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Solution-processible small-molecular host materials for high-performance phosphorescent organic light-emitting diodes

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ABSTRACT

Two novel carbazole-based molecules were synthesized by attaching 3,6-bis(3,6-di-tert-butyl-carbazol-9-yl)-carbazole and pyrazole to the dimethylbiphenyl core in a symmetric and asymmetric way. Owing to highly twisted configuration, they exhibit high triplet energy of 2.90 eV. They are suitable for spin coating to make thin films. The complete spatial separation of frontier molecular orbitals and the single-carrier devices study confirm the bipolar feature of pyrazole-containing material. They were used as hosts to fabricate phosphorescent organic light-emitting diodes by wet method. The green devices exhibited maximum efficiencies of 33 cd A^{-1} and 35 cd A^{-1}, which far exceed that (23 cd A^{-1}) of the control device with the polyvinylcarbazole host. The versatility of these hosts also spread to orange devices and peak efficiencies of 35 cd A^{-1} and 39 cd A^{-1} were achieved. The pyrazole-containing material always performed better due to its bipolar charge transporting nature.

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1. Introduction

Organic light-emitting diodes (OLEDs) are promising for practical applications in flat-panel displays as well as solid-state lighting [1–3]. Among these devices, tremendous efforts have been made in the development of phosphorescent organic light-emitting diodes (PhOLEDs) because they can approach 100% internal quantum efficiency in theory by harvesting both singlet and triplet excitons [4–6]. In general, host/dopant systems are used for PhOLEDs to suppress the severe concentration quenching and triplet–triplet annihilation [7]. Since the host occupies the majority weight of the emitting layer, the host is also essential to determine the overall device performance. In principle, an ideal phosphorescent host material should possess the following merits, such as (1) certain charge transporting ability to facilitate electric conduction of the whole device, (2) proper highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels to guarantee efficient charge injections and low driving voltage, (3) suitably high triplet energy to guarantee efficient forward energy transfer from the host to the metal complex dopant and to prevent reverse energy transfer [8,9]. In addition, the film-forming method of the host material directly determines the fabrication strategy of the OLED. It is well established that the solution-processing would be the final fabrication method of OLEDs for industrial application due to its easiness and low-price. Therefore, to be solution-processible is one valuable requirement for host materials. Accordingly, the solution-processed PhOLEDs have been extensively investigated in recent years [10–12]. The low-molecular-weight host materials that can be solution-processed seem to be one of the new trends, and more and more small-molecular host materials have been developed for PhOLEDs in recent years [13–15]. However, in addition to the tendency to crystallize, the poor solubility and weak film-forming ability by wet method are big challenges for small molecules. Therefore, it is of significant importance to develop amorphous small molecules with highly solubility and high glass transition temperature (T_g). Meanwhile, bipolar type host materials with asymmetric molecular structures have been demonstrated to perform better than the unipolar hosts due to the capability to balance charge transportations [16].

We report the design and synthesis of two carbazole-based small molecular compounds, **CMP** and **CPMP** (structures shown in Scheme 1), for application as hosts in solution-processed phosphorescent OLEDs. In these molecules, the twisted 2,2′-dimethylbiphenyl is selected as the molecular core in order to control the effective conjugation length of the whole molecules and thus to realize high triplet energy. The 3,6-bis(3,6-di-tert-butyl-carbazol-9-

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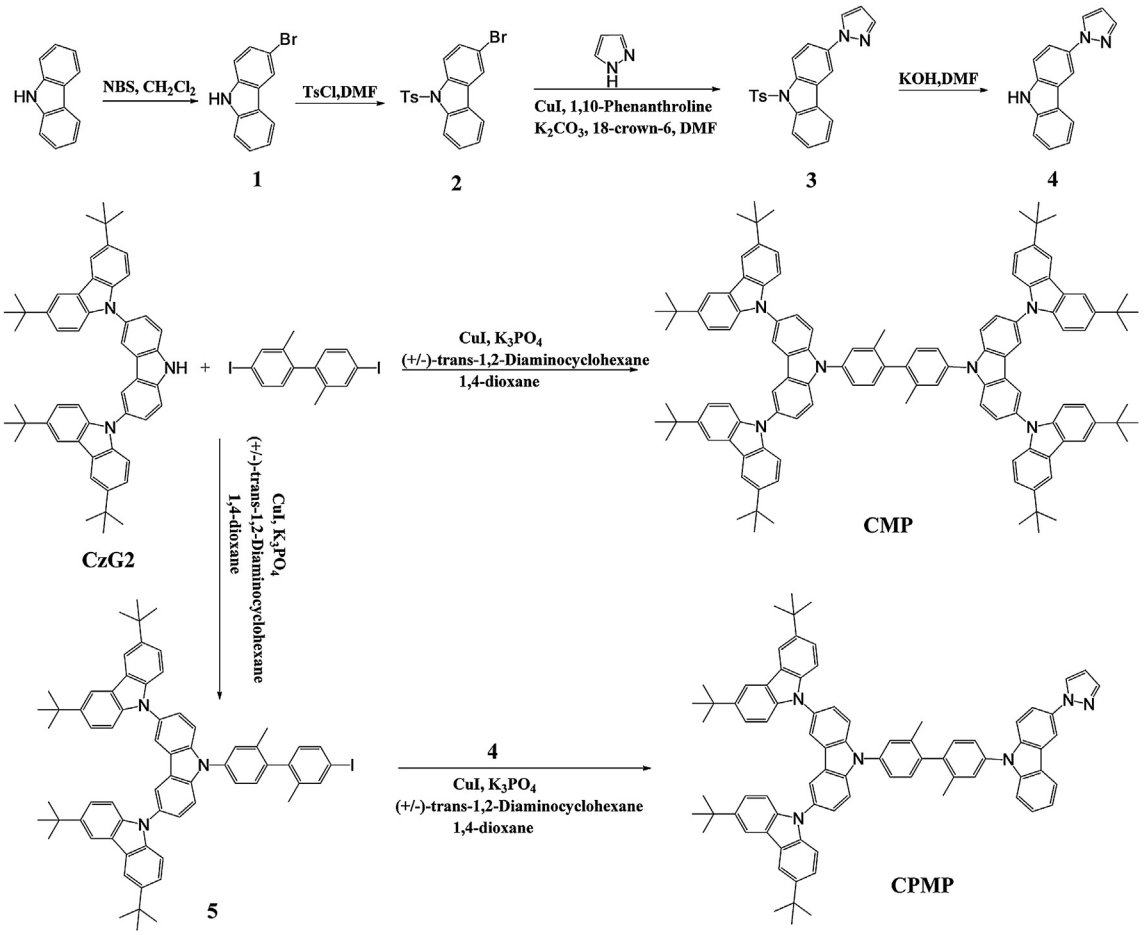

Scheme 1. Synthetic routes of **CMP** and **CPMP**.

yl)-carbazole was frequently used to construct host materials in literatures [7,17,18] and selected as the important building block in present study based on the following facts. First, it has high triplet energy because the linkage through sp^2-N does not cause effective conjugation between the outer and inner carbazoles. Second, the compounds containing this group are usually amorphous and possess high glass transition temperatures. **CMP** is designed by symmetrically attaching two 3,6-bis(3,6-di-tert-butyl-carbazol-9-yl)-carbazole groups to the terminals of the dimethylbiphenyl core. In the case of **CPMP**, pyrazole is attached to the 3-position of the inner carbazole unit on one side. Based on the electronegativity of some atoms in this heterocycle, it is expected to form the bipolar molecule by this asymmetric structure. *tert*-Butyls are grafted at the 3- and 6-sites of the outer carbazole rings to improve the solubility of these molecules. Both **CMP** and **CPMP** exhibit high triplet energies of 2.9 eV, amorphous feature with high T_g close to 300 °C, and good solubility and good film-forming ability by spin coating. Additionally, they show significantly shallow HOMO levels (−5.32 and −5.30 eV), which are close to that (−5.20 eV) of the widely used hole injecting material poly(ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and favourable for efficient hole injection when used as the emitting layer in OLEDs. Efficient green and orange PhOLEDs were fabricated with the emitting layer obtained by spin coating. The **CMP** and **CPMP**-hosted green PhOLEDs using $Ir(ppy)_3$ (Hppy = 2-phenyl-pyridine) as the dopant exhibited the maximum current efficiencies of 33 cd A^{-1} and 35 cd A^{-1}, respectively, which far exceed that (23 cd A^{-1}) of the control device with the traditional polyvinylcarbazole (PVK) host. The orange devices reached maximum current efficiencies of 35 cd A^{-1} and 39 cd A^{-1}, respectively. The single-carrier devices were fabricated for both materials. It was observed that **CPMP** exhibited higher electron current than **CMP** in the electron-only device, indicating the improved electron-transporting ability due to the incorporation of the pyrazole unit. The better performance of **CPMP** than **CMP** should benefit from its bipolar nature and the improved charge balance in emitting layer. As far as we know, this is the first report using pyrazole as n-type moiety to construct bipolar phosphorescent host materials for OLEDs application.

2. Experimentals

General information. ^{1}H NMR and ^{13}C NMR spectra were recorded on a 400 MHz and 100 MHz Varian Unity Inova spectrophotometer. Mass spectra were taken on MALDI micro MX and HP1100LC/MSD MS spectrometers. The fluorescence and UV–vis absorption spectra measurements were performed on a Perkin–Elmer LS55 spectrometer and a Perkin–Elmer Lambda 35 spectrophotometer, respectively. Phosphorescence spectra were measured on an Edinburgh FLS920 Spectrometer at 77 K in 2-MeTHF. Thermogravimetry analyses (TGA) and differential scanning calorimetry (DSC) measurements were carried out using Perkin–Elmer thermogravimeter (Model TGA7) and a Netzsch DSC 201 at a heating rate of 10 °C min^{-1} under a nitrogen atmosphere, respectively.

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