Interface Engineering with Co-Self-Assembled Monolayers for Improved Charge Extraction and Morphology in Organic Photovoltaics

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Abstract Carbazole-based self-assembled monolayers (SAMs), such as the widely used [2-(9H-carbazol-9-yl)ethyl]phosphonic acid (2PACz), have significantly improved the performance of conventional organic photovoltaics (OPVs) by serving as efficient hole-

selective layers (HSLs). However, the intrinsic limitations of 2PACz have hindered further enhancements in device performance. Although the molecular modification of 2PACz is a common strategy to overcome these constraints, it often introduces challenges. In this study, we present a coself-assembled monolayer (co-SAM) approach that blends 2PACz with 4PDACB to address the drawbacks associated with single-component SAMs. This co-SAM strategy effectively balances electrode work function tuning, interfacial quality improvement, and active layer blend morphology optimization, thereby enabling enhanced overall performance in OPVs. By optimizing PM6:Y6-based OPVs using the co-SAM strategy, we achieved an outstanding power conversion efficiency (PCE) of 17.55%, significantly exceeding the PCEs of devices with single-layer 2PACz (17.08%) and 4PDACB (16.13%). The effectiveness of this approach was further demonstrated in ternary OPVs, where co-SAMs enabled a maximum PCE of 18.69%. These results underscore the critical role of surface engineering in controlling film morphology and provide valuable insights for advancing OPV performance.

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