



## Semi-orthogonal solution-processed polyfluorene derivative for multilayer blue polymer light-emitting diodes

Yawei Ma<sup>a</sup>, Feng Peng<sup>a,b</sup>, Ting Guo<sup>a</sup>, Congbiao Jiang<sup>a</sup>, Zhiming Zhong<sup>a,b,\*\*</sup>, Lei Ying<sup>a,b,\*</sup>, Jian Wang<sup>a</sup>, Wei Yang<sup>a</sup>, Junbiao Peng<sup>a,\*\*\*</sup>, Yong Cao<sup>a</sup>

<sup>a</sup> Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou, 510640, China

<sup>b</sup> South China Institute of Collaborative Innovation, Dongguan, 523808, China

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### ABSTRACT

Semi-orthogonal solution processing can circumvent interface intermixing issue of construction of multilayer polymer light-emitting diode devices (PLEDs). In this manuscript, we developed a new strategy to construct multilayer PLEDs by the semi-orthogonal solvents based on the difference between the solubilities of a poly(9,9-dioctylfluorene) that end-capped with a triphenylamine derivative (PFO-TF) (processed with *p*-xylene) and the light-emitting polymer PPF-SO15 or PPF-SO15-BT1 (processed with 1,4-dioxane). As the highest occupied molecular orbital energy level of PFO-TF (−5.70 eV) lies in between PEDOT:PSS (−5.20 eV) and the light-emitting copolymer PPF-SO15 (−5.94 eV), it can serve as the hole transport layer (HTL) to facilitate hole injection. The blue PLED based on PFO-TF as the HTL exhibits lower turn-on voltage and nearly identical electroluminescent spectra as that of the control device without HTL. Of particular importance is that the blue device with PFO-TF as the HTL exhibits the luminous efficiency over 5.0 cd A<sup>−1</sup>, which is much higher than that observed from a device without an HTL (3.51 cd A<sup>−1</sup>) or one with traditional poly(9-vinylcarbazole) as the HTL (4.74 cd A<sup>−1</sup>). Similar trend is also realized for PLEDs based on green-light-emitting polymer PPF-SO15-BT1. These observations indicate that processing of a multilayer device with this specific semi-orthogonal system may be a promising strategy for the construction of efficient PLEDs.

### 1. Introduction

Solution-processed organic light-emitting diodes have attracted much attention from both academia and the industrial community due to their great potential for the fabrication of large-area devices by using a low-cost printing technique [1–5]. To achieve high device performance, one of the most effective strategies is to use multilayer device structures (with tens of nanometers of each functional layers) that can facilitate charge injection and confine the excitons in the emissive layer (EML) [6–9]. Even though such multi-layer structure is easy to access with the thermo-evaporation process under high vacuum that can precisely control over the film thickness up to 1 nm, it remains a great challenge for the solution-processed technique because of the similar solubilities of organic semiconducting materials in organic solvents [10,11]. Thus, interface intermixing between different layers usually

occurs during the subsequent solution processing [12,13]. A variety of attempts have addressed this issue, such as developing certain materials with thermal cleavable side chains and introducing cross-linkable functional groups using orthogonal solution processing [14–24].

An alternative promising processing strategy to construct a multilayer device is the semi-orthogonal processing technique, which relies on the discrepant solubilities of the bottom layer and the adjacent layer [25–28]. The key issue with this processing technique is that the solvent used for processing the subsequent layer is a poor solvent for the bottom layer, such that the pre-fabricated bottom layer would not be damaged in the successive processing procedure. Compared to the other strategies, the semi-orthogonal solution-processing technique is very simple as it only requires certain materials that can be dissolved in appropriate solvents and does not require additional post-treatment or additional functional groups to facilitate solidification. For example,

\* Corresponding author. Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou, 510640, China.

\*\* Corresponding author. Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou, 510640, China.

\*\*\* Corresponding author.

E-mail addresses: [zmzhong@scici.cn](mailto:zmzhong@scici.cn) (Z. Zhong), [msleiyong@scut.edu.cn](mailto:msleiyong@scut.edu.cn) (L. Ying), [psjbpeng@scut.edu.cn](mailto:psjbpeng@scut.edu.cn) (J. Peng).

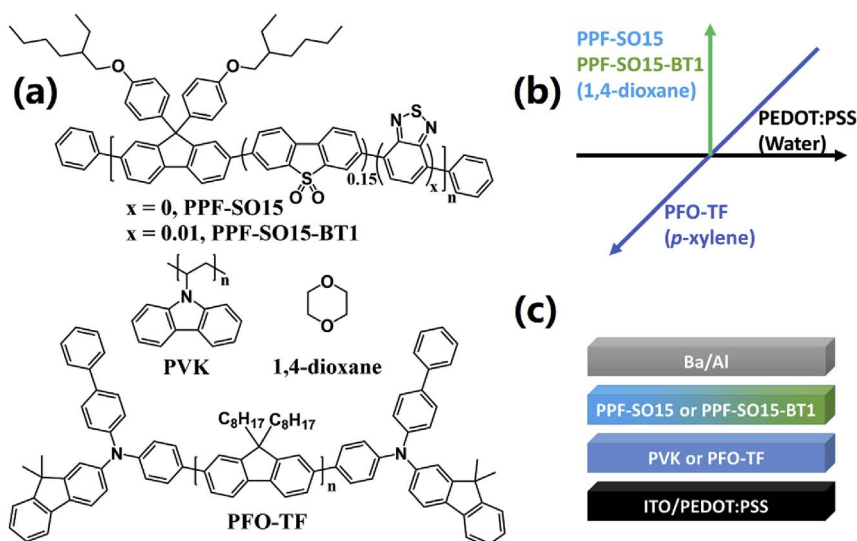


Fig. 1. (a) The molecular structure of the semiconducting polymers and the solvent 1,4-dioxane. (b) The three dimensional semi-orthogonal relationship of PEDOT:PSS, PFO-TF, and PPF-SO15 (or PPF-SO15-BT1). (c) The configuration of multilayer device structure.

one can deposit a light-emitting polymer from its *p*-xylene solution on the top of a prefabricated high molecular weight poly(9-vinyl-carbazole) (PVK) layer because the PVK can only be dissolved in chlorobenzene solution while emitter also exhibiting good resistance to *p*-xylene [9,29,30]. It is also interesting to note that the constructed multi-layer device presents obviously enhanced efficiency relative to single-layer devices, which is favorable for attaining high-performance devices [23].

Based on the aforementioned merits of the semi-orthogonal solution-processing technique, herein we introduced the construction of multi-layer devices by using a semi-orthogonal solvent system. In this device, we used a *p*-xylene-soluble polyfluorene derivative PFO-TF, which is end-capped with a triarylamine derivative *N*-([1,1'-biphenyl]-4-yl)-9,9-dimethyl-*N*-phenyl-9*H*-fluoren-2-amine as the HTL, and a blue light-emitting polyfluorene PPF-SO15, which comprises an *S,S*-dioxidibenzothiophene (15 mol%) moiety in the backbone and the alkoxy-substituted phenyl groups in the side groups, as the EML [28,31]. Fig. 1a shows the relevant molecular structures of these materials. Although both copolymers can be dissolved in *p*-xylene, only PPF-SO15 can be dissolved in 1,4-dioxane, whereas PFO-TF is not soluble. Unlike the traditional orthogonal solution-processed using solvents with extremely different polarity (e.g. *p*-xylene v.s. water, alcohol), the polarity of solvents for semi-orthogonal solution-processed can be relatively closed (e.g. *p*-xylene v.s. alkane [25]). This specific discrepancy in solubility allows for the construction of multi-layer device by casting PPF-SO15 from 1,4-dioxane solution on the top of the pre-fabricated PFO-TF layer. This semi-orthogonal solution processing material system provides an opportunity to utilize the polyfluorene derivative PFO-TF as the HTL. Compared to the single-layer PPF-SO15 device, the bilayer device with PFO-TF as the hole-transport and electron-blocking layer has a two-step hole-injection barrier, which is from the anode PEDOT:PSS to the PFO-TF and then from the PFO-TF to the EML of PPF-SO15. As a result, an efficient blue polymer light-emitting diode device (PLED) based on the PFO-TF/PPF-SO15 stacking structure was achieved through this semi-orthogonal solution processing procedure. Similar trend can also be realized by based on a green-light-emitting polymer PPF-SO15-BT1, which exhibits an impressively high performance that outperforms those based on PVK as the HTL.

## 2. Results and discussion

### 2.1. Solubility and solvent resistance

Both PPF-SO15 and PFO-TF can be easily dissolved in both *p*-xylene

at room temperature, with solubilities higher than 20 mg mL<sup>-1</sup>. However, the copolymer PPF-SO15 can be easily dissolved in 1,4-dioxane, whereas PFO-TF is barely dissolvable in 1,4-dioxane. Such obviously different solubilities of these two copolymers in 1,4-dioxane (see Table S2 in the Supporting Information, SI) can be attributed to their different side groups. From Fig. 1a one notes that the side groups of PFO-TF are composed entirely of hydrocarbon, whereas the side groups of PPF-SO15 consist of a 4-(2-ethylhexyloxy)phenyl group. Therefore, the better solubility of PPF-SO15 in 1,4-dioxane could be correlated to the bulkier side group than that of PFO-TF, or the enhanced interaction with 1,4-dioxane of the ether moiety in the side group of PPF-SO15, or both factors together. To confirm the solvent resistance of PFO-TF against 1,4-dioxane, the crude product of PFO-TF was particularly purified by Soxhlet extraction using 1,4-dioxane as the eluent after extraction with methanol, acetone and *n*-hexane.

According to the Beer-Lambert Law, absorption intensity is proportional to film thickness [18,20,32]. To confirm the semi-orthogonal solvent system for processing PPF-SO15 and PFO-TF, we measured the UV-vis absorption of these films before and after solvent rinsing. From Fig. 2a one notes that the UV-vis absorbance intensity of the PFO-TF thin film remains nearly unchanged before and after rinsing with 1,4-dioxane. This observation indicates the excellent 1,4-dioxane-resisting property of PFO-TF, which is better than the *p*-xylene-resisting property of the extensively used PVK (Fig. 2a). It is interesting to note that the absorbance edge of PFO-TF (Fig. 2b) shows an unobvious shift, which might be correlated with the  $\beta$ -phase of PFO-TF affected by solvent treatment [33–36]. Moreover, we measured the surface morphologies of the PFO-TF film before and after treatment with 1,4-dioxane by atomic force microscopy (AFM), with relevant images shown in Fig. 3. Both of the resulting films show aggregations across the entire film, whereas the morphology of the film treated with 1,4-dioxane is nearly identical with the pristine film, with both having quite similar root-mean-square roughness (RMS) values of about 2.2  $\pm$  0.1 nm [37].

### 2.2. Energy level alignment

The frontier molecular orbital energy levels of the used semiconducting polymers were evaluated by cyclic voltammetry measurements (Fig. 3a), in which a saturated calomel electrode is used as the reference electrode, a graphite is used as the working electrode, and tetra-*n*-butylammonium hexafluorophosphate (0.1 mol L<sup>-1</sup> in acetonitrile) is used as the electrolyte. Here, we used the ferrocene/ferrocenium (Fc/Fc<sup>+</sup>) redox couple as the standard. It is assumed that the Fc/Fc<sup>+</sup> redox couple has an absolute energy level of -4.8 eV relative to

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