



Enabling high-efficiency organic light-emitting diodes with a cross-linkable electron confining hole transporting material



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ABSTRACT

Wet-process enables flexible, large area-size organic devices to be fabricated cost-effectively via roll-to-roll manufacturing. However, wet-processed devices often show comparatively poor performance due to the lack of solution-process feasible functional materials that exhibit robust mechanical properties. We demonstrate here a cross-linkable material, 3,6-bis(4-vinylphenyl)-9-ethylcarbazole (VPEC), to facilitate the injection of hole and meanwhile effectively confine electron to realize, for examples, high efficiency organic light-emitting diodes, especially at high luminance. The VPEC shows a hole mobility of $1 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and a triplet energy of 2.88 eV. Most importantly, the VPEC not only works for devices containing low band-gap red or green emitters, but also for the counterpart with high band-gap blue emitter. With the electron confining hole transporting material, the power efficiency of a studied red device, at $1,000 \text{ cd m}^{-2}$ for example, is increased from 8.5 to 13.5 lm W^{-1} , an increment of 59%, and the maximum luminance enhanced from 13,000 to $19,000 \text{ cd m}^{-2}$, an increment of 46%. For a high triplet energy blue emitter containing device, it is increased from 6.9 to 8.9 lm W^{-1} , an increment of 29%, and the maximum luminance enhanced from 9,000 to $11,000 \text{ cd m}^{-2}$, an increment of 22%.

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1. Introduction

Organic light-emitting diodes (OLEDs) have attracted much attention due to their potential in high quality full-color displays and solid-state lighting applications [1–11]. In order to expand their applicability, new fabrication techniques are constantly being developed. Typically, OLEDs are fabricated using either vacuum evaporation or solution process. The latter is deemed more superior in enabling flexible, large area size, roll-to-roll production, and consequently more cost-effective [12,13]. However, wet-processed OLED devices generally exhibit efficiency much lower than their dry-processed counterparts, especially at high luminance. Markedly improving the efficiency of solution-processable OLEDs is hence crucial.

Over the past years, numerous efficiency effective architectural approaches have been proposed. They include the employment of low interfacial resistance P–I–N structures [14–18], low

carrier-injection-barriers [19–22], balanced carrier injection [23–28], carrier and exciton confinement [29–33], stepwise emissive layers [34], carrier modulation layers [35–37], structures enabling exciton to generate on host or on both host and guest [38], structures facilitating host-to-guest energy transfer [39,40], and co-host structures [41,42], etc. All these approaches are valid for OLED devices fabricated via dry-process, but not all applicable if via wet-process. In particular, structures with ultra-thin layers, such as multi-emissive layers, modulation nanolayers, carrier and exciton confining layers, can hardly be realized into wet-processed OLED devices, unless certain special methods are adopted. This may explain why wet-processed OLEDs generally have device efficiency much lower than their dry-processed counterparts.

Nevertheless, several efficient wet-processed OLEDs, especially with a solution-processable emissive layer, have been reported. They include the employment of electroluminescence effective dyes [43–47] and hosts [48–53] as well as high mobility light emitting auxiliary materials [54,55,25], including hole transporting layers (HTLs), and electron transporting layers (ETLs). However, it is

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quite challenging to devise an organic material for sequential wet-process deposition of each layer without partial dissolution of underlying counterpart.

In past years, two major approaches have been developed to address the dissolution issue. The first approach is orthogonal sequential layer deposition. For example, Jenekhe's group reported a maximum current efficiency of 37.7 cd A^{-1} at $1,300 \text{ cd m}^{-2}$ for all solution-process blue phosphorescent OLED with a cesium carbonate (CsCO_3) doped 1,3,5-tris(m-pyrid-3-yl-phenyl)benzene (TmPyPB) ETL [56]. His group also reported a current efficiency of 53.8 cd A^{-1} at 5900 cd m^{-2} for a green device with a 4,7-diphenyl-1,10-phenanthroline (BPhen) ETL [57]. Perumal et al. reported a maximum efficacy of 22 lm W^{-1} (47 cd A^{-1}) at $1,000 \text{ cd m}^{-2}$ for a green device with copper thiocyanate as HIL/HTL function layer [58]. However, the choice of molecular organic materials available for orthogonal sequential deposition are rather limited because of their similar solubility characteristics.

The other alternative approach to realize all-solution-process fabricating is to use cross-linkable materials in the emissive layers (EMLs) and light emitting auxiliary layers, including hole injection layers (HILs), HTLs, ETLs, and electron injection layers (EILs). For example, Jen's group reported 5.6 lm W^{-1} at 800 cd m^{-2} for a white device by using a tris(4-carbazole)triphenylamine based cross-linkable HTL [59], and an external quantum efficiency (EQE) of 2.8% at 100 cd m^{-2} for a deep blue device by employing cross-linkable double HTLs [60]. Wong's group reported a maximum EQE of 1.4% for a green device by employing a 9,9-diarylfuorene-based triaryldiamine HTL [61]. Kido's group reported a maximum EQE of 2.3% for a blue device by using a carbazole derivative cross-linkable host material [62]. By using cross-linkable spirobifluorene-co-fluorene electroluminescent polymers, Meerholz's group reported a maximum current efficiency of 1.1 cd A^{-1} for a red device, 6.5 cd A^{-1} for a green device, and 3.0 cd A^{-1} for a blue device [63], and a maximum efficacy of 11.3 lm W^{-1} for a blue device with a cross-linkable TAPC-based HTL to reduce efficiency roll-off [64]. Lee's group reported 19.3 lm W^{-1} at 1000 cd m^{-2} for a green device by using a photo cross-linkable HTL [65]. Kim's group realized a maximum efficacy of 10.4 lm W^{-1} for green device by using a poly(azido-styrene)-random-poly(triphenylamine) copolymer as photo cross-linkable HTL [66]. So's group reported a current efficiency of 18 cd A^{-1} at 1000 cd m^{-2} for orange red device by employing a thermally cross-linkable N^2,N^7 -di(naphthalen-1-yl)- N^2,N^7 -diphenyl-9H-fluorene-2,7-diamine core based PLEXCORE[®] HTL [67]. Marder's group obtained, at 1000 cd m^{-2} , an efficacy of 11.8 lm W^{-1} and current efficiency of 41.9 cd A^{-1} for green device by using a rapid thermal processable copolymer as HTL with 3,6-bis(carbazol-9-yl)carbazole hole transporting unit and oxetane cross-linking unit [68].

To further improve the device efficiency especially at high applied voltage or high luminance, the employed HTL material, for example, should possess a lowest unoccupied molecular orbital (LUMO) higher than that of the guest or host so that the electrons entering in the EML can be effectively confined therein to lead to a higher device efficiency. This may explain why the wet-processed device by Meerholz's group showed a much less efficiency roll-off [64]. Many of such electron confining HTLs, however, fail to improve the device efficiency in the phosphorescent blue or even green light-emitting devices due to the lack of a high triplet energy to prevent the out-diffusion of the triplet excitons. It hence becomes highly challenging but necessary to design and synthesize a cross-linkable HTL material with an effective electron confining function, and meanwhile a high triplet energy. Specifically, the ideal triplet energy should be no less than 2.7 eV so that the highly desirable light-blue or even blue emission can be significantly enhanced accordingly.

We demonstrate here a novel cross-linkable HTL, 3,6-bis(4-vinylphenyl)-9-ethylcarbazole (VPEC), containing a carbazolyl moiety and two vinylphenyl groups, designed for solution-based OLEDs, where the carbazolyl moiety serves to provide a high triplet energy, while the vinylphenyl fragments to provide cross-linking function. Importantly, the material works not only for devices containing low band-gap red or green emitters, but also for the high band-gap blue or white counterparts for having a relatively high triplet energy (2.88 eV). Device efficiency improvement is especially marked at high luminance, also due to its relatively high LUMO energy-level (-2.0 eV). Taking the studied blue device at $1,000 \text{ cd m}^{-2}$ for example, its power efficiency is increased from 6.9 to 8.9 lm W^{-1} , an increment of 29%, and the maximum luminance enhanced from 9,000 to $11,000 \text{ cd m}^{-2}$, an increment of 22%.

2. Results and discussion

2.1. Synthesis

The cross-linkable derivative (3) was prepared as shown in Scheme 1. 3,6-Diiodo-9-ethylcarbazole (2) as a key material was synthesized firstly from commercially available 9-ethylcarbazole (1) by Tucker iodination with KI/KIO₃ in acetic acid. 3,6-bis(4-vinylphenyl)-9-ethylcarbazole (3) was prepared by Suzuki reaction of the di-iodo-derivative with an excess of 4-vinylphenyl boronic acid. The synthesized derivatives were identified by mass spectrometry, ¹H and ¹³C NMR spectroscopy. The data were found to be in good agreement with the proposed structures.

2.2. Hole mobility measurement by time of flight method

The hole mobility (μ_{th}) of the VPEC compound was characterized by the time of flight (TOF) technique [69,70]. In the double-logarithmic representation (Fig. S1), the carrier transit time (t_T) was determined from the kink point in the transient photocurrent curves. Inset of Fig. S1 shows the TOF transient curve for the holes of VPEC. The t_T with the applied bias (V) indicates the passage of holes through the $1.4 \mu\text{m}$ VPEC layer. The hole mobility was calculated by the formula of $\mu_{th} = d^2/(V \cdot t_T)$, where d is the VPEC layer thickness, V is the applied voltage [71,72]. Hole mobility determined for VPEC is shown as a function of square root of electric field ($E^{1/2}$) in Fig. 1. The resultant VPEC compound shows hole mobility of $1.0 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at an electric field of $3.3 \times 10^5 \text{ V cm}^{-1}$ (Table 1). As well established in literature, the resultant rational hole mobility of VPEC may be attributed because of the carbazole core group [73–75].

2.3. Thermal photophysical and electrochemical properties

As determined using Thermo-gravimetric analysis (TGA), the VPEC shows a 5% weight loss decomposition temperature of $350 \text{ }^\circ\text{C}$ (Fig. S2). Differential scanning calorimetry (DSC) was performed under nitrogen atmosphere at a heating rate of $10 \text{ }^\circ\text{C min}^{-1}$. The VPEC molecule exhibits a glass transition temperature (T_g) of $109 \text{ }^\circ\text{C}$, and a cross-linking temperature (T_x) ranging from 240 to $250 \text{ }^\circ\text{C}$ (Table 1 and Fig. 2). The exothermic peak of the VPEC compound was observed at $244 \text{ }^\circ\text{C}$ in the first heating curve of DSC thermogram corresponds to thermal polymerization of vinyl group, while no peak was resulted in the second heating scan (Fig. 2). The stable second heating thermogram is resulted because of cross-linking of VPEC during the thermal polymerization, which is extremely consistent with the previously reported cross-linkable hole transporting materials [76–81]. The sample from DSC pan is completely insoluble in common organic solvents, which further confirmed the cross-linking of VPEC compound.

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