

Benzimidazole/Amine-Based Compounds Capable of Ambipolar Transport for Application in Single-Layer Blue-Emitting OLEDs and as Hosts for Phosphorescent Emitters**

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Organic light-emitting diodes (OLEDs) have attracted a great deal of interest because of their potential applications in full-color flat-panel displays and lighting sources.^[1] Considerable progress has been made on both small-molecule- and polymer-based OLEDs. Though blue-, green-, and red-emitting materials are all needed for OLED applications, currently development of deep-blue emitters with good stability and high luminescence efficiency is deemed most critical in effectively reducing the power consumption and generating emission of different colors (including white light). In recent years considerable efforts have been shifted to the development of luminescent transition-metal complexes, particularly second- and third-row transition metals.^[2] As a result of efficient spin-orbit coupling in these complexes, both singlet and triplet excitons can be harvested, and theoretically up to 100% internal quantum efficiencies can be attained. However, metal complexes are typically crystalline and must be used as a dopant in an appropriate host. Furthermore, the host and materials used for carrier transport or carrier blocking must have sufficiently large triplet (T_1) energy to prevent the loss of triplet excitons from the metal complexes.^[3] Therefore, phosphorescence-based devices frequently have complicated structures. Since blue-emitting phosphorescent materials have relatively a large triplet energy, it becomes increasingly difficult to find host materials with a suitably high triplet state.^[4] Lastly, there are only few blue-emitting phosphorescent devices showing Commission Internationale de l'Eclairage (CIE) y values below 0.2 yet still with a respectable efficiency for the conversion of electrical into light energy.^[4c,e] Obviously, blue-emitting fluorescent

materials will still play a major role in the foreseeable future, and it would be useful if a single fluorescent molecule could be fabricated for application in a single-layer blue-emitting device. The simplified device structure would also help to limit the overall cost.

A prerequisite for high-performance single-layer OLEDs with a single component is to use molecules with high fluorescent quantum yields and balanced electron and hole mobilities. Equally important are good film-forming properties and an energy level matching that of the electrodes. Ambipolar molecules capable of carrying electrons and holes are likely to be candidates for single-layer devices. So far, quite a few ambipolar molecules have been used for OLEDs.^[5] However, most of them could not be fabricated as single-layer devices or exhibited only low efficiencies. Recently, a yellow-emitting single-layer device using a boron-containing material was reported to have good performance.^[6] We also successfully developed various high-efficiency single-layer devices, using materials ranging from yellow-emitting 3-cyano-9-diarylamino-carbazoles^[7] to a green-emitting dibenzothiophene-*S,S*-dioxide-based compound containing an arylamine unit.^[8] In the first case, external quantum efficiencies up to 1.4% and current efficiencies up to 5.1 cd A⁻¹ at 100 mA cm⁻² could be achieved. In the second case, a high external quantum efficiency of 3.1% and a current efficiency of 7.5 cd A⁻¹ were achieved at a current density of 100 mA cm⁻².

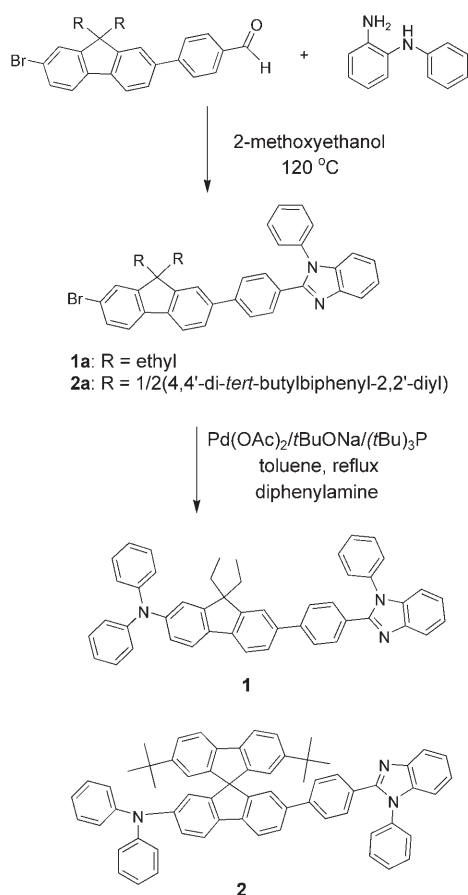
Herein, we report blue-emitting fluorescent molecules for high-performance single-layer devices. To the best of our knowledge, no single-layer blue-emitting devices using small molecules as the sole component have ever been reported to have acceptable efficiencies. In designing a blue-emitting molecule, one should avoid an extended conjugated π system and as well as strong charge-transfer character in the molecular skeleton. Moreover, the molecule should possess segments capable of effectively transporting electrons and holes suitable for the single-component single-layer devices. In view of the good electron mobility ($\approx 10^{-5}$ cm² V⁻¹ s⁻¹) of 1,3,5-tris(phenyl-2-benzimidazolyl)benzene (TPBI)^[9] and good hole mobility of arylamines ($\approx 10^{-3}$ – 10^{-5} cm² V⁻¹ s⁻¹),^[10] we decided to link an arylamine unit and a (phenyl-2-benzimidazolyl)benzene unit with a fluorene spacer that may also prevent strong charge transfer from the amine to imidazole.

Scheme 1 illustrates the synthesis of the new compounds **1** and **2** examined in this study. Standard procedures^[11] were followed to construct benzimidazoles from *N*-phenyl-*o*-phe-

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Scheme 1. Synthesis of compounds **1** and **2**.

nylenediamine and fluorene-containing aromatic aldehydes. The palladium-catalyzed C–N coupling reaction developed by Hartwig et al.^[12] was then adopted to incorporate the arylamine moiety. The compounds were isolated in good yields ($\approx 70\%$).

The thermal properties of compounds **1** and **2** were determined by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) (Table 1). Though the compounds are crystalline, they readily form a glass from their melts upon fast cooling, and the glassy state persists in the subsequent heating cycles. Replacement of fluorene by spirobifluorene^[13] increases the glass transition temperature by $\approx 60^\circ\text{C}$ (**2** vs. **1**). The thermal decomposition temperatures of the compounds are 373°C (**1**) and 467°C (**2**).

Photophysical properties of compounds **1** and **2** are also presented in Table 1. The absorption bands in the region around 370 nm are attributed to π – π^* transitions of the conjugated aromatic segments. The compounds are blue-emitting with very high solution quantum yields ($> 70\%$ in toluene and in the film).^[14] Each compound displays a quasi-reversible wave in its cyclic voltammogram (Table 1), which is attributable to the oxidation of the arylamine unit. No reduction waves were detected. The HOMO energy levels of the compounds were calculated from cyclic voltammetry and by comparison with ferrocene.^[15] These together with absorption spectra were then used to obtain the LUMO energy levels (Table 1).

Table 1: Physical data of the compounds.

	1	2
$T_m/T_g/T_d$ [$^\circ\text{C}$] ^[a]	284/104/373	396/170/467
λ_{abs} [nm] EA ^[b]	306, 370	309, 373
λ_{em} [nm] (Φ_f [%]) toluene ^[c]	428 (89)	428 (76)
λ_{em} [nm] (Φ_f [%]) film ^[c]	466 (80)	457 (72)
E_T [eV] ^[d]	2.3 (2.2)	2.3 (2.2)
E_{ox} (ΔE_p) [mV] ^[e]	386 (91)	413 (70)
HOMO/LUMO [eV] ^[f]	5.19/2.20	5.21/2.21

[a] The heating and cooling rates were 10Kmin^{-1} and 30Kmin^{-1} , respectively. T_m : melting point; T_g : glass transition temperature; T_d : decomposition temperature. [b] λ_{abs} : absorption maximum. EA = ethyl acetate. [c] λ_{em} : emission maximum. Measurement of quantum yields (Φ_f) in solution and in thin films are described in Ref. [14]. [d] Triplet energy measured in toluene at 77 K (and measured in film). [e] $E_{\text{ox}} = 1/2(E_{\text{pa}} + E_{\text{pc}})$, $\Delta E_p = E_{\text{pa}} - E_{\text{pc}}$, where E_{pa} and E_{pc} are anodic and cathodic potentials, respectively. Measured in CH_2Cl_2 . All the potentials are reported relative to ferrocene, which was used as the internal standard in each experiment. Ferrocene oxidation potential was located at $+272\text{ mV}$ relative to the Ag/AgNO_3 nonaqueous reference electrode. [f] The HOMO and LUMO energies were determined from cyclic voltammetry and absorption data.

Devices of different structures were fabricated for electroluminescence (EL) studies: Device A: indium tin oxide (ITO)/**1** or **2** (80 nm)/LiF (1 nm)/Al (150 nm); Device B: ITO/1,4-bis[(1-naphthylphenyl)amino]biphenyl (NPB, 40 nm)/**1** or **2** (40 nm)/LiF (1 nm)/Al (150 nm); Device C: ITO/**1** (40 nm)/1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBI, 40 nm)/LiF (1 nm)/Al (150 nm). 1,4-Bis[(1-naphthylphenyl)amino]biphenyl^[16] and 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene^[17] were used as the hole- and the electron-transporting materials, respectively, while LiF and Al were used as the electron-injection layer and the cathode. All devices are blue-emitting, and representative EL spectra are shown in Figure 1. The device performances are summarized in Table 2. Current–luminance (I – L) characteristics for representative devices are shown in Figure 2. It is worth noting that the unoptimized single-layer devices A show very promising performances: external quantum efficiency (η_{ext}) 2.4 %, maximum power efficiency ($\eta_{\text{p,max}}$) 1.2 lm W^{-1} , current efficiency (η_c) 2.4 cd A^{-1} , luminance (L) 2378 cd m^{-2} , and CIE indices [x,y] (0.15, 0.12) at 100 mA cm^{-2} for **1**, and respective values of 1.4 %, 0.9 lm W^{-1} , 1.8 cd A^{-1} , 1750 cd m^{-2} , and (0.15, 0.15) at 100 mA cm^{-2} for **2**. The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates of the two devices are shown in Figure 3. The performance of the single-layer device of **1** compares favorably with those of non-doped blue-emitting devices reported in literature.^[18] Among the devices A–C for the same compound, device B has the best performance, but device A gives performances similar to those of device B. No discernible light emission was detected for device C made with compound **2**. Device B with **1** appears to have the best performance of all the blue-emitting devices reported here: 2.8 %, 1.9 lm W^{-1} , 2.6 cd A^{-1} , and 2626 cd m^{-2} at 100 mA cm^{-2} .

The triplet energies of **1** and **2** were measured to be 2.2 and 2.3 eV in the film state and in toluene solution, respectively.^[19] We thus estimate that they may act as appropriate hosts for orange/red-emitting phosphorescent

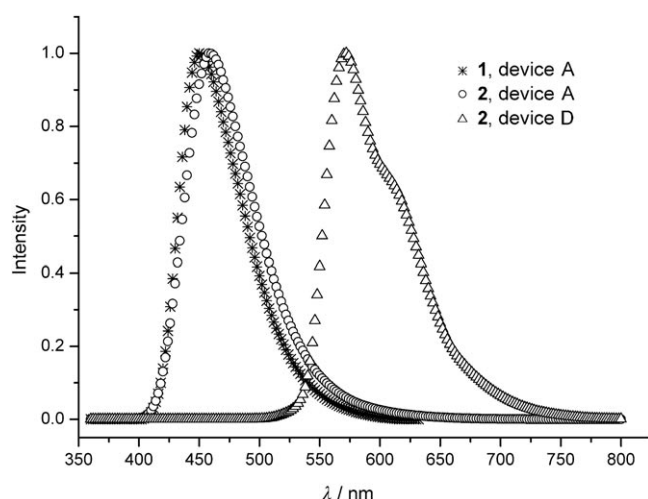


Figure 1. The EL spectra of the devices, in which, for example, “1, device A” denotes device A made with compound **1**. Note the EL spectra of devices B and C made with compounds **1** and **2** are essentially the same as those of device A made with **1** and **2**, respectively (not shown here).

Table 2: Electroluminescent data of the devices.^[a]

	1	2
V_{on} [V]	2.9; 2.7; 5.0	2.9; 2.7; 3.1
L_{max} [cd m^{-2}]	16178 (13.5);	10011 (13.5);
(V at L_{max} , V)	21663 (14.0);	10418 (14.0);
	5713 (20.5)	79750 (21.5)
λ_{em} [nm]	452, 456, 446	458; 454; 572
CIE [x,y]	0.15, 0.12;	0.15, 0.15;
	0.14, 0.11;	0.15, 0.12;
	0.15, 0.12	0.53, 0.47
fwhm [nm]	60; 62; 64	68; 62; 76
$\eta_{ext,max}$ [%]	2.5; 3.0; 2.0	1.4; 1.6; 7.8
$\eta_{p,max}$ [lm W^{-1}]	2.0; 2.3; 0.7	1.1; 1.6; 7.5
$\eta_{c,max}$ [cd A^{-1}]	2.5; 2.8; 2.2	1.8; 1.7; 22
L [cd m^{-2}] ^[b]	2378; 2626; 1125	1750; 1432; 19637
η_{ext} [%] ^[b]	2.4; 2.8; 1.2	1.4; 1.4; 6.9
η_p [lm W^{-1}] ^[b]	1.2; 1.9; 0.24	0.9; 0.9; 4.4
η_c [cd A^{-1}] ^[b]	2.4; 2.6; 1.2	1.8; 1.4; 20

[a] The measured values are given in order of the devices A, B, and C for **1** and A, B, and D for **2**. Abbreviations: L_{max} : maximum luminance; V_{on} : turn-on voltage; V: voltage; λ_{em} : emission wavelength; CIE [x,y]: Commission Internationale de l'Eclairage coordinates; fwhm: full width at half maximum; $\eta_{ext,max}$: maximum external quantum efficiency; $\eta_{p,max}$: maximum power efficiency; $\eta_{c,max}$: maximum current efficiency; L : luminance; η_{ext} : external quantum efficiency; η_p : power efficiency; η_c : current efficiency. V_{on} was obtained from the x-intercept of $\log(\text{luminance})$ vs applied voltage plot. [b] Measured at a current density of 100 mA cm^{-2} .

materials. Indeed, we successfully used compound **2** as the host for an orange-yellow phosphorescent iridium complex, $[(\text{fbi})_2\text{Ir}(\text{acac})]$ ($\text{fbi} = 2-(9,9\text{-diethyl-9H-fluoren-2-yl})-1\text{-phenyl-1H-benzimidazole}$; $\text{acac} = \text{acetylacetonate}$).^[20] The single-layer device D (ITO/**2** with 5% $[(\text{fbi})_2\text{Ir}(\text{acac})]$ (80 nm)/LiF (1 nm)/Al (150 nm)) has efficiencies (6.9%, 4.4 lm W^{-1} , 20 cd A^{-1} , 19637 cd m^{-2} at 100 mA cm^{-2}) rivaling those of the multilayer device with 4,4'- N,N' -dicarbazole-

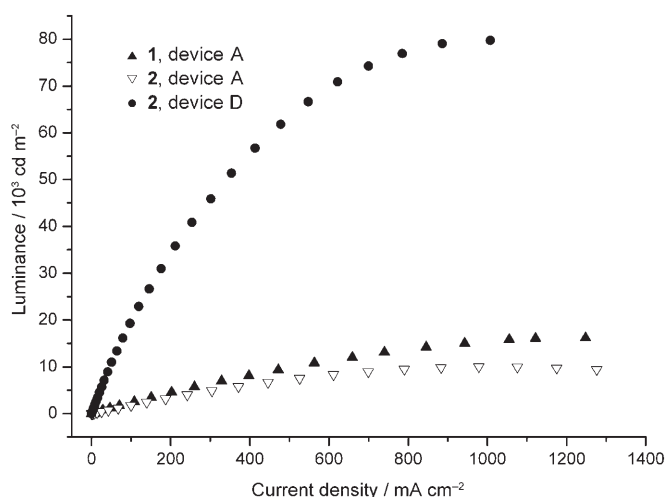


Figure 2. Plot of luminance versus current density for devices A and C.

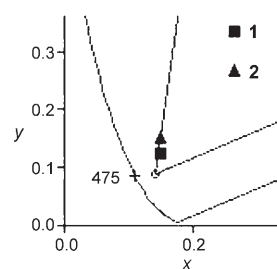


Figure 3. CIE coordinates for devices A made with compounds **1** and **2** (the cross denotes 475 nm).

biphenyl (CBP) as the host.^[20] The EL spectrum and the I - L characteristics of device D are shown in Figure 1 and Figure 2, respectively, and relevant performance parameters for the device are summarized in Table 2.

The carrier mobilities for the amorphous compounds **1** and **2** were measured by the time-of-flight (TOF) transient photocurrent technique^[21] in vacuum at room temperature. Both compounds exhibit nondispersive electron- and hole-transport characteristics as indicated by the TOF transients. The field-dependence of mobilities determined from the TOF transients (Figure 3) follows the nearly universal Poole-Frenkel relationship: $\mu \propto \exp(\beta E^{1/2})$, where β is the Poole-Frenkel factor.^[22] The electron mobilities ($\approx 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) for both compounds are comparable to those reported for commonly used the electron-transporting materials TPBI and tris(8-hydroxyquinoline) aluminum (Alq3).^[23] In contrast, the hole mobilities ($\approx 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) for **1** and **2** are lower than that of the typical HTL material NPB ($\approx 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$).^[24] Compound **1** has slightly higher mobility for electrons than for holes, and the reverse trend was found in **2** (Figure 4). The rather balanced and effective bipolar carrier transport (with hole and electron mobilities both in the $10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ range) is further proof of the successful single-layer devices in this study.

In summary, we have developed a convenient synthesis of highly emissive compounds containing benzimidazole and

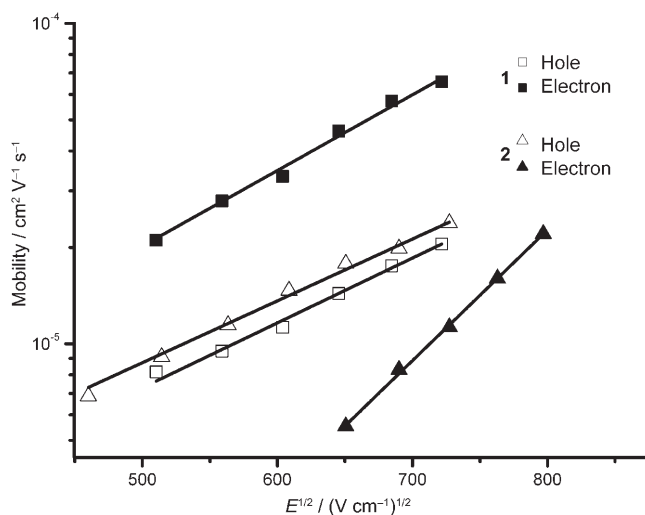


Figure 4. Electron and hole mobilities versus $E^{1/2}$ for films of 1 and 2.

arylamine units. These blue-emitting compounds exhibit intriguing ambipolar carrier-transport properties and were used to fabricate single-layer blue-emitting EL devices with very promising performances comparable to those of multi-layer blue-emitting devices. High-performance single-layer devices with a phosphorescent dopant and these compounds as the hosts were also demonstrated. The concept presented could be extended potentially to further generate a new class of advanced materials for full-color OLED displays or white-emitting OLEDs.

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carriers. Depending on the polarity of the applied bias, selected photogenerated carriers (holes or electrons) are swept across the sample thickness D , the applied electric field E is then V/D , and the carrier mobility is given by $\mu = D/(t_T E) = D^2/(V t_T)$.

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