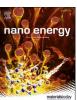


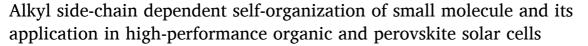
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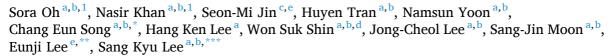
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ABSTRACT

The molecular self-organization of organic semiconductors, which is mainly determined by the structural design, film processing, and device configuration, is one of the crucial factors for achieving high-performance organic photovoltaics (OPVs) and perovskite solar cells (PvSCs). In this study, we newly synthesized and developed strongly self-organized small molecules via alkyl side-chain engineering. Replacing "H" to "C₆H₁₃" on the thienyl group, SM2 showed a well-ordered face-on orientation. Due to favorable self-organization leading to effective charge carrier dynamics, including enhanced charge transfer/transport and suppressed recombination, SM2based OPVs and PvSCs exhibited improved device performance compared to the devices based on SM1 without an additional hexyl side-chain. The best fullerene-based OPV and planar PvSC with SM2 as a smallmolecule donor and as a hole transport layer (HTL) achieved an unprecedentedly high efficiency of 9.38% and 20.56%, in contrast with SM1-based devices showing lower efficiency of 8,70% and 15,37%. Furthermore, the planar PvSCs based on undoped-SM2 HTL exhibited comparable efficiency but provided excellent heat and humidity stability compared with doped spiro-OMeTAD-based devices. These results clearly indicated that SM2 with highly-ordered and favorable self-organization is a promising organic semiconductor for future applications of high-performance organic and inorganic-organic hybrid electronics.

1. Introduction

Solution-processed organic photovoltaics (OPVs) and perovskite solar cells (PvSCs) are both promising candidates for emerging renewable energy sources owing to their flexibility, lightweight, good transparency, and color rendering from tunable absorption spectra and are easily manufactured devices [1-14]. In recent years, the development of organic semiconductors as a photoactive component for OPVs and as a charge transport material for PvSCs is rapidly accelerating to further improve the photovoltaic performance [15-20].

For photoactive materials in high-efficiency OPVs, not only broadened and efficient light absorption for increasing the short-circuit current density (JSC) but also energy level matching and morphology control of the photoactive layer for maximizing the open-circuit voltage

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