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Efficiency enhancement of non-fullerene organic solar cells using PEDOT:PSS diluted with alcohol solvents as the hole transport layer

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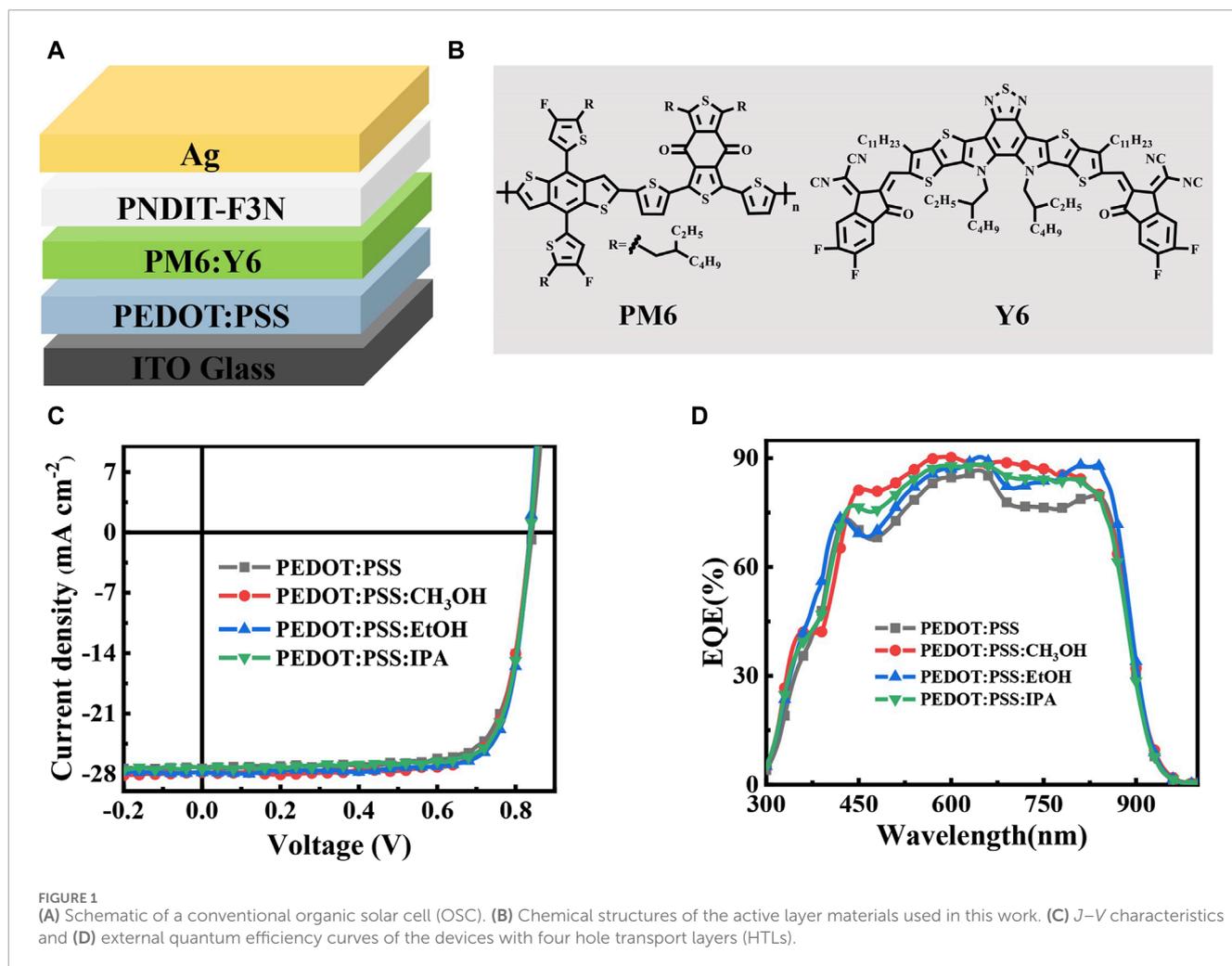
The poly(3,4-ethylenedioxythiophene) poly(styrene sulfonate) (PEDOT:PSS) hole transport layer (HTL) has always played a crucial role in achieving high-efficiency organic solar cells (OSCs) owing to its unique advantages of suitable energy levels and high optical transparency. However, the inherent insulation and easy aggregation property of PSS results in relatively low conductivity and high surface roughness of the PEDOT:PSS film, which is unfavorable for charge transport and the morphology of the top layer. To address these problems, we use PEDOT:PSS diluted in a series of alcoholic solvents and evaluate them on the PM6:Y6 system. Among these, the PM6:Y6 devices using PEDOT:PSS modified with ethanol as the HTL demonstrate the best vertical phase segregation and carrier extraction. In addition, the PEDOT:PSS film with added ethanol also has the smoothest surface compared to those diluted in the other alcoholic solvents. Finally, a high power conversion efficiency of 18.13% was obtained with the PM6:Y6 devices based on PEDOT:PSS modified by ethanol. This work provides a sufficient reference for the alcoholic modification of PEDOT:PSS and also proposes a feasible solution for high-efficiency OSCs.

KEYWORDS

organic solar cell, PEDOT:PSS, alcoholic solvent, phase component, morphology

1 Introduction

Organic solar cells (OSCs) have attracted extensive attention for solving the energy problem owing to their advantages, such as light weight, semitransparency, flexibility, and roll-to-roll production (Lin et al., 2015; Li et al., 2016; Lin et al., 2017; Zhao et al., 2017; Hou et al., 2018; Li et al., 2018; Li et al., 2024; Peng et al., 2024). Recently, the power conversion efficiency (PCE) of OSCs has improved significantly upon the emergence of non-fullerene acceptors that are matched well with the absorption spectra and energy levels of the donor (Sonar et al., 2011; Lin and Zhan, 2014; Cheng et al., 2018; Hou et al., 2018; Yan et al., 2018). Through interface modifications, semiconductor material design, and morphological optimization, the PCE of OSCs has



exceeded 19% (Cui et al., 2021; Li et al., 2021; Zhang et al., 2021; Chong et al., 2022; Sun et al., 2022; Zhu et al., 2022).

Suitable interface modifications can enhance charge extraction and suppress carrier recombination, thereby resulting in large photovoltages and photocurrent densities (Cheng et al., 2009; Zhang et al., 2018; Zhao et al., 2018; Zhu et al., 2021; Zhou X. M. et al., 2022; Liao et al., 2022; Yu et al., 2022; Zheng et al., 2022). The currently available high-efficiency OSCs are based on the conductive polymer poly(3,4-ethylenedioxy-thiophene) poly(styrene-sulfonate) (PEDOT:PSS) as the hole transport layer (HTL) because of its suitable energy levels, good optical transparency, and solution processability. In addition, the large work function of PEDOT:PSS (~ 5.1 eV) approaches the energy level of the least unoccupied molecular orbital of the most polymeric donor material, which is conductive enough to form good ohmic contacts at the anode/active layer interface and enhance hole transport efficiency (Potsavage et al., 2009; Bouthinon et al., 2015; Xu et al., 2020; Zhang et al., 2020; Zhu et al., 2021). However, there are also some drawbacks to using PEDOT:PSS, such as its relatively moderate conductivity, strong acidity, and structural inhomogeneities caused by the insulating PSS structure, which affect the photovoltaic performance and stability of the device (Kemerink et al.,

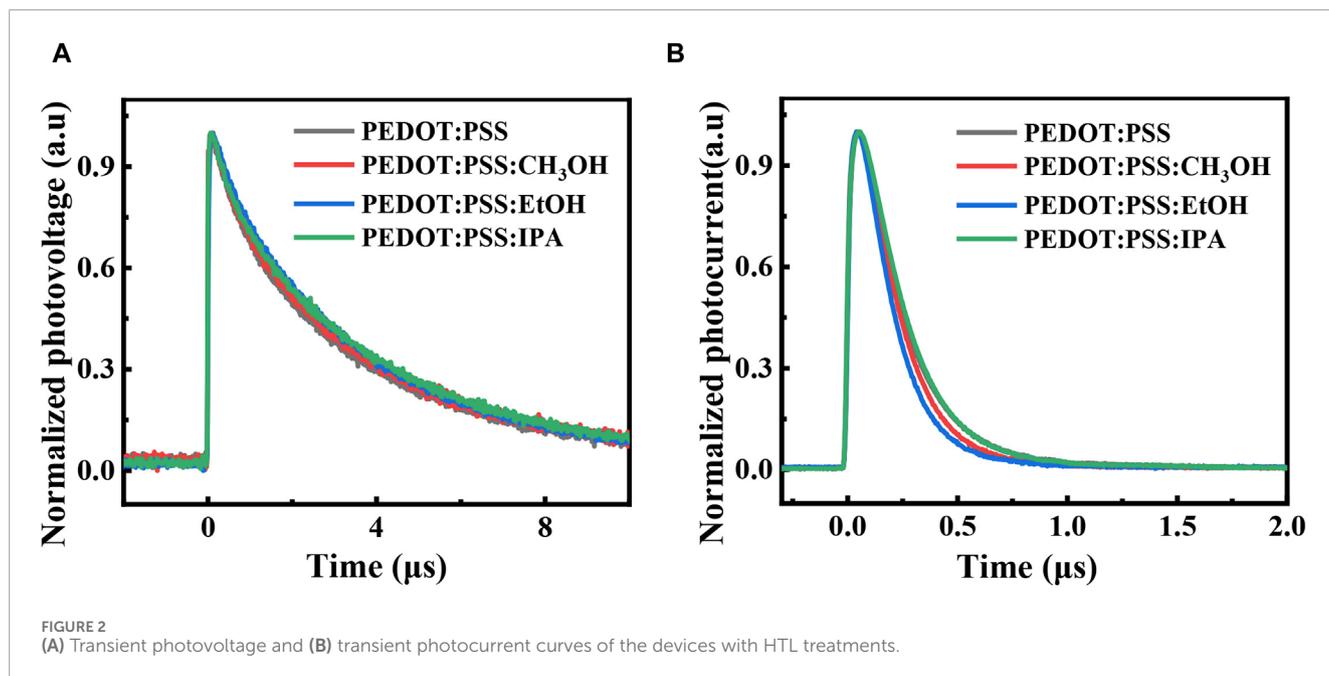
2004; Lee et al., 2014; Shi et al., 2015; Zeng et al., 2020; Zhou K. K. et al., 2022; Bertrandie et al., 2022).

It has been reported that isopropanol (IPA) can adjust the morphology, charge transport, and optical properties of a PEDOT:PSS film, thereby significantly improving its hole extraction and transport efficiencies (Ouyang, 2013; Donoval et al., 2017; Aimukhanov et al., 2021). Moreover, isopropanol can also hinder the insulating PSS portion in PEDOT:PSS, thereby hindering the aggregation of PSS and leading to a higher PCE in OSCs (Mengistie et al., 2015; Zhu et al., 2016; Aimukhanov et al., 2021; Cassinelli et al., 2021). Nevertheless, research on the effects of the types of alcohols used with the PEDOT:PSS solution on the photovoltaic performances of OSCs remains insufficient. Moreover, the effects of alcohol-modified PEDOT:PSS on the vertical component distribution of the active layer are not yet clear, seriously limiting the application of PEDOT:PSS to high-efficiency OSCs in the future.

Herein, we selected methanol, ethanol, and IPA as the diluents for the PEDOT:PSS solution to enhance the photovoltaic performances of the OSCs. Based on the transient photovoltage (TPV) and transient photocurrent (TPC) measurements, the effects of alcoholic solvent dilution of PEDOT:PSS on the

TABLE 1 Extracted performance parameters of the OSCs equipped with different HTLs under AM 1.5 G illumination at 100 mW cm^{-2} .

	V_{OC} (V) ^a	J_{SC} (mA cm^{-2}) ^a	FF (%) ^a	PCE (%) ^a	R_s (ohm)	R_{sh} (ohm)
PEDOT:PSS	0.841 ± 0.003	27.27 ± 0.27	76.00 ± 0.18	17.42 ± 0.21	34.07	37106.66
PEDOT:PSS:CH ₃ OH	0.835 ± 0.004	27.85 ± 0.14	77.19 ± 0.26	17.94 ± 0.05	24.15	25046.39
PEDOT:PSS:EtOH	0.834 ± 0.001	27.63 ± 0.32	78.66 ± 0.31	18.13 ± 0.19	18.31	54773.50
PEDOT:PSS:IPA	0.835 ± 0.001	27.32 ± 0.27	78.23 ± 0.43	17.86 ± 0.13	23.09	43787.40



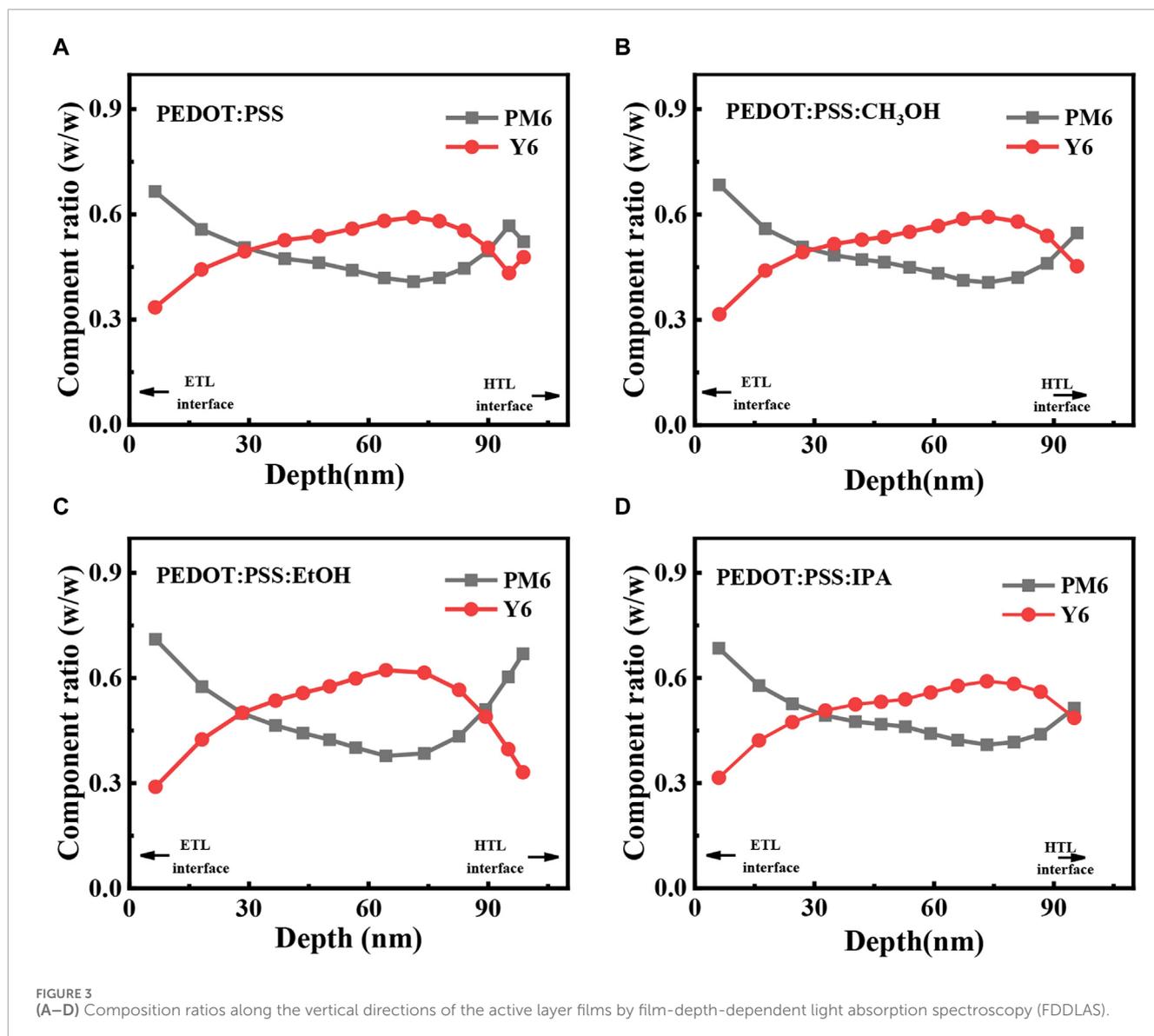
carrier recombinations and extraction process in devices were studied thoroughly. In addition, film-depth-dependent light absorption spectroscopy (FDDLAS) was employed to understand the phase component information in the active layer. The results of the investigations indicate that PM6:Y6 devices based on ethanol-modified PEDOT:PSS HTLs enable faster charge extractions and suitable phase distributions, which are conducive to efficient carrier and lower charge combinations. Thus, the corresponding OSCs exhibit a maximum PCE of 18.13%, which is much higher than the 17.42% of the control OSCs.

2 Materials and methods

PM6 and Y6 were purchased from Solarmer Material Inc. (Beijing, China), PNDIT-F3N was purchased from eFlexPV Limited (Guangdong, China), and PEDOT:PSS (Clevios PVP 4083) was purchased from Heraeus Inc. (Hanau, Germany). All of the other reagents and chemicals were purchased from Sigma-Aldrich or Aladdin and used as received.

3 Results and discussion

The schematics of the device structure and chemical structure of the photoactive layer are shown in Figures 1A, B, respectively. Clearly, the polymer donor PM6 and non-fullerene acceptor Y6 are selected as active layers, and the corresponding device architecture was stacked in the form of ITO/HTL/PM6:Y6/PNDIT-F3N/Ag. The related chemical structures of PEDOT:PSS and PNDIT-F3N are shown in Supplementary Figure S1. Subsequently, the current density vs. voltage (J - V) curves of OSCs with four different HTLs were measured under AM 1.5 G illumination at 100 mW cm^{-2} , as shown in Figure 1C. The extracted photovoltaic performance parameters from the J - V curves are summarized in Table 1. When the pure PEDOT:PSS was used as the HTL, a relatively low short-circuit current density (J_{SC}) of 27.27 mA cm^{-2} and poor fill factor (FF) of 76.00% were obtained, which resulted in a low PCE of 17.42%. When the alcoholic solvents (CH₃OH, EtOH, and IPA) were added to PEDOT:PSS, the PCE of the OSCs may be increased by the enhanced J_{SC} and large FF. The detailed optimizations of the volume ratio between PEDOT:PSS and the alcoholic solvent are shown in Supplementary Figure S2 and Supplementary Table S1. For the PEDOT:PSS:CH₃OH cast device, the J_{SC} and FF reached

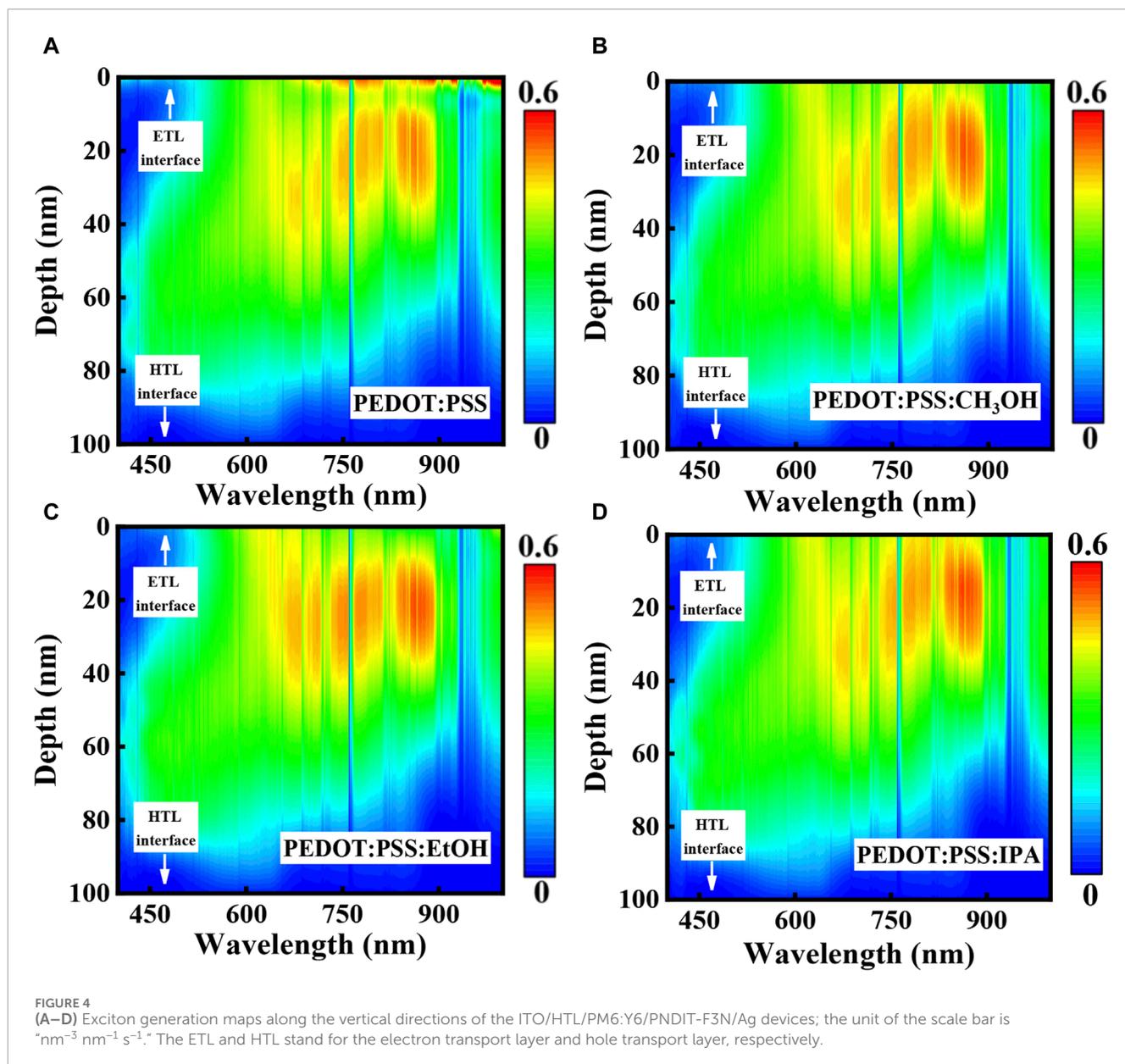


27.85 mA cm⁻² and 77.19%, respectively, resulting in a high PCE of 17.94%. When EtOH was introduced into the PEDOT:PSS solution, the FF of the device was greatly improved from 76.00% (PEDOT:PSS) to 78.66% (PEDOT:PSS:EtOH), resulting in an increased PCE of 18.15%. After deposition of PEDOT:PSS:IPA onto the ITO electrode, the PCE of the OSCs based on the PM6:Y6 system increased to 17.86%, which is a result of the modest increments in J_{SC} and FF (27.32 mA cm⁻² and 78.23%). Moreover, the external quantum efficiency (EQE) spectra of the OSCs with different HTLs were measured, as shown in Figure 1D. It can be seen that the EQE spectra of the devices equipped with PEDOT:PSS along with added alcoholic solvents exhibit remarkable enhancements in the range of 400–900 nm compared to the control device using pure PEDOT:PSS as the HTL. The stabilities of OSCs with different HTLs were measured in an N₂ atmosphere (Supplementary Figure S3). After 600 h of storage, the PCEs of the devices with PEDOT:PSS modified by alcoholic solvents retain above 95% of their initial values, higher than that of the control device (92.9%). In particular,

the PCE of the OSCs with PEDOT:PSS:EtOH as the HTL could still reach 96.3% of that of the pristine device after 600 h of exposure in the N₂ atmosphere.

To explore the reasons behind the improved photovoltaic performance, the TPV and TPC curves of the OSCs were analyzed for the effects of these HTLs on carrier recombinations and extraction processes (Figure 2A; Figure 2B). Based on the TPV curve, the extracted carrier lifetime ($\tau_{lifetime}$) values of the devices are 3.056 μ s (PEDOT:PSS), 3.232 μ s (PEDOT:PSS:CH₃OH), 3.338 μ s (PEDOT:PSS:EtOH), and 3.433 μ s (PEDOT:PSS:IPA). From Figure 2B, the related charge extraction time constant ($\tau_{extract}$) values are 0.227 μ s (PEDOT:PSS), 0.203 μ s (PEDOT:PSS:CH₃OH), 0.182 μ s (PEDOT:PSS:EtOH), and 0.226 μ s (PEDOT:PSS:IPA). After adding EtOH to PEDOT:PSS, the device exhibits the shortest $\tau_{extract}$ and suitable $\tau_{lifetime}$ as compared to the other OSCs, indicating reduced carrier loss and effective extraction.

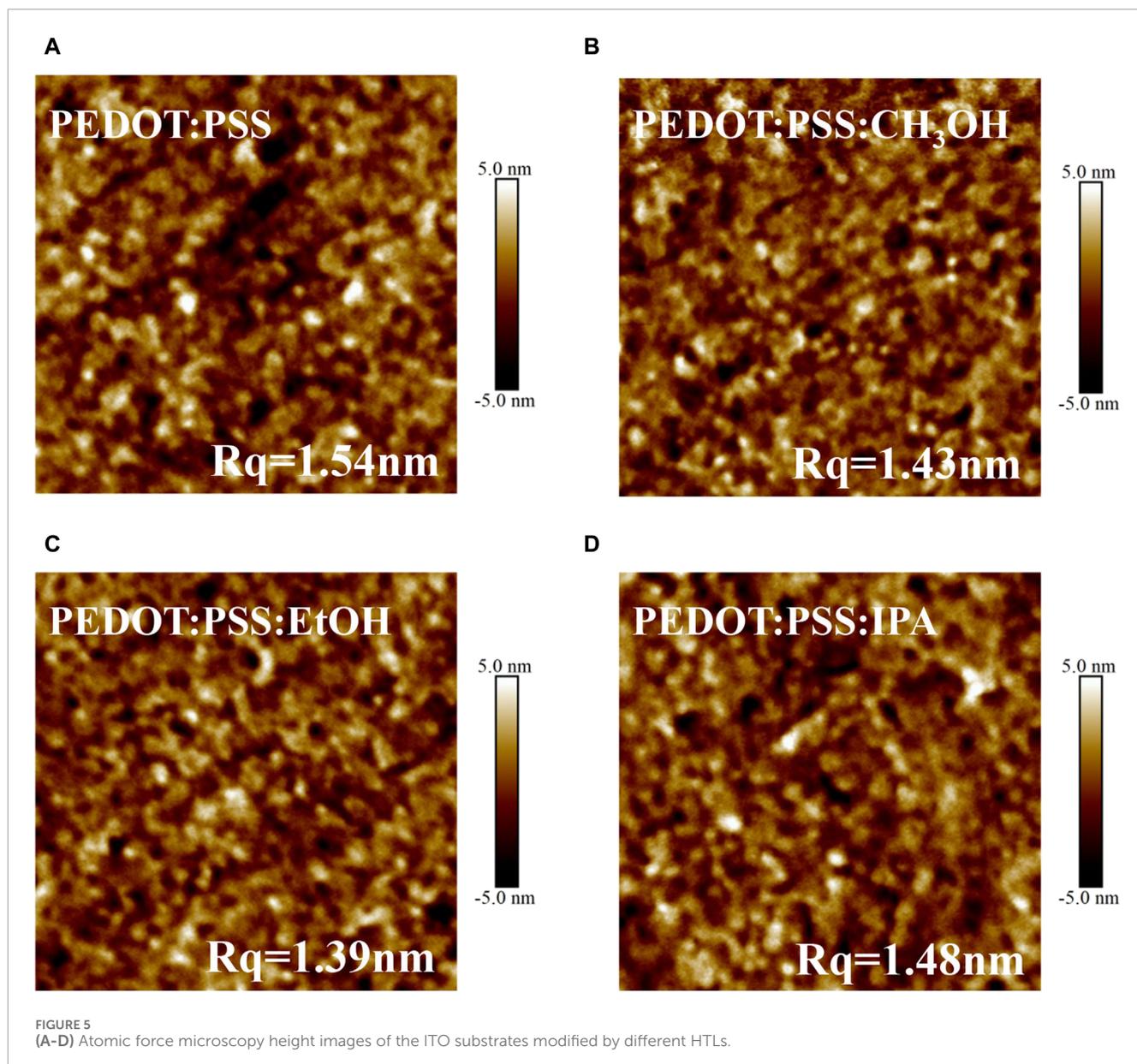
FDDLAS was used to obtain the vertical distribution information of the donors and acceptors in the OSCs to analyze



the effects of the HTL on the vertical gradient in the active layer (Bu et al., 2016; Wang et al., 2019). The FDDLAS data of various PM6:Y6 films with different HTLs are shown in [Supplementary Figure S4](#). Furthermore, the vertical distributions of PM6 and Y6 were extracted by fitting the FDDLAS data with the absorption values of the PM6 and Y6 films, as displayed in [Figures 3A–D](#). It is observed that the HTLs are able to adjust the composition distributions of the PM6:Y6 film in the top (0–30 nm) and bottom (90–100 nm) regions. Concretely, the proportions of PM6 in the active layers of the films near the HTL interfaces were 52.22% (PEDOT:PSS), 54.72% (PEDOT:PSS:CH₃OH), 66.89% (PEDOT:PSS:EtOH), and 51.38% (PEDOT:PSS:IPA). The PEDOT:PSS:EtOH/PM6:Y6 structure thus achieves the highest PM6 content near the HTL interface and an optimal Y6 content near the electron transport layer (ETL) interface, which is conducive for enhancing hole transfer and

reducing charge recombination at the interface between the active layer and electrode.

By combining the optical interferences of the different layers in the device, the exciton distribution profile was also extracted from the FDDLAS data based on the modified optical transfer matrix model ([Figures 4A–D](#)) (Bu et al., 2016; Gao et al., 2017). It can be seen that almost all the excitons generated by the PM6 donor (absorption at 500–620 nm) are distributed within the depth range of 0–50 nm for all sample films. However, this phenomenon is modestly different for the excitons produced by the Y6 acceptor in the blend film. For the PM6:Y6 films based on the PEDOT:PSS, PEDOT:PSS:CH₃OH, and PEDOT:PSS:IPA HTLs, significant numbers of the excitons generated by the Y6 acceptor (absorption at 620–900 nm) were observed on the surfaces of the blend films near the ETL regions (depth at 0–10 nm). This implies that the free holes generated after exciton separation must travel a



greater distance to reach the HTL interface, which could damage the efficiency of electron transport. When EtOH was added to PEDOT:PSS, the exciton-rich region near the surface generated by Y6 shifted from the top to the middle region in the blend film, indicating ameliorative transport of the free electrons generated by exciton separation.

To study the influences of the alcoholic solvents on the morphology of PEDOT:PSS in greater depth, the height images of the ITO/HTL films were tested by atomic force microscopy (AFM) using the tapping mode. As shown in Figure 5, the root mean-squared (R_q) values of the heights in these ITO/HTL films are 1.54 nm (PEDOT:PSS), 1.43 nm (PEDOT:PSS:CH₃OH), 1.39 nm (PEDOT:PSS:EtOH), and 1.48 nm (PEDOT:PSS:IPA). These results indicate that the use of alcoholic solvents can reduce the roughness of PEDOT:PSS and facilitate good interfacial contact with the active layer.

4 Conclusion

In this study, we propose a method of improving the photovoltaic performances of OSCs using diluted PEDOT:PSS with alcoholic solvents as the HTLs. The PCEs of the PM6:Y6 devices with alcohol-modified PEDOT:PSS as the HTLs are 17.94% (PEDOT:PSS:CH₃OH), 18.13% (PEDOT:PSS:EtOH), and 17.86% (PEDOT:PSS:IPA), which are higher than that of OSCs based on the traditional PEDOT:PSS (17.42%). The optimal photovoltaic performance of the device with the PEDOT:PSS:EtOH HTL was attributable to the smooth surface morphology and high PM6 content near the HTL interface, which could achieve a suitable carrier lifetime and fast charge extraction. This study shows that diluting PEDOT:PSS with alcoholic solvents is an effective method of deriving high-efficiency OSCs.

Data availability statement

The original contributions presented in the study are included in the article/[Supplementary Material](#), and any further inquiries may be directed to the corresponding authors.

Author contributions

TH: data curation, methodology, and writing—original draft. CG: data curation, formal analysis, and writing—review and editing. KS: data curation, methodology, and writing—original draft. LZ: investigation, software, and writing—original draft. LW: investigation, methodology, and writing—review and editing. WL: investigation, formal analysis, and writing—original draft. YM: conceptualization, data curation, funding acquisition, methodology, and writing—review and editing. YW: data curation, methodology, formal analysis, investigation, and writing—original draft. MQ: formal analysis, methodology, supervision, and writing—original draft. SL: formal analysis, funding acquisition, investigation, supervision, and writing—review and editing. GZ: data curation, funding acquisition, supervision, and writing—review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmats.2024.1383816/full#supplementary-material>

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