



## Configuration effect of novel bipolar triazole/carbazole-based host materials on the performance of phosphorescent OLED devices

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### ABSTRACT

A series of structurally isomeric carbazole/triazole (TAZ)-based bipolar host materials **1–4** were designed and synthesized. These new materials were found to exhibit wide energy gaps ( $E_g$ : 3.29–3.52 eV), high triplet energies ( $E_T$ : 2.56–2.76 eV), high thermal stability ( $T_d$ : 426–454 °C), high glass-transition temperatures ( $T_g$ : 116–156 °C) and excellent film-forming property. Green and blue emitting devices with *fac*-tris(2-phenylpyridine)iridium ( $\text{Ir}(\text{ppy})_3$ ) and iridium(III) bis(4,6-(di-fluorophenyl)pyridinato- $N,C^2$ )picolinate (Flrpic) as phosphorescent dopants have been fabricated. The measurements of turn-on voltages, efficiencies and luminance suggested that the practice of combining carbazole's high triplet energy and excellent hole-transporting ability with TAZ's electron-transporting ability at the molecular level was effectively translated into better performance at the device level. The molecular structure of compound **4** is well-correlated with its efficiencies, which (32.7 and 21.1 cd/A for green and blue devices, respectively) were the best among the four materials.

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### 1. Introduction

Organic light emitting diodes (OLEDs) have attracted much attention from both the academic and industrial communities for their practical applications in displays and solid state lighting [1–3]. Compared to fluorescent dopants, the phosphorescent dopants have become a main stream materials system due to their much higher internal quantum efficiencies (close to 100%) as a result of harvesting both singlet and triplet excitons [4–13]. Choosing dopant materials is mainly focused on the cyclometalated iridium(III) complexes. In recent years, platinum(II) [14,15]

and copper(I) [16,17] complexes have also received increased attention because of their better device stability and the lower cost, respectively. On the other hand, the design of a proper host material matching well with its corresponding phosphorescent dopants is of crucial importance for OLED performance improvement [18]. To ensure an exothermic energy transfer from the host to the dopant and a confinement of triplet excitons on the dopant, a larger HOMO–LUMO (HOMO – the highest occupied molecular orbital; LUMO – the lowest unoccupied molecular orbital) energy gap ( $E_g$ ) and a higher triplet energy ( $E_T$ ) level of the host materials than those of the phosphorescent dopants are needed. Furthermore, keeping the balance of carrier mobilities is very important to achieve high-efficiency phosphorescent OLEDs [19]. Generally, the hole mobilities of hole-transport materials such as 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) or 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPD) [20,21] are about three orders of magnitude higher than the electron mobilities of

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electron-transport materials (ETMs) such as tris(8-quinolinolato) aluminum ( $\text{Alq}_3$ ) and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) [22]. One successful and effective approach to balancing the hole and electron mobilities is to use bipolar host materials [23–25]. Recently many groups have been working on bipolar host materials using different electron-donating and electron-accepting units, which greatly enriched the host materials [26–33]. In these work, the devices based on the bipolar host materials demonstrated better performance than those based on 4,4'-*N,N'*-dicarbazole-biphenyl (CBP) and 1,3-bis(9-carbazolyl)benzene (mCP), which are the common references for green and blue host materials.

Carbazole moiety, which possesses sufficiently high triplet energy and hole mobility, has been widely used as a structural unit for developing OLED host materials. Some well-known examples are CBP [34], mCP [35], 3,5-di(*N'*-carbazolyl)tetraphenylsilane (SimCP) [36] and 4,4'-*N,N'*-dicarbazole-2,2'-dimethyl-biphenyl (CDBP) [37]. 1,2,4-triazole derivatives, on the other hand, have been proved to have excellent electron-transporting and hole-blocking abilities due to their highly electron-deficient TAZ moiety [38–42]. A good example of its derivative is 3-phenyl-4-(1'-naphthyl)-5-phenyl-1,2,4-triazole, which has been used as host for green emitters [43].

The efforts in searching better bipolar materials have never been exhausted. In this article, we report the design and synthesis of a series of novel bipolar host materials containing carbazole and 1,2,4-triazole building blocks (Scheme 1). We expect that varying coupling mode can tune the photophysical and electrochemical properties, and then modulate their energy gaps, energy levels and triplet energies of the four isomers. We incorporated these two functional moieties by different coupling modes or at different positions of the bridging phenyl ring, and anticipated that these host materials would have both electron- and hole-transporting properties. The thermal,

morphological, photophysical and electrochemical properties of the compound **1–4**, as well as the performance of the OLEDs hosted by the compounds have been investigated. Devices using  $\text{Ir}(\text{ppy})_3$  and  $\text{Flrpic}$  as the green and the blue dopants have been fabricated and evaluated. The relationship between the structures of these four isomers and properties is also discussed.

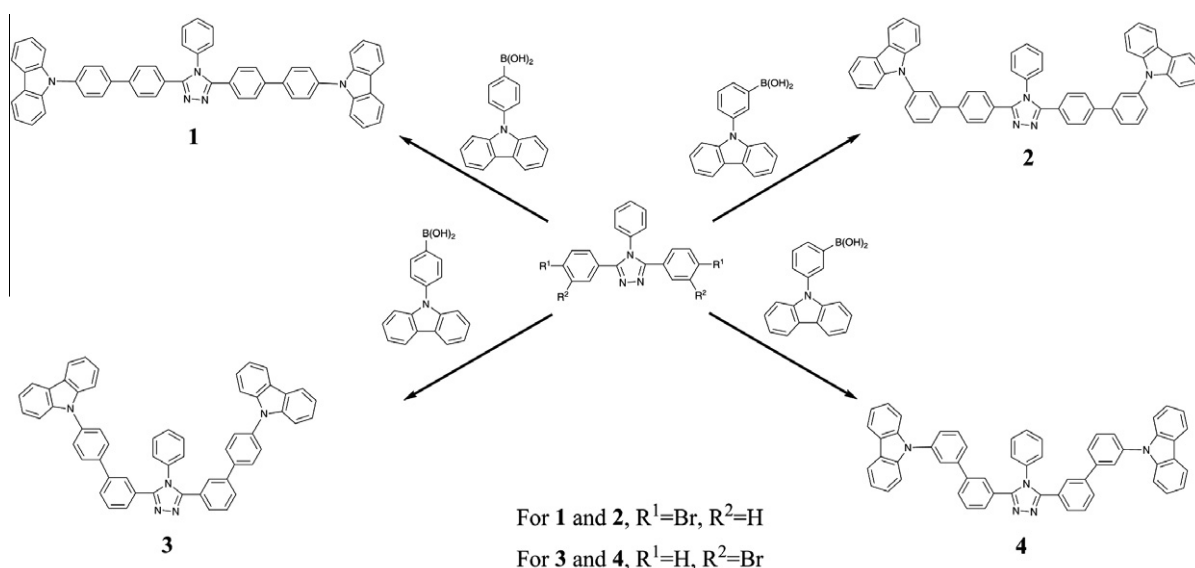
## 2. Results and discussion

### 2.1. Synthesis and characterization

Four bipolar isomeric host materials (**1–4**) were synthesized following the same Suzuki coupling reaction, starting from dibromo-substituted 1,2,4-triazole and boronic acid in this work. Their chemical structures and the synthetic routes to them are shown in Scheme 1. Four precursors, i.e., two dibromo-substituted 1,2,4-triazole and two 3-(or 4)-(9-phenyl-9*H*-carbazole) phenylboronic acid were prepared according to the previously reported procedure [44–46]. All compounds were purified with silica columns using petroleum ether, ethyl acetate and dichloromethane as eluants. Finally, these compounds were structurally confirmed by  $^1\text{H}$  NMR,  $^{13}\text{C}$  NMR, and mass spectrometry.

### 2.2. Thermal and morphological analysis

The thermal properties of these materials were investigated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) (Fig. S1 and S2 in Supporting information). Their decomposition temperatures ( $T_d$ ) are in the range 426–454 °C, as defined by 5% weight loss. The high  $T_d$  values suggest that these compounds can endure vacuum thermal sublimation at a reasonably high temperature during OLED device fabrication. They were also found to have very high glass-transition temperatures ( $T_g$ ) ranging from 116 to 156 °C, which are much higher



**Scheme 1.** Synthetic routes to **1–4** (reaction condition: reflux under argon with  $\text{Pd}(\text{PPh}_3)_4$  and  $\text{Na}_2\text{CO}_3$  in toluene/EtOH solvent).

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