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DIO-Induced Interface Intermixing Limits in Pseudo-Bilayer PM6:Y6 Organic Solar Cell Blends

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Organic solar cells (OSCs) based on PM6:Y6 bulk heterojunction (BHJ) active layers have achieved power conversion efficiencies (PCEs) approaching 20%,[1] establishing them among the most promising non-fullerene acceptor systems. However, the inherent morphological disorder of single-step BHJ deposition introduces challenges in controlling donor–acceptor phase separation, promoting charge recombination and limiting long-term stability.[2] The two-step pseudo-bilayer heterojunction (PPHJ) architecture offers improved vertical phase separation control through sequential sublayer deposition,[3] yet PM6:Y6 PCEs typically plateau near 17%,[4] underscoring the need for a deeper understanding of interfacial intermixing and aggregation/phase separation dynamics.

In this work, we systematically investigate the role of the solvent additive 1,8-diiodooctane (DIO) on phase aggregation dynamics in PM6:Y6 PPHJ active layers cross four sublayer modification configurations: unmodified, acceptor-only, donor-only, and dual modification of both sublayers. Using UV-VIS absorption spectroscopy and time-resolved photoluminescence (TRPL), we track sublayer crystallinity, intermixing, and morphological evolution under thermal stress.

We demonstrate that unmodified PM6:Y6 PPHJ films are thermally unstable, exhibiting significant morphological evolution under thermal stress. Introducing DIO during deposition of a single sublayer partially mitigates this instability: the plasticising effect of DIO promotes sufficient initial intermixing, yielding a stable interface that is notably resistant to large-scale morphological disruption. However, this stabilising effect is highly sensitive to the modification strategy employed. Extending DIO treatment to both sublayers induces excessive phase separation, effectively collapsing the intermixed interface into a more discrete layer-by-layer morphology and disrupting the balance achieved through single-layer modification.

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