

An efficient blue emitter based on a naphthalene indenofluorene core

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ABSTRACT

We designed and synthesized a novel sky-blue fluorescent dye, 7,7,13,13-tetramethyl-*N*⁵,*N*⁵,*N*¹¹,*N*¹¹-tetra-*p*-tolyl-7,13-dihydrobenzo[*g*]indeno[1,2-*b*]fluorene-5,11-diamine (DANIF), which contains a naphthalene indenofluorene (NIF) core and two diphenylamine end-capped groups. Because of the presence of steric diphenylamine terminal groups, DANIF showed a high glass transition temperature (T_g) of 161 °C and decomposition temperature (5% weight loss temperature) of 390 °C, which is suitable for the evaporation process. With 9,10-di(2-naphthyl)anthracene as the host and DANIF as the emitter, the fabricated vacuum-deposited organic light-emitting diodes exhibited a relatively low turn-on voltage of 2.8 V and a maximum luminous efficiency of 11.0 cd A⁻¹. The Commission Internationale de l'Eclairage coordinates of (0.15, 0.22) were achieved with a DANIF weight ratio of 7%. The combination of low driving voltage and high efficiency of the device gave rise to an impressive operational lifetime of 160 h (initial at 1000 cd m⁻², decay to 85% of the initial luminance). These results indicate that DANIF is a promising candidate for the development of highly efficient blue organic light-emitting diodes.

1. Introduction

Organic light-emitting diodes (OLEDs) have attracted considerable attention because of their potential application for the next generation full-color flat-panel displays and lighting applications [1–6]. In recent years, many efforts have been focused on the development of new light-emitting materials based on rational molecular design and optimization of device structures to improve the exciton utilization efficiency [7]. To achieve practical applications of OLEDs for full-color flat-panel displays, it is important to realize three primary color materials with high electroluminescent efficiencies, good thermal properties, and long device lifetimes [8,9]. Although great success has been achieved for red and green emitters, highly efficient blue emitters with long-term stability remain a challenge due to the inherent tradeoff between their energy gaps and ionization potential [10–12]. Typically, blue emitters have large band gaps (E_g) and relatively deep highest occupied molecular orbital energy levels (E_{HOMO}) [13–19]. Thus, it is difficult to inject charge carriers into the emitters and confine excitons exclusively to the emissive layer [20,21].

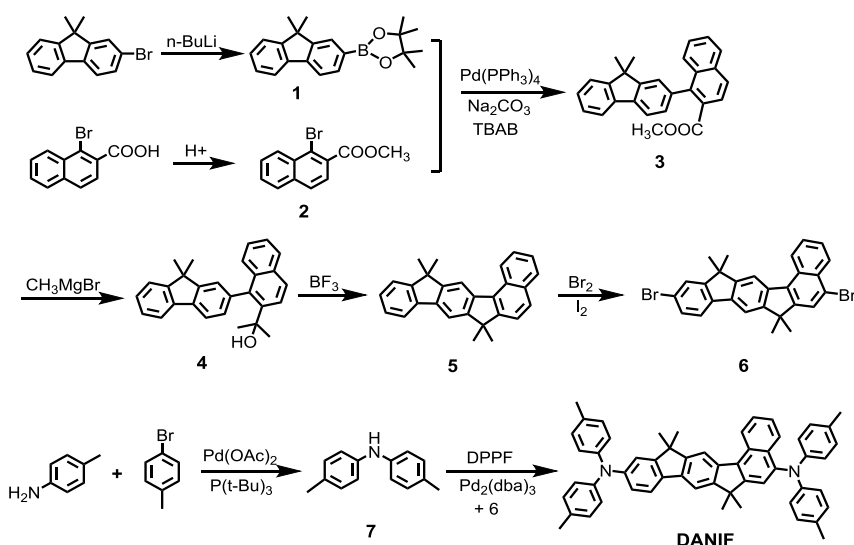
Although the use of an appropriate hole transport layer can mitigate the injection barriers between the anode and the emitters [22], unexpected low-energy emission from the exciplex or excimer may occur, which is detrimental to the color purity [23–25]. Recently, a range of potential small-molecule blue dyes consisting of styryl groups have been developed, such as *p*-bis(*p*-*N,N*-diphenyl-aminostyryl)-benzene (DSA-Ph) [26], 4,4'-bis(2,2-diphenylvinyl)-1,10-biphenyl (BCzVBi) [27], 4,4'-bis[2-(4-(*N,N*-diphenylamino)phenyl)vinyl]biphenyl (DPAVBi) [28], and so forth (Table S1 in the supporting information, SI). In general, these blue light-emitting devices are achieved by incorporating a certain ratio of these small-molecule dyes into appropriate host materials [29–34]. Besides the styryl group, large polycyclic aromatic building blocks are of particular interest due to the large π -conjugated length, high fluorescence quantum yield, and appropriate charge mobility [35,36]. For instance, Cha et al. reported a blue dopant materials 3,9-di(di(*p*-tolyl)aminospiro[benzo[*de*]-anthracene-7,9'-fluorene]) (DTSBAF), which exhibited a high luminous efficiency (LE) of 10.12 cd A⁻¹ with Commission Internationale de l'Eclairage (CIE) coordinates of (0.14, 0.25) [35]. Cha et al. reported a benzoanthracene-

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Scheme 1. Synthetic route of DANIF.

cored deep blue-emitting compound N^5, N^9 -bis(4-cyano-phenyl)- N^3, N^9 -diphenylspiro[benzo[de]anthracene-7,9'-fluorene]-3,9-diamine (CN-SBAF), while blended in a 10-(naphthalene-2-yl)-3-(phenanthrene-9-yl)spiro[benzo[*ij*]tetraphene-7,90-fluorene] (SBTF) host with a weight ratio of 10%, the resulted devices presented a high LE of 10.3 cd A^{-1} at 1000 cd m^{-2} with CIE coordinates of (0.14, 0.11) [37].

In this study, we developed a blue emitter with large polycyclic aromatic unit named as 7,7,13,13-tetramethyl- N^5, N^5, N^{11}, N^{11} -tetra-*p*-tolyl-7,13-dihydrobenzo[*g*]indeno[1,2-*b*]fluorene-5,11-diamine (DANIF) with comparable or even better performance with the reported blue small-molecule emitters (Table S1, SI). The DANIF consisted of diphenylamine terminal groups at the para-position of a naphthalene indenofluorene center, which could suppress the fluorescence-quenching interactions and decrease the non-radiative energy decay caused by intermolecular interactions. Considering that the central indenofluorene has expanded planar architecture, it should have good thermal stability and relatively high fluorescent quantum efficiency. With a widely used host material of 9,10-di(2-naphthyl)anthracene (ADN), the fabricated light-emitting diodes exhibited a high maximum LE of 11.0 cd A^{-1} with CIE coordinates of (0.15, 0.22). Of particular interest is that the OLED device based on DANIF exhibited impressive long-term operational stability, in which the luminance remained 85% of the initial value over 160 h at the initial luminance of 1000 cd m^{-2} .

2. Results and discussion

2.1. Synthesis and characterization

Scheme 1 shows the synthetic route of the resulting compound. Compound 2-(9,9-dimethyl-9H-fluorene-2-yl)-4,4,5,5-tetramethyl-1,3,2-

dioxaborolane (1) was synthesized based on 2-bromo-9,9-dimethyl-9H-fluorene by boron esterification at -78°C . Compound methyl 1-bromo-2-naphthoate (2) was prepared by esterification of 1-bromo-2-naphthoic acid and methanol as reaction monomers. A Suzuki cross-coupling reaction of 1 and 2 gave the intermediate methyl 1-(9,9-dimethyl-9H-fluorene-2-yl)-2-naphthoate (3) with a good yield of 80%. By treating 3 with the Grignard reagent CH_3MgBr at -78°C , followed by reaction with the Lewis acid BF_3 , compound 7,7,13,13-tetramethyl-7,13-dihydrobenzo[*g*]indeno[1,2-*b*]fluorene (5) was achieved with a good yield of 80%. The bromination of 5 with element bromine generated 5,11-dibromo-7,7,13,13-tetramethyl-7,13-dihydrobenzo[*g*]indeno[1,2-*b*]fluorene (6) with a good yield of 80%. The target 7,7,13,13-tetramethyl- N^5, N^5, N^{11}, N^{11} -tetra-*p*-tolyl-7,13-dihydrobenzo[*g*]indeno[1,2-*b*]fluorene-5,11-diamine (DANIF) was synthesized using a palladium catalyzed C–N coupling reaction of 6 with di-*p*-tolylamine (7) with a good yield of 78%. The molecular structure of DANIF was confirmed by ^1H and ^{13}C nuclear magnetic resonance (NMR) spectra (Fig. S1, SI) and MALDI TOF mass spectra (Fig. S2, SI).

2.2. Thermal properties

Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were used to evaluate the thermal properties of DANIF. As illustrated in Fig. 1a, DANIF exhibited a high decomposition temperature (T_d , 5% weight loss temperature) of 390°C . DSC (Fig. 1b) curve only showed an endothermic baseline shift yet without crystallization or melting peaks at raised temperatures. The glass transition temperature (T_g) was estimated to be 161°C . These results indicate that the DANIF possessed good thermal stability with amorphous properties and has the potential as an emitter for a vacuum thermal evaporation optoelectronic device [38].

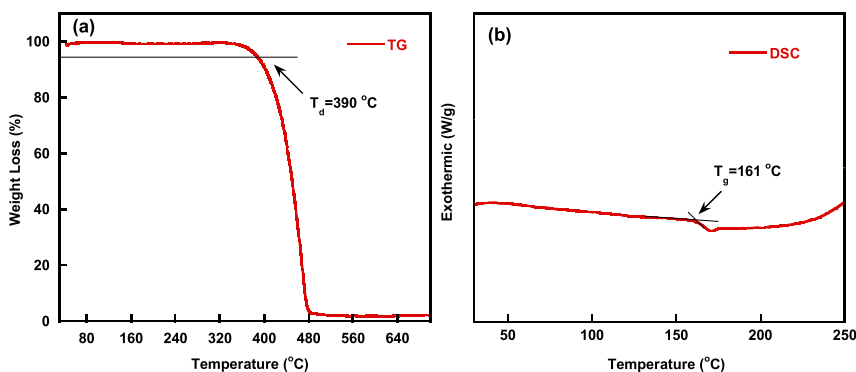


Fig. 1. (a)TGA and (b) DSC curves of DANIF.

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