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Crosslinkable hole-transporting small molecule as a mixed host for efficient solution-processed red organic light emitting diodes

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ABSTRACT

The fabrication of multilayer organic light-emitting diodes through solution processing presents numerous challenges, primarily concerning dissolution of the first layer during deposition of a second layer. Here, we demonstrate a highly efficient solution processed red organic light emitting diode utilizing an electron confining cross linkable small molecule, 9,9'-bis(4-vinylphenylmethylene)[3,3']bi-carbazole, as a hole transporting and co-host material. Compared with the corresponding single host device, the luminance of the mixed host device is enhanced from 8351 cd m⁻² to 15,200 cd m⁻², an increment of 82%, in addition, power efficiency and current efficiency also show an increment of 25% and 20% at 100 cd m⁻², respectively. Results show that use of the crosslinking material makes the hole injecting, poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate), layer more intact during the deposition of the emissive layer which improves the film quality and hence device brightness. The enhanced charge carrier balance and broadened exciton recombination zone due to the mixed host contribute to the improvement of efficiency and efficiency roll-off.

1. Introduction

Over the past few decades, organic light emitting diode (OLED) has gained incredible scientific and industrial awareness due to their potential in high quality full-color displays and solid state lighting applications [1–4]. OLEDs are usually fabricated either through thermal evaporation under high vacuum or through solution based depositions. Characteristically, former approach provides straightforward access to stacks of multiple layers such as charge transport or carrier blocking layers providing high-performance devices, however, bears the drawbacks of low material consumption rates, pitiable scalability, high capital cost, and difficulty in patterning [5]. The latter one is deemed more superior in enabling flexible, larger area size, roll-to-roll production, and subsequently more cost effective with high efficiency if we stay away from the dissolution or intermixing of previously applied layers during the sequential deposition of multilayer stacks [6]. To triumph over these problems one most promising approach is to insububilize the prior deposited layers using cross-linking [7].

Depending on the reactive monomers, crosslinking mechanism can be discriminated into photochemical triggered and thermally initiated processes [8]. Photo cross-linking enables the layer patterning at room temperature in very short period with the help of photoinitiators such

as oxetanes [9], cinnamates [10], chalcones [11], thiolene-click-reactions [12], azides [13], perfluoro azides [14] etc. This process has one very thoughtful weakness, formation of undesired impurities from the photoinitiators which may affect the operation of the devices. On the other hand, thermal crosslinking can be efficiently applied for any type of material via anneal at high temperatures (> 100 °C), the only one requirements being the presence of cross-linkable moieties in the material. Styrenes [15], acrylates [16], azides [17], trifluorovinylethers [18], phenylethynyl-groups [19], silanes [20], benzocyclobutenes [21], or alkyne-azide-cycloaddition [22] are thermal crosslinking functionalities and mechanisms which are mostly studied in solution-processable OLEDs.

From the perspective of high-efficiency device structures, multilayer structures with separated charge-transporting layers and emitting layers are essential. Nevertheless, it is not easy to comprehend multilayer structures by simple solution-processing due to the erosion of the underlying layers. In the recent years, numerous solution-processable hole-transporting materials have been synthesized, it is necessary to develop novel cross-linkable polymers to form robust films. In 2013, Lee's group reported a solution-processed hole-transporting layer based on the "Thiol-ene" photo cross-linking reaction [12]. In 2018, Ying's group stated a thermally cross-linkable hole-transporting poly

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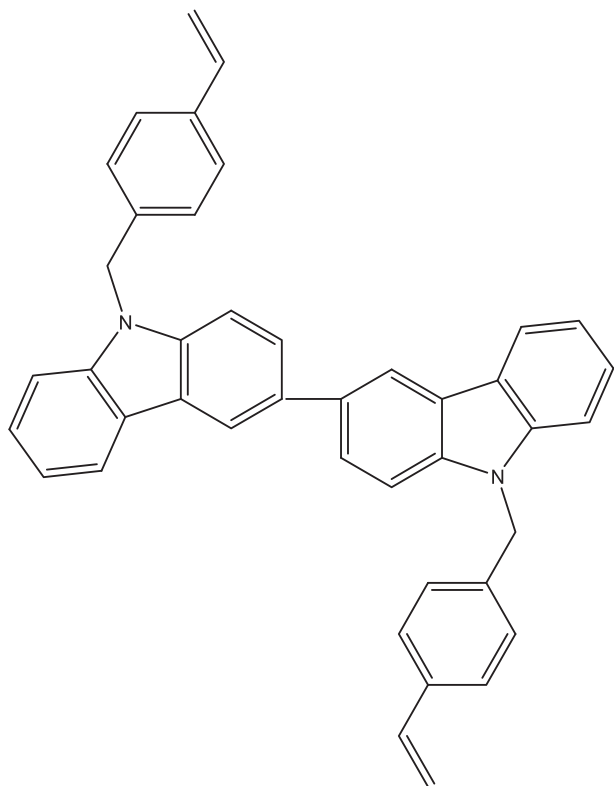


Fig. 1. Shows the molecular structure of the co-host material VPMC.

(indenofluorene-co-triphenylamine) copolymer (X-IFTPA) containing a vinyl-functionalized triphenylamine moiety for solution-processed OLED [23].

In this study, we demonstrate here a thermally activated electron confining cross-linkable hole-transporting material, 9,9'-bis(4-vinylphenylmethyl)[3,3']bi-carbazole (VPMC), as shown in Fig. 1, as a hole transport layer (HTL) and co-host with a bipolar material *N,N'*-Dicarbazolyl-4,4'-biphenyl (CBP) for red OLEDs. The incorporation of cross-linked HTL not only prevented dissolution of the initial layer while processing the subsequent wet depositions, but also lowered the hole-injection barrier of the OLEDs. Compared with the corresponding single-host device, co-host containing device shows better performance. Most prominently, we did not find any unfavorable influence of the crosslinking process on the OLED performance. The approach presented herein could become a general method for the decreasing the hole-injection barriers of OLEDs, improving the thin film quality of HTL and emissive layers (EML) and, eventually, lead to higher device performance.

Table 1

Effects of the presence and absence of hole transporting layer and co-host on the operation voltage (OV), power efficiency (PE), current efficiency (CE), external quantum efficiency (EQE), and CIE coordinates of the red OLED devices.

Device	Host CBP:DM 73	@ 100/1000 cd m ⁻²					Maximum Luminance (cd m ⁻²)
		OV[V]	PE[lm W ⁻¹]	CE[cd A ⁻¹]	EQE[%]	CIExy. coordinates	
R-1	100: 0	4.9/6.4	14.6/8.9	22.8/18.2	10.1/8.1	(0.57, 0.43)	8531
R-2	100: 10	4.3/5.9	15.9/9.4	23.5/18.6	10.6/8.5	(0.56, 0.44)	10,200
R-3	100: 20	4.2/5.6	18.3/11.5	27.4/21.4	13.6/9.9	(0.58, 0.42)	15,200
R-4	100: 30	4.5/6.0	13.2/8.2	19.9/17.0	8.8/7.4	(0.57, 0.42)	11,260
R-5	100: 40	4.9/6.5	12.5/7.9	18.0/15.2	8.4/6.7	(0.57, 0.43)	6563

2. Experimental section

2.1. Materials

Indium tin oxide (ITO) coated glass substrates with a sheet resistance of $15 \Omega^{-1}$ and light transmittance $> 84\%$ was purchased from the Luminescence Technology Corporation, Taiwan. The hole injection material poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) with a purity of 99.9% was purchased from UniRegion Bio-Tech Co., Taiwan. The sublimated grade 2,2',2''-(1,3,5-Benzinetriyl)-tris(-phenyl-1-H-benzimidazole) (TPBi) as an electron transporting material was purchased from the Luminescence Technology Corporation and 4,4'-bis(9-carbazolyl)-1,1'-biphenyl,4,4-*N,N'*-dicarbazole-1,1'-biphenyl (CBP) as the host material was purchased from Wang Shine Co. Lithium fluoride (LiF) (purity 99.95%) was purchased from Sigma-Aldrich. Aluminum (Al) ingots (99.999%) were purchased from Showa Chemical Co. Ltd. All materials were used without further purification.

2.2. Device fabrication

The solution-processed OLED device was fabricated with 150 nm thick patterned indium-tin oxide (ITO) glass. The ITO glass was cleaned with acetone and isopropyl alcohol with a sonication process and was rinsed in deionized water. Then, the ITO glass substrates were treated with UV-ozone to eliminate all organic impurities. The fabrication process included first spin-coating an aqueous solution of PEDOT:PSS at 4000 rpm for 20 s to form a hole injection layer (HIL) on pre-cleaned ITO anode and then dried at 120 °C for 20 min to remove residual solvent. The HTL was dissolved in chlorobenzene for 0.5 h and spin-coated at 3000 rpm for 20 s onto the HIL. After coating the HTL, the substrate was baked at 120 °C for 20 min to remove residual solvent, and then it was heated at 250 °C for 40 min for crosslinking reaction. Before depositing the following EML, the solution was prepared by dissolving the host and guest molecule in tetrahydrofuran at 40 °C for 0.5 h with stirring. The resulting solution was then spin-coated at 2500 rpm for 20 s under nitrogen. Followed by the TPBi, LiF, and Al, were deposited by thermal evaporation under high vacuum 6×10^{-4} Torr.

2.3. Thin films characterization and device measurements

The luminance, Commission Internationale de l'Eclairage (CIE) chromatic coordinates, and electroluminescence spectrum of the resulting OLED devices were measured by using Photo Research PR-655 spectra scan.

Keithley 2400 electrometer was used to measure the current-voltage (I-V) characteristics. The emission area of the devices was 2.5 mm², and only the luminance in the forward direction was measured. Surface topography of the films was studied using atomic force microscopy (AFM, Bruker, USA-INNOVA 1B3BE) in the trapping-mode.

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