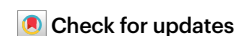


Organic photovoltaics surpass the 20% efficiency milestone

Francesco Furlan & Nicola Gasparini



Crystallization dynamics manipulation leads to vertically separated donor and acceptor phases in thick films, improving charge mobility and device efficiency.

Organic photovoltaics (OPVs) are a promising technology to produce green energy. Combining unique solution-processing properties, materials tunability and potentially low costs, OPVs represent an exciting solar technology for various applications, including building-integrated photovoltaics and agrivoltaics¹. OPVs have faced multiple phases since the first device discovery in 1986. For at least two decades, fullerene-based OPVs have dominated the field, with the 10% power conversion efficiency (PCE) barrier being difficult to overcome due to the poor absorption properties and reduced voltage of these devices. A breakthrough was the discovery of non-fullerene acceptors (NFAs) with complementary absorption, with the donor materials, tunable energetics and similar transport properties of fullerene. NFA-based OPVs, in particular the Y-family, allowed superior PCE and opened up applications, that is, semi-transparent devices, not achievable with fullerene-based OPVs and inorganic counterparts².

The state-of-the-art photoactive layer in OPVs is made of a donor and an NFA, forming an interpenetrating network (bulk heterojunction; BHJ)³. Separation of photogenerated charge carriers (excitons) requires the formation of a well-interconnected BHJ, and charge carrier transport towards the electrode requires phase-pure domains close to the bottom and top contacts. However, the vertical crystallization of a BHJ is not homogeneous and low crystallinity regions form far from the substrate driving the crystallization process. Increasing the thickness of the BHJ exacerbates this phenomenon, leading to solar cell performance losses. One of the strategies to achieve vertical phase separation is sequential layer-by-layer deposition of donor and acceptor materials to form a pseudo bilayer. However, the region forming a BHJ is limited in this case, leading to poor exciton separation⁴.

Now, writing in *Nature Materials*, Yaowen Li, Yongfang Li and colleagues report a strategy to improve the crystallization of BHJ, resulting in devices surpassing the 20% PCE barrier⁵. They designed and synthesized an organic semiconductor regulator, AT-b2O, to fine-tune the BHJ nanomorphology, consisting of the donor polymer D18-Cl and the acceptor N3. Owing to hydrogen bonding interactions between the hydrophilic glycolate side chains of AT-b2O and N3, the authors can precisely regulate the components' thin-film crystallization. In fact, the ternary D18:4Cl:AT-b2O:N3 blends form a BHJ gradient vertical phase separation consisting of a donor-rich (D18-4Cl) phase close to the substrate, a uniform BHJ region in the middle and an acceptor-rich phase (N3) with enhanced crystallinity near the top of the active layer (Fig. 1). This configuration combines the advantages of both BHJ and layer-by-layer deposition, where the former maximizes exciton

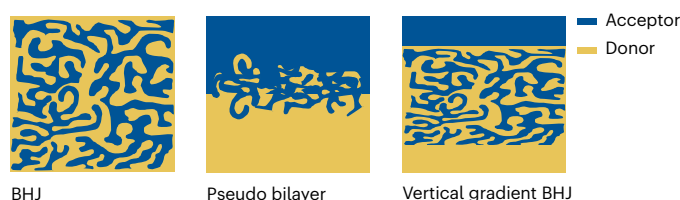


Fig. 1 | Photoactive layer configurations in OPVs. Schematic of BHJ, pseudo bilayer and vertical gradient BHJ configuration in organic solar cells. Figure adapted from ref. 8.

splitting, while the latter allows the formation of pure donor (and acceptor) phases to enhance charge transport. When introduced in the ternary blend, AT-b2O promotes higher crystallinity and a longer-range order of N3, with long diffusion lengths approaching 50 nm, facilitating exciton separation and higher electron mobilities in the vertical direction. With this in mind, D18-4Cl:AT-b2O:N3-based OPVs delivered a certified PCE of 20.43% (20.82% achieved in the laboratories) with high BHJ thickness tolerance (~18% PCE at 400 nm BHJ thickness). An anti-reflective coating was employed to reduce reflection losses and 2PCAz as a hole-transport layer to reduce parasitic absorption compared with PEDOT:PSS.

The effect of AT-b2O on N3 crystallinity is independent of the donor material employed in the BHJ, the thickness of the layer or the type of underlying electrode. Cross-sectional scanning electron microscopy and energy dispersive spectroscopy mapping confirms how the binary blend of D18-4Cl:N3 has a homogeneous distribution of donor and acceptor components throughout the whole blend, while D18-4Cl:AT-b2O:N3 shows F-rich domains (corresponding to N3) close to the top electrode and Cl-rich domains close to the bottom electrode (corresponding to D18-Cl). This is valid for both thin (100 nm) and thick (400 nm) BHJs and supported by the ion spatial mapping via time-of-flight secondary ion mass spectrometry measurements. Halogenated and non-halogenated solvents and different processing methods were tested, demonstrating the universality of this approach, as the efficiency of all devices with AT-b2O had very low thickness-dependency for all conditions.

The capability of fabricating OPVs with a thick BHJ (>100 nm) is critical for fabricating large-area devices and modules, as industry-scale deposition methods struggle to deposit thin photoactive layers with high film continuity. However, large-area devices delivered lower efficiency compared with small-area devices^{6,7}. Li and co-workers demonstrated that highly homogeneous thick layers (250 nm) of D18-4Cl:AT-b2O:N3 can be deposited via blade-coating with pinhole-free morphology, fabricating mini modules (area of 15 cm²) with a record certified efficiency of 18.04% (18.36% obtained in the laboratory). The stability of the devices with AT-b2O was also enhanced, maintaining 83% of their initial PCE for over 1,200 hours.

This research features many ingredients to move OPVs closer to commercialization. In fact, surpassing the 20% efficiency target on small-area devices and 18% on mini modules puts this technology a step closer to its inorganic counterparts. Future research efforts should target industrial requirements to close the lab-to-fab gap of OPV technology. This includes green solvent formulation, scalable donor and acceptor materials, and printable (and flexible) electrodes, aiming for stable devices under real-world operation.

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Competing interests

The authors declare no competing interests.