



Highly efficient non-doped organic light emitting diodes (OLEDs) using anthracene derivatives with triphenylsilane unit



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ABSTRACT

A blue fluorescent emitting material for organic light emitting diodes (OLEDs) based on anthracene derivatives, 10-(4-(triphenylsilyl) phenyl)-9-(10-(4-(triphenylsilyl) phenyl) anthracen-9-yl) anthracene (**1**), was designed and synthesized via Suzuki coupling reaction. Multilayered OLED was fabricated with a device structure of: ITO (180 nm)/NPB (50 nm)/blue material (30 nm)/Bphen (30 nm)/LiQ (2 nm)/Al (100 nm). A device using **1** exhibits high efficiencies (1.85 cd/A, 1.05 lm/W and 0.63% at 100 mA/cm²). Also, the device incorporating **1** shows sky-blue emission with the CIE_{x,y} coordinates of (0.26, 0.49) at 6.0 V.

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1. Introduction

Organic light-emitting diodes (OLEDs) have attracted scientific and commercial attention for next generation flat panel displays because of wide-viewing angle, low-voltage operation, high resolution, and fast response time [1]. Even though many studies have been led to design and synthesize organic light-emitting materials for improving performance for full color OLEDs, efficient and stable blue materials are still insufficient compared to green and red materials [2]. Recently, a variety of blue-emitting materials based on anthracene [3–5], styrylarylene [6], fluorene [7], fluoranthene [8] and pyrene [9] have been reported. Among those, anthracene derivatives have attracted much research interests due to their outstanding photoluminescence (PL) and electroluminescence (EL) properties as well as good thermal properties [10–12]. Recently, triphenylsilane containing-anthracene derivatives have been reported to show outstanding EL performances [13,14].

Introduction of a triphenylsilane group into anthracene derivatives would reduce self-aggregation through steric hindrance within the bulky triphenylsilane moieties and lead the improved EL performances [15].

In this paper, we have designed and synthesized a blue emitting material, 10-(4-(triphenylsilyl) phenyl)-9-(10-(4-(triphenylsilyl) phenyl) anthracen-9-yl) anthracene (**1**), based on anthracene derivatives with triphenylsilane unit. This material has two non-planar triphenylsilane units bridged with bianthryl moiety, which consists of two anthracene unit connected by a single bond at the 9- and 9'-position.

In bianthryl moiety, two anthracene rings are almost perpendicular to each other with the dihedral angle of 89.4° because of the repulsive interaction of hydrogen atoms at the 1,1' and 8,8' position [16]. Therefore, bianthryl core as well as triphenylsilane moieties of material **1** provide the steric hindrance to the close-packing of the molecules and thus leads to improve EL performances. Furthermore, the non-planar structure of material **1** leads to larger intermolecular distances and more amorphous behavior in thin film consisting of material **1**.

2. Experimental details

2.1. Synthesis and characterization

¹H- and ¹³C-NMR were recorded on a Varian Unity Inova 300Nb spectrometer. FT-IR spectra were recorded using a Bruker VERTEX70 FT-IR spectrometer. Low-resolution mass spectra were measured using a Jeol JMS-600 spectrometer in the EI mode.

General procedure for the Suzuki cross-coupling reaction: Triphenyl(4-(4,4,5,5-tetramethyl-1,3-dioxolan-2-yl) phenyl) silane (2.3 mol) and the corresponding anthracene containing

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arylbromides (1.0 mol), Pd(PPh₃)₄ (0.08 mol), aqueous 2.0 M K₂CO₃ (10.0 mol), Aliquat 336, and toluene were mixed in a flask. The mixture was refluxed for 4 h at 120 °C. After the reaction had finished, the reaction mixture was extracted with ethyl acetate and washed with water. The organic layer was dried with anhydrous MgSO₄ and filtered with silica gel. The solution was then evaporated. The crude product was purified recrystallization from CH₂Cl₂/EtOH.

10-(4-(Triphenylsilyl) phenyl)-9-(4-(triphenylsilyl) phenyl) anthracen-9-yl) anthracene (**1**): ¹H-NMR (300 MHz, CDCl₃) [δ ppm]: 7.87 (t, *J* = 8.1 Hz, 9H), 7.76–7.73 (m, H), 7.65 (d, *J* = 8.1 Hz, 4H), 7.54–7.44 (m, 16H), 7.35 (t, *J* = 7.8 Hz, 5H), 7.24 (d, *J* = 7.2 Hz, 5H), 7.18–7.13 (m, 5H); ¹³C-NMR (75 MHz, CDCl₃) [δ ppm]: 140.4, 137.6, 136.6, 136.4, 133.6, 133.5, 131.4, 131.1, 130.0, 129.8, 128.1, 127.2, 127.1, 125.6, 125.3, FT-IR [ATR]: ν 3081, 2774, 1738, 1366, 1216 cm⁻¹. MS (EI⁺) *m/z* 22.9 (M⁺).

9,10-Bis(4-(triphenylsilyl) phenyl) anthracene [14] (**2**): ¹H-NMR (300 MHz, CDCl₃) [δ ppm]: 8.81 (d, *J* = 7.8 Hz, 4H), 7.75–7.6 (m, 14H), 7.52–7.43 (m, 22H), 7.38–7.34 (m, 6H), FT-IR [ATR]: ν 3033, 2774, 1738, 1366, 1216 cm⁻¹. MS (EI⁺) *m/z* 846.7 (M⁺).

2.2. Photophysical measurements

The UV–vis absorption and photoluminescence spectra of the new host materials were measured in dichloromethane (10⁻⁵ M) using Shimadzu UV-1650PC and Amincobrowman series 2 luminescence spectrometers. The fluorescence quantum yields of the emitting materials were determined in dichloromethane at 293 K against DPA (9,10-diphenylanthracene) as a reference ($\Phi_{\text{DPA}} = 0.90$). The HOMO (highest occupied molecular orbital) energy levels were measured with a low-energy photoelectron spectrometer (Riken-Keiki, AC-2). The energy band gaps were determined from the intersection of the absorption and photoluminescence spectra. LUMO (lowest unoccupied molecular orbital) energy levels were calculated by subtracting the corresponding optical band gap energies from the HOMO energy values.

2.3. Device fabrication and measurements

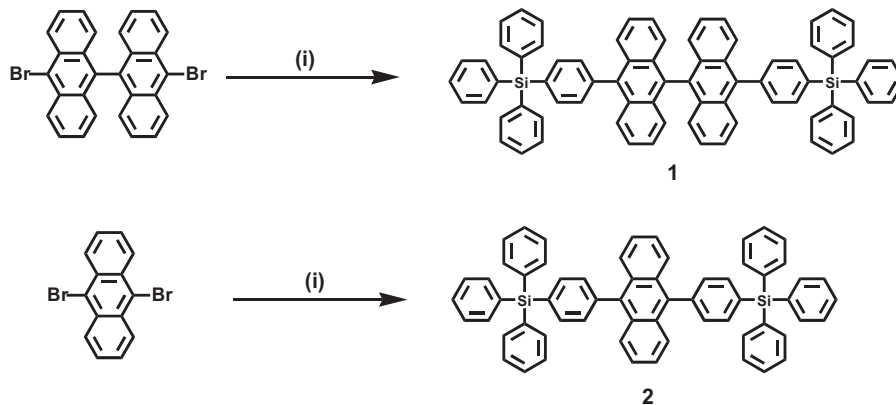
OLEDs using blue emitting molecules were fabricated by vacuum (5×10^{-7} Torr) thermal evaporation onto precleaned ITO-coated glass substrates. The OLEDs were fabricated in the following sequence: ITO (180 nm)/4,4-bis(*N*-(1-naphthyl)-*N*-phenylamino) biphenyl (NPB) (50 nm)/**1** or **2** (30 nm)/4,7-diphenyl-1,10-phenanthroline (Bphen) (30 nm)/lithium quinolate (Liq) (2 nm)/Al (100 nm). The current density (*J*), luminance (*L*), luminous efficiency (LE), and CIE chromaticity coordinates of

the OLEDs were measured with a Keithly 2400, Chroma meter CS-1000A. Electroluminescence was measured using a Roper Scientific Pro 300i.

3. Results and discussion

The molecular structure and synthetic route of anthracene derivative **1** are outlined in Scheme 1. The fluorescent-emitting material (**1**) was synthesized by Suzuki-coupling reaction with moderate yields. After purification by column chromatography and recrystallization, these designed compounds were fully characterized using ¹H- and ¹³C-NMR, infrared (IR), mass spectroscopy. For the comparison, material **2** (9,10-bis(4-(triphenylsilyl) phenyl) anthracene) was prepared by following the literature procedure [14]. The UV–vis absorption and photoluminescence (PL) spectra of emitting materials **1** and **2** in dichloromethane are shown in Fig. 1, and these results are summarized in Table 1. The absorption spectra showed three major peak with characteristic vibrational pattern at 350–410 nm because of the π–π* transitions of the anthracene core. The maximum absorption wavelengths of **1** and **2** appeared at 403 and 376 nm, respectively. Also, the maximum emission wavelengths of **1** and **2** were shown at 449 and 419 nm, respectively. Compared to material **2**, the maximum absorption and emission wavelengths of **1** showed red-shifts by 27 and 30 nm, respectively, in dichloromethane due to the extended π-conjugation length of material **1**. Interestingly, the maximum emission peak of **1** in solid film state showed slightly blue-shift by 10 nm in comparison with that in dichloromethane solution, while the maximum emission peak of **2** in solid film state showed red-shift by 22 nm in comparison with that in dichloromethane solution. These observations implied that the bianthryl moiety in material **1** provides the large steric hindrance to prevent the excimer-formation and thus induces blue-shift of emission spectrum of **1** in solid state.

To explore electroluminescent properties of these molecules, we fabricated devices using thermal deposition in the following configuration: ITO (180 nm)/4,4-bis(*N*-(1-naphthyl)-*N*-phenylamino) biphenyl (NPB) (50 nm)/**1** or **2** (30 nm)/4,7-diphenyl-1,10-phenanthroline (Bphen) (30 nm)/lithium quinolate (Liq) (2 nm)/Al (100 nm). NPB as the hole-transporting layer, Bphen as the electron-transporting layer, and Liq:Al as the composite cathode. The EL properties of devices **1** and **2** using material **1** and **2** as the emitting materials, respectively, are summarized in Table 2. Fig. 2(a) showed the EL spectra of devices **1** and **2**. The maximum EL peak of device **1** and **2** were 509 and 454 nm, respectively. Compared to device **2**, the EL spectra of device **1** showed red-shift by 55 nm due to differences in π-conjugation lengths of the



Scheme 1. Molecular structures and synthetic routes for emitting materials **1** and **2**. Reagents: (i) Triphenyl(4-(4,4,5,5-tetramethyl-1,3-dioxolan-2-yl) phenyl) silane, Pd (PPh₃)₄, K₂CO₃(2M), toluene, Aliquat 336.

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