MINIMIZING INTERFACIAL ENERGY LOSS VIA POLYMER-ASSISTED D–A INTERFACE FOR INVERTED PEROVSKITE SOLAR CELLS WITH 25.78% EFFICIENCY

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ABSTRACT

Severe energy losses caused by carrier thermalization persist at heterojunction perovskite/PCBM interfaces, limiting further performance enhancement of inverted PSCs. Here, tetrakis(pentafluorophenyl)porphyrin (5FTPP) is introduced between 3D/2D perovskite heterojunction and PCBM. Compared to tetraphenylporphyrin without electron-withdrawing fluoro-substituents, 5FTPP can self-assemble with PCBM at interface into donor-acceptor (D-A) complex with stronger supramolecular interaction and lower energy transfer losses. This rapid energy transfer from donor (5FTPP) to acceptor (PCBM) within femtosecond scale is demonstrated to enlarge hot carrier extraction rates and ranges, reducing thermalization losses. Furthermore, the incorporation of polystyrene derivative (PD) reinforces D-A interaction by inhibiting self-π-π stacking of 5FTPP. Notably, the multidentate anchoring of PD-5FTPP mitigates the adverse effects of FA$^+$ volatilization during thermal stress. Ultimately, devices with PD-5FTPP achieve a power conversion efficiency of 25.78% (certified: 25.36%), maintaining over 90% of initial efficiency after 1000 hours of continuous illumination at the maximum power point (65 °C) under ISOS-L-2 protocol.

KEY WORDS

D-A supramolecular interface, thermalization loss, perovskite solar cells

1. INTRODUCTION

Minimizing energy loss caused by nonradiative recombination, carrier thermalization, and conduction band offset at perovskite/electron transport layer (ETL) interface of inverted PSCs remains a challenge. While the 3D/2D perovskite heterojunction passivation strategy is effective in combating defect-assisted surface nonradiative recombination, it falls short in mitigating interlayer recombination, thermalization losses, and conduction band offset losses. Notably, these existing interfacial methods focus on modulating perovskites while neglecting the crucial role of the ETL. Here, polystyrene derivatives (PD) and tetrakis(pentafluorophenyl) porphyrin (5FTPP) mixture is incorporated between 3D/2D perovskite heterojunction and PCBM (ETL) to establish a polymer-assisted D-A supramolecular self-assembly interface including 5FTPP as donor and PCBM as acceptor. This pioneering strategy is demonstrated to minimize interfacial energy loss, while mitigating FA$^+$ volatilization under thermal stress.

2. RESEARCH PROJECTS

2.1 D–A Supramolecular System

The supramolecular interaction between 5FTPP and PCBM was investigated through theoretical calculations based on density functional theory (DFT). The results (Fig.1a) suggest that 5FTPP, relying on C-F···π, π···π, and N-H···π supramolecular interactions, tends to form a more stable supramolecular complex with PCBM by self-assembly. Subsequently, we calculated the HOMO and LUMO energy levels of the supramolecular complex of 5FTPP and PCBM. HOMO was localized on 5FTPP component while the LUMO was positioned on PCBM component, confirming the donor characteristics of 5FTPP and the acceptor characteristics of PCBM (Fig.1b). When 5FTPP is introduced between PVK and PCBM, it tends to self-assemble into a more stable supramolecular D-A system with PCBM.

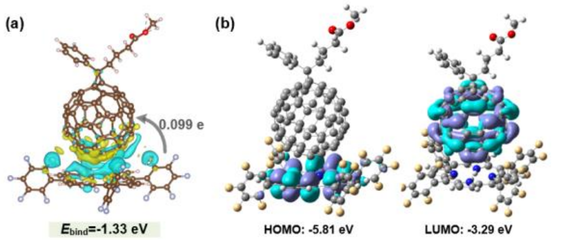


Fig.1: (a) Schematic diagram of the differential charge density (b) HOMO and LUMO energy levels of 5FTPP/PCBM systems based on theoretical calculation.

2.2 Reduce Thermalization Losses of Carriers

Compared with tetraphenylporphyrin without electron-withdrawing fluoro-substituents, 5FTPP can self-assemble with PCBM at the interface to form a donor-acceptor (D-A) complex with stronger supramolecular interactions including C-F···π, π···π, and N-H···π bonding and lower loss of the energy transfer from excited state (5FTPP*) to CS state (5FTPP$^{\cdot+}$-PCBM$^{\cdot-}$). This rapid energy transfer within the femtosecond scale from the donor (5FTPP) to the acceptor (PCBM) can shorten the extraction time of hot carriers by 12% and expand the temperature extraction range by 8%, thus effectively suppressing carriers thermalization loss.

2.3 Suppress 5FTPP π-π stacking with PD

To further reinforce this D-A effect, different polymers were screened to modulate the interaction between 5FTPP and PCBM. The results show that self-synthesized polystyrene derivative (PD) exhibits effective inhibition of self-π-π stacking of 5FTPP (Fig.2a-f), thereby mitigating the increase of height and surface potential roughness (Fig.2g-i) stemming from the excessive enrichment of 5FTPP itself, promoting the formation of a more uniform donor-acceptor interface between 5FTPP and PCBM, and enhancing electron transport.

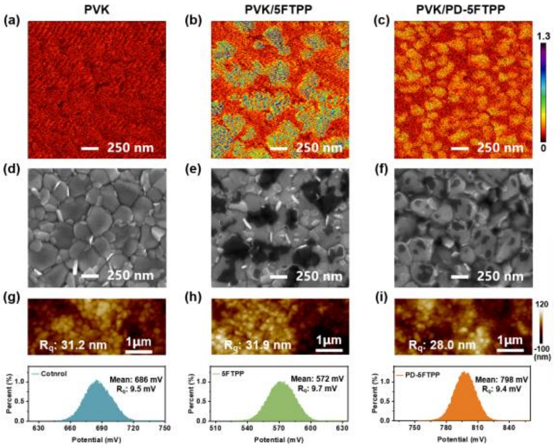


Fig.2: AFM-IR image at 992 cm^{-1} of (a) PVK, (b)PVK/5FTPP, and (c)PVK/PD-5FTPP films. SEM images of perovskite (d) without treatment, and treated by (e) 5FTPP and (f)PD-5FTPP. AFM height image and KPFM potential distribution diagram of perovskite (g) without treatment, and treated by (h) 5FTPP and (i) PD-5FTPP.

2.4 Multidentate anchoring inhabits FA^+ escaping

Synchronously multidentate anchoring (Fig.3) of PD, including the strong electron-donating C=O groups in PD interact with FA^+ and Pb through hydrogen bonds and coordination bonds, and with I ions through N-H⋯I- hydrogen bonds, suppresses the escaping of organic cation FA+ and formation of PbI2, thus enhancing the film's stability under thermal stress.

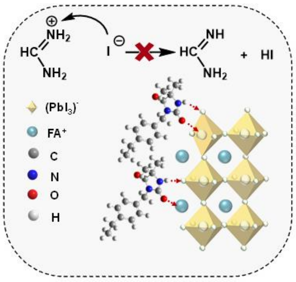
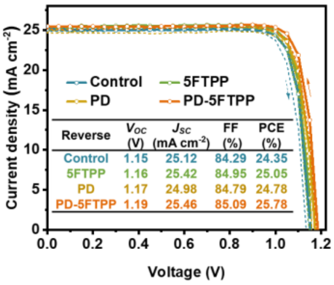


Fig.3: Multidentate anchoring diagram of PD.

2.5 Photovoltaics and stability

device with PD-5FTPP exhibits a PCE improvement from 24.35% to 25.78% (certificated: 25.36%) with a Voc of 1.19 V, retaining above 90% of the initial PCE after 1000 hours of continuous illumination at the maximum power point (65 °C) according to the ISOS-L-2 standard protocol.



Reverse	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
Control	1.15	25.12	84.29	24.35
5FTPP	1.16	25.42	84.95	25.05
PD	1.17	24.98	84.79	24.78
PD-5FTPP	1.19	25.46	85.09	25.78

Fig.4: *J-V* curves of champion PSCs.

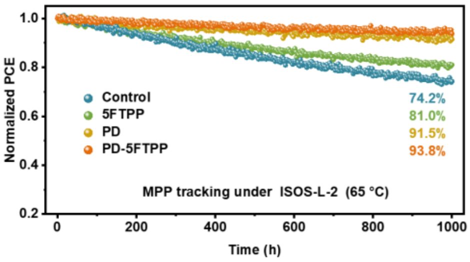


Fig.5: MPP tracking of the devices under SOS-L-2 standard protocol.

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