



Non-doped red-green-blue electroluminescence for fumaronitrile and fluorene bridge with pyrenyl or phenanthrylamino group



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ABSTRACT

A red-emitting bis(4-(*N*-(1-pyrenyl)-3,5-dimethylphenylamino)phenyl)fumaronitrile (**3**) and a green-emitting 2,7-bis(*N*-1-pyrenyl-3,5-dimethylphenylamino)-9,9-diethylfluorene (**4**) were synthesized, while a red emissive bis(4-(*N*-(9-phenanthryl)-3,5-dimethylphenylamino)phenyl)fumaronitrile (**5**) and a blue 2,7-bis(*N*-9-phenanthryl-3,5-dimethylphenylamino)-9,9-diethylfluorene (**6**) were reported previously. The bulky and non-planar phenanthryl or pyrenyl amino substituent created steric torsional hindrance to restrict intermolecular aggregation and to promote high carrier mobility, thermal stability and luminous efficiency. The red non-doped devices for donor– π –acceptor **3** and **5** showed electroluminescent (EL) peaks at 668 and 652 nm with luminance (external quantum) efficiencies of 3.45 cd/A (6.89%), and 2.52 cd/A (3.74%), respectively. **4** exhibited EL peak at 500 nm with efficiency of 2.18 cd/A (0.81%). The device based on **6** was centered at 460 nm with efficiency of 1.86 cd/A (1.16%). The large planar pyrenyl group had stronger electron-donating ability than phenanthryl group, so the EL emissions from **3** and **4** redshifted compared with those from the homologous **5** and **6**. EL efficiency was improved more effectively by pyrenyl unit than by phenanthryl unit. The prepared compounds were multifunctional materials with emitting and hole-transporting properties.

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1. Introduction

Non-doped red fluorescent pigments have attracted increasing attention in terms of simplifying and improving the efficiency of electroluminescent (EL) devices [1]. Donor–acceptor-substituted red pigment is normally highly polar and vulnerable to aggregation in solid state. A pair of antiparallel dipoles [2–4] has an electron acceptor and arylamino donor [5] to suppress fluorescence quenching in solid state and to balance the mobility of hole and electron carriers within the device. In particular, arylamino fumaronitrile derivatives, such as 1-naphthyl, 2-naphthyl, 9,9-diethyl-2-fluorenyl amino fumaronitrile [5–7], and diphenylamino spirobifluorenylfumaronitrile derivatives [8], possess a non-planar arylamino group and fumaronitrile core to form donor– π –acceptor structure and exhibit high EL efficiency. In addition, these derivatives exhibit aggregation-induced emission [9]. Photoluminescence (PL) of dicyano-bis(4-(carbazolyl)phenyl)ethylene in film or fluorescent organic nanoparticle is much brighter than that in solution [10].

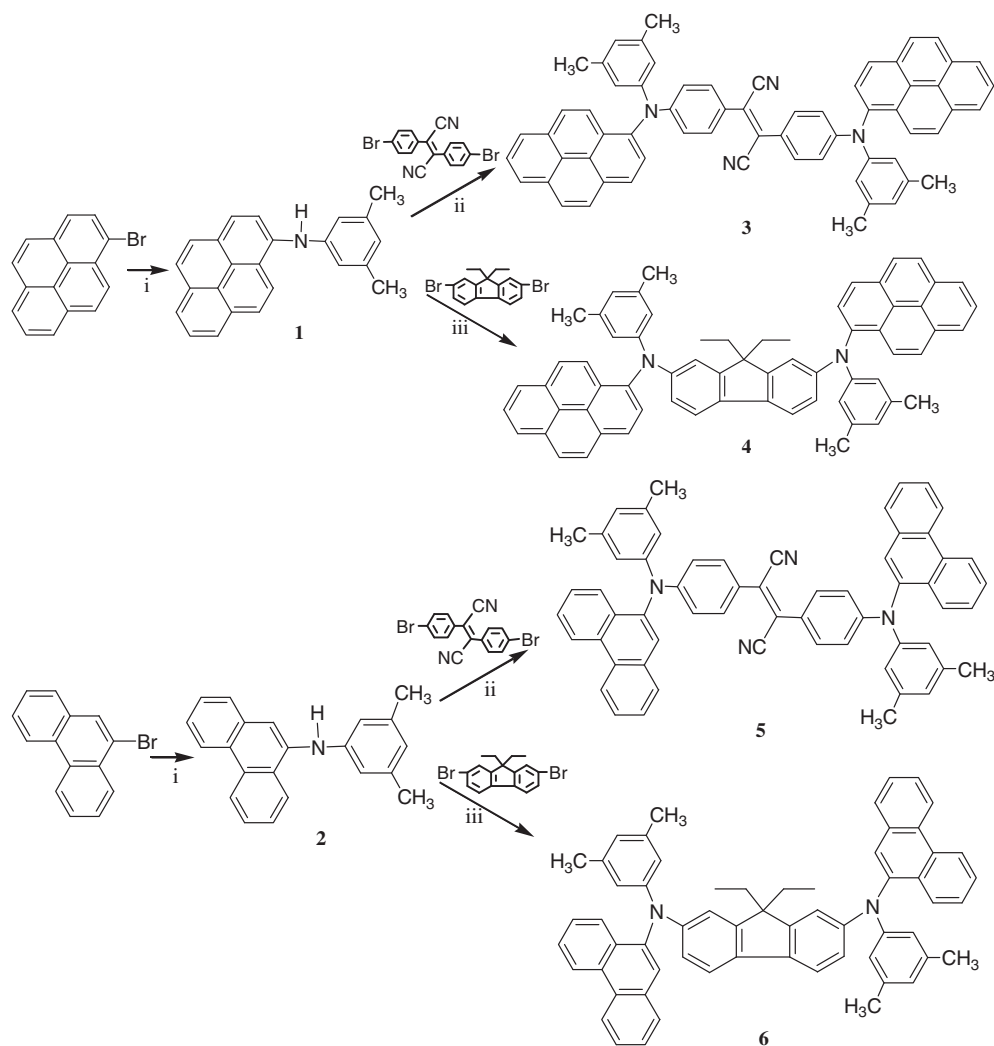
Typical organic triarylaminos possess excellent hole-transporting property [11]; however, their low morphological stability easily degrades the device [12]. Polycyclic aromatic hydrocarbons, such as naphthalene, fluorene and phenanthrene, especially the strong π electron

delocalization energy of large planar pyrene [13,14], greatly promote thermal stability, high carrier mobility, hole injection, and fluorescence quantum yield [15–19]. Fluorene-bridged triarylamine is reported mainly as a hole-transporting or hole-injecting material but only seldom as an emitter [20–22].

In our previous work, a red-emitting bis(4-(*N*-(9-phenanthryl)-3,5-dimethyl-phenylamino)phenyl)fumaronitrile (PhMPAFN, **5**) and a blue-emitting 2,7-bis(*N*-9-phenanthryl-3,5-dimethylphenylamino)-9,9-diethylfluorene (PhMPAEF, **6**) were reported [23]. In addition, devices ITO/NPB/**5**/BCP/AlQ₃/LiF/Al, ITO/NPB/**6**/AlQ₃/LiF/Al and ITO/**6**/BCP/AlQ₃/LiF/Al were characterized. Here, ITO stands for indium tin oxide glass substrate, NPB for 4,4'-bis(*N*-(1-naphthyl-*N*-phenylamino)biphenyl, BCP for 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline and AlQ₃ for tris(8-hydroxyquino)aluminum. In this study, the bulky and large planar conjugated pyrenyl group was incorporated to nitrogen atom in fumaronitrile and fluorene bridge, the non-planar pyrenylamino substituent created steric torsional hindrance to restrict intermolecular aggregation and to promote high carrier mobility, thermal stability and luminous efficiency. A red-emitting bis(4-(*N*-(1-pyrenyl)-3,5-dimethylphenylamino)phenyl)fumaronitrile (PyMPAFN, **3**) and a green-emitting 2,7-bis(*N*-1-pyrenyl-3,5-dimethylphenylamino)-9,9-diethylfluorene (PyMPAEF, **4**) were prepared. Red non-doped devices based on **3** and **5**, a green-emitting one based on **4**, and a blue-emitting one based on **6** were fabricated and characterized. Furthermore, the

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Scheme 1. The schematic diagram for synthetic routes of **3**, **4**, **5** and **6**. Reagents and conditions are: (i) 3,5-dimethylphenylamine, Pd(OAc)₂, bis(2-(diphenylphosphino)phenyl)ether, toluene, sodium *tert*-butoxide, purged with nitrogen, 85 °C, 24 h; (ii) bis(4-bromophenyl)fumaronitrile, Pd(OAc)₂/P(*t*-Bu)₃, toluene, Cs₂CO₃, 110 °C, 18 h; (iii) 2,7-dibromo-9,9-diethylfluorene, Pd(dba)₂/P(*t*-Bu)₃, *o*-xylene, sodium *tert*-butoxide, 110 °C.

effects of polycyclic aromatic phenanthryl and pyrenyl units on the EL emissions from fluorene bridge and fumaronitrile core were investigated.

2. Experimental details

2.1. Materials and instruments

Fluorene, pyrene, 3,5-dimethylphenylamine, Cs₂CO₃, sodium *tert*-butoxide, palladium acetate (Pd(OAc)₂), bis(dibenzylideneacetone)palladium (Pd(dba)₂), and tris(*tert*-butyl)phosphine (P(*t*-Bu)₃) were purchased from Acros Organics. *o*-Xylene, toluene, cyclohexane and tetrahydrofuran (THF) were refluxed with sodium and benzophenone followed by distillation. Bis(4-bromophenyl)fumaronitrile [7,24], 2,7-dibromo-9,9-diethylfluorene [20,25], *N*-3,5-dimethylphenyl-9-phenanthrylamine (**2**), **5** and **6** were synthesized according to the reference [23].

Synthetic compounds were characterized through elemental analysis (Carlo Erba 1106) and proton nuclear magnetic resonance (HNMR) spectroscopy (Bruker DMX-300). Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed by Netzsch TG-209 and Netzsch DSC-200PC. Cyclic voltammetry curves were recorded using Zahner Zennium electrochemical workstation. Tetra-*n*-butylammonium hexafluorophosphate in anhydrous dichloromethane

formed a supporting electrolyte (0.1 M), whereas ferrocene (Fc) served as a reference. A platinum disk electrode was composed of a platinum disk (a diameter of 3 mm) within a polytetrafluoroethylene coat (an outer diameter of 6 mm and a length of 60 mm) as the working electrode, the total length of an electrode was 80 mm. A platinum wire electrode consisted of a polytetrafluoroethylene coat (an outer diameter of 6 mm and a length of 25 mm) connecting a platinum wire (a diameter of 0.5 mm and a length of 37 mm) as the counter electrode, the total length of an electrode was 80 mm. Connection scheme was set as 2 electrodes. An Hg/Hg₂Cl₂ electrode (a saturated calomel electrode, SCE, the potential value was 0.241 V) acted as a reference electrode. The surfaces of an Hg/Hg₂Cl₂ electrode and a platinum wire electrode were cleaned with an alcohol cotton ball. The surface of a platinum disk electrode was polished on Al₂O₃ powder (grain size is 0.05 μm), rinsed with distilled water and dried. The concentration of the test sample solution with the saturated argon ranged from around 10⁻³ to 10⁻⁴ M. The lower potential was set at 0, or +0.2 V, respectively, the upper potential was at +1.5 V. The scanning rate was 50 mV/s, and the scanning cycles were 3. PL spectrum was recorded using the Perkin Elmer LS-55 luminescence spectrometer. UV–vis absorption spectrum was measured using the GBC Cintra 303 spectrometer.

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