keywords:
Pyridine
Carbazole
Bromo
Excimer
Orange
White

ABSTRACT

Seven compounds with pyridine as the backbone modified by carbazole moiety, bromine atom and fluorine atom were synthesized. Compounds **1, 2, 3** with bromo substitution at the 2-position and carbazole modification at the 5-position of pyridine emit not only a sharp blue singlet fluorescence but also a wide banded excimer-based orange emission. The two colors coming from a single molecule can be used to fabricate a simplified white light emitting device. The electroluminescence based on **1** and **2** exhibits white-light emission with CIE coordinates of $x = 0.25$ and $y = 0.30$ for **1** and $x = 0.33$ and $y = 0.37$ for **2** at high current densities, very close to pure white emission. In addition, the role of bromo-substitution at pyridine is concluded to be essential to generate molecular interaction thus an excimer emission.

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1. Introduction

Organic light-emitting diodes (OLEDs) have received significant attention over the past two decades, demonstrating their great potential in full-color flat-panel displays and solid-state white-lighting [1]. Considerable achievements in efficiency and life-time as monochromatic emission have led to the development of devices with broad band white emission [2]. One common approach to obtain white emission is to fabricate multi-emission-layer devices, in which each layer emits a primary color [3]. Another strategy is via a single layer structure in which dyes emitting different colors are doped in a host [4] and mixing molecular emission (usually blue) or exciplex emission (usually orange) are obtained [5]. White organic light-emitting diodes (WOLEDs) based on a single emitter have advantages, such as improved color stability and device stability, better reproducibility, and simpler fabrication [6].

For real-world application in optoelectronic devices, light emitters usually have to be fabricated into thin solid films. However, the concentration quenching effect caused by the strong intermolecular π–π interaction or intermolecular hydrogen bonding between neighboring fluorophores in the solid state generally debases or quenches the emission [7]. Nevertheless, aggregation-induced enhanced emission (AIEE) [8] or excimer emission [9] is welcome. The excimer exhibits characteristic featureless, red-shifted emission spectra as compared to their monomer counterparts. The excimer emission plus the monomer emission usually make up a white light emission and becomes useful in white lighting. Because few materials are known to emit "pure" white light as a single-emitting component [10], the search for new organic light-emitting materials based on excimers for use in single-emitting-component WOLEDs is of obvious interest and importance.

In this paper, we report on the synthesis, photoluminescence and electroluminescence properties of several molecular compounds modified on the pyridine backbone. Of the seven materials synthesized, **1, 2, 3** emit sharp blue singlet fluorescence and wide band excimer-based orange emission and can act as white light emitting materials. The device based on **1** and **2** exhibits white-light emission very close to the international standard.

2. Experimental

2.1. Materials

1,4-Dibromobenzene, 1-bromo-4-fluorobenzene, iodobenzene, CuI, carbazole, *n*-BuLi, $B(OCH_3)_3$, 1-bromo-4-methylbenzene, 4-bromo-1,1′-biphenyl, 1-bromo-4-fluorobenzene, 2,5-dibromopyridine, 1,10-Phenanthroline, $Pd(PPh_3)_4$ and NBS were bought from Alfa Aesar. 9-phenyl-9H-carbazole (**a**″), 9-(*p*-fluorophenyl)-9H-carbazole (**b**″) and 9-(*p*-bromophenyl)-9H-carbazole (**c**′) were synthesized by Ullmann reaction, a procedure that can be commonly found in the literature [11]. A series of different substrate pydrine/carbazole-based bipolar compounds **1**–**7** were synthesized by suzuki reaction (Scheme 1) [12]. All solvents used in the reaction were purified by routine procedures. Other reagents were used as received from commercial sources, unless otherwise stated.

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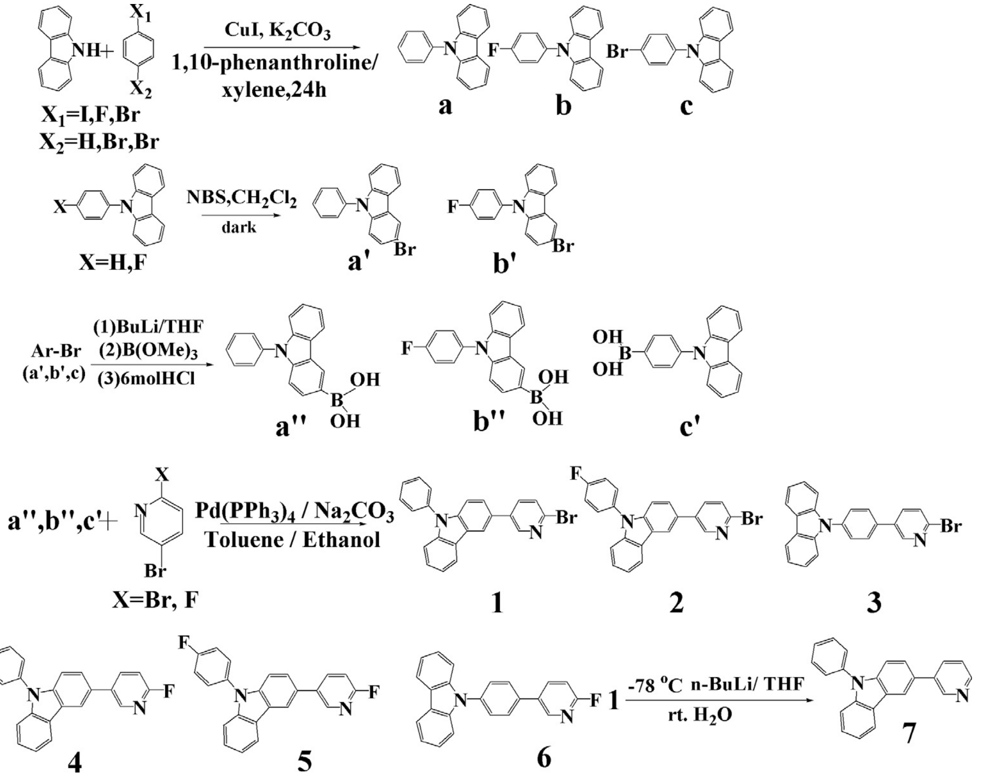
http://dx.doi.org/10.1016/j.displa.2013.06.002



Scheme 1. Synthetic route and the molecular structure of **1–7**.

2.2. Measurements and OLED fabrication

^{1}H NMR was collected by a Bruker 400 MHz spectrometer. Elemental analysis was carried out on a Vario III elemental analyzer. UV–vis absorption spectra were recorded on a Perkin–Elmer Lambda-35 spectrometer. Photoluminescence (PL) spectra were obtained using a Hitachi F-4500 florescence spectrophotometer.

Fabrication of light-emitting devices: The OLEDs were prepared by vacuum deposition system. Organic functional layers were fabricated on a clean glass substrate pre-coated with a 150 nm thick, ~15 Ω/sq indium tin oxide layer (emitting area was 2 × 2 mm^2). Prior to usage, the substrate was degreased in an ultrasonic bath by the following sequence: in detergent, de-ionized water, acetone, and isopropanol, and then cleaned in a UV–ozone chamber for 10 min. Hole injection layer (HIL), emitting layer (EML) and electron transport layer (ETL) were subsequently evaporated in high vacuum (~10^{-4} mbar). The devices were measured with a current voltage source/measure unit (Keithley 2400) and a PR-650 luminance color meter. The color meter was placed on the bottom of the test device to measure the front luminance.

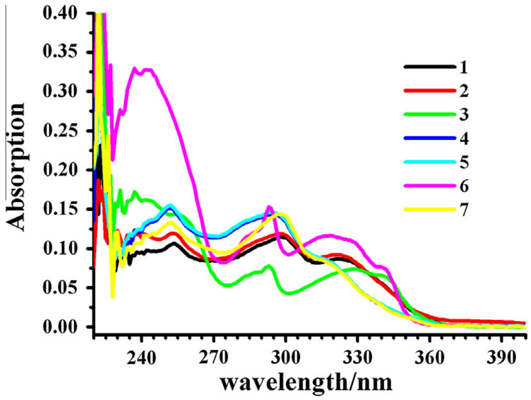


Fig. 1. Absorption spectra **1–7** in CH$_2$Cl$_2$.

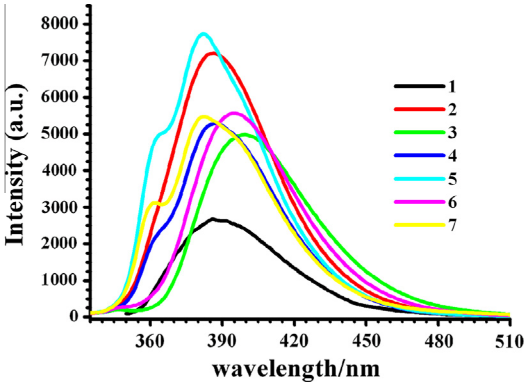


Fig. 2. Emission spectra of **1–7** in CH$_2$Cl$_2$, excited by 313 nm.

3. Results and discussion

The chemical structure and the synthetic procedure for the target compounds**1**–**7**are illustrated inScheme 1.Detailed synthetic procedure and characterization can be found in the supplementary material.

Fig. 1 shows the absorption spectra of **1**–**7** in CH$_2$Cl$_2$. The strong absorption peaks around the high energy 270–310 nm region correspond to the spin-allowed, π–π^* absorption of the conjugated phenyl rings. The lower energy, weaker bands around 310–360 nm are attributed to the forbidden n–π^* absorption localized at the N atoms.

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