



Bulk heterojunction organic photovoltaics from water-processable nanomaterials and their facile fabrication approaches



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ABSTRACT

Organic thin film photovoltaics based on bulk-heterojunction donor–acceptor combinations have received significant interest due to their potential for low-cost, large-scale solution processing. However, current state-of-the-art cells utilise materials soluble mainly in halogenated solvents which pose processing challenges due to their toxicity and thus environmental hazards. In this contribution, we look at various nanomaterials, and alternative processing of these solar cells using environmentally friendly solvents, and review recently reported different strategies and approaches that are making inroads in this field. Specifically, we focus on the use of water-dispersible donors and acceptors, use of aqueous solvents for fabrication and discuss the merits of the two main approaches of water-processable solar cells; namely, through the use of water-soluble materials and the use of aqueous dispersion rather than a solution, as well as review some of the recent advances in alternative fabrication techniques.

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1. Introduction

Organic solar cells (OSCs) have significant potential to supplement the world's drive for renewable energy; however, current OSCs still cannot compete with traditional, more mature systems such as silicon solar

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cells. The continual development of other technologies means that in the near future, bulk solar power generation will still be dominated by the existing technologies. However, one of the major advantages OSCs have over other solar cell technologies is their flexibility and their potential for niche applications. This advantage arises from properties such as mechanical flexibility, transparency and processing flexibility where OSC can be fabricated on arbitrary shapes or flexible substrates. In this regard, OSCs have a real potential to add functionality to many devices and be integrated into new applications. The possibility of fabricating OSCs by low-cost, high-throughput methods is highly attractive for new applications such as flexible electronics and building-integrated solar cells.

The ease of fabrication of OSCs comes from the use of solution processing, which enables the use of bubble jet printing (another type of inkjet printing, where inside each nozzle there is a tiny heating element, rather than a piezo crystal as in conventional inkjet printing. In this printer, when an electric impulse reaches the heating element, it vaporizes a tiny amount of ink), die-slot coating, screen printing and other existing fabrication methods. Current solution processing of OSCs however relies on halogenated solvents for processing, which are toxic and pose an environmental hazard. For large-scale processing, this would ideally be replaced with aqueous or more environmentally friendly solvents. To date, the main challenge in the aqueous fabrication of OSCs has been the active layer. While water-soluble or water-processable materials are widely used for the hole and electron transport layers, the active layer of OSCs based on poly(3-hexylthiophene) (P3HT) and phenyl-C61-butyric acid methyl ester (PCBM)–P3HT:PCBM is generally fabricated using halogenated solvents such as dichlorobenzene, and the use of non-halogenated solvents has generally led to poorer efficiencies. For environmental reasons and cost-effectiveness, it is desirable to also replace such solvents with water. In this case, the challenge lies in how to achieve the optimum morphology, as a change to aqueous solvents would have a significant effect on OSC morphology and therefore its efficiency. Also, using facile solution processing or emulsion, large-scale, low-cost production is possible for rapid manufacturing process with quicker energy payback time.

1.1. Parameters affecting solar cell morphology and efficiency

The key performance characteristic of OSCs is their efficiency, which depends on both material and processing factors. The efficiency of a solar cell depends on the open circuit voltage (V_{oc}), the short circuit current (I_{sc}), the fill factor (FF) and the incident light intensity (P_{in}) according to Eq. (1)

$$\eta = \frac{V_{oc} \times I_{sc} \times FF}{P_{in}} \quad (1)$$

And thus, in order to maximise efficiency, one needs to maximise the V_{oc} , I_{sc} and FF. The V_{oc} is theoretically limited by the energy difference of the donor–acceptor HOMO–LUMO levels [1,2]; however, it also depends on the processing factors such as the material contacts, which may lead to resistive losses or current leakage. Similarly, the I_{sc} is highly affected by the absorption spectrum of the material, but is also dependent on the right morphology to facilitate charge separation and transport. The FF is affected by all the processes involved, and thus manipulation of the film morphology greatly affects it, which in turn affects the achievable efficiency.

Kirchartz et al. [3] calculated the radiative efficiency limits of organic bulk heterojunction (BHJ) solar cells, and analysed the losses leading to the large difference between theoretical and real efficiencies (of 20% and 4.2%, respectively). They found that the losses can be attributed to (i) optical losses, (ii) exciton losses due to insufficient transport or inefficient exciton dissociation, (iii) non-radiative recombination losses and (iv) charge carrier collection losses due to insufficient mobility. Amongst these losses, the most dominant one is the non-radiative

recombination at the donor–acceptor interface, which poses a challenge to solar cell material design. In BHJ solar cells it is desirable that all the generated excitons are dissociated at the donor–acceptor interface, and minimising this recombination loss would go a long way towards increasing cell efficiency.

In terms of morphology, the ideal BHJ solar cell would contain an active layer that possesses a large area of interface between the donor and the acceptor material as well as a continuous percolation pathway for both electron and hole transport [4]. These phases should also be similar to the exciton diffusion length in order to minimise losses through recombination. However, although it is believed that this active layer is composed of interconnected, phase-separated morphologies, some recent studies have shown that both crystalline and amorphous domains of donor, acceptor and donor–acceptor mixtures play a part. Yin and Dadmun [5] found through small-angle neutron scattering (SANS) study that the optimal model structure of the active layer contains crystals of donor and acceptor materials in a matrix of miscible, amorphous donor–acceptor mixture. This model postulates that the charges form within the amorphous donor–acceptor mixture as they contain a large amount of donor–acceptor interfaces, and these charges then diffuse towards the crystalline donor or acceptor domains for transport to the electrodes. This hypothesis implies that a certain degree of miscibility is required between the donor and acceptor material; but interconnected, crystalline, phase separated domains are still required for effective charge transport.

Adding to the complexity is the fact that such a morphology depends on many parameters, such as the donor–acceptor combination, solvent, substrate, fabrication techniques and conditions, additives, post-processing and other external influences that may occur during the fabrication process [6]. For example, it has been well established in P3HT:PCBM-based systems fabricated from organic solvents that post-fabrication annealing can improve OSC efficiency. This is because as-fabricated P3HT:PCBM blends from solvents such as dichlorobenzene show no obvious phase separation. A simple thermal treatment can increase the crystallinity of P3HT and drive the phase separation in the active layer, as well as improving material composition and polymer chain orientation near the cathode. Similarly, this phase separation can also be induced by solvent vapour annealing. By swelling the active layer with solvent vapour, the diffusive motions of both P3HT and PCBM are enhanced, leading to P3HT crystallisation and PCBM phase aggregation. It was observed that a poor solvent yields better results because it enhances PCBM segregation; causing them to migrate to the surface. All these factors have been extensively studied for OSCs cast from organic solvents; however, aqueous fabrication shares some common challenges in its optimization, as well as unique challenges due to the difference in structure or morphology for aqueous processed OSCs.

1.2. Characterisation of the active layer morphology in organic solar cells

In addition to the material selection and design, the analysis and characterisation of the morphology of the active layer across a wide range of length scales poses its own unique challenge, and many techniques have been used to provide complementary information on parameters such as polymer orientation, donor–acceptor interface, phase separation and domain sizes. Several in-depth reviews already exist on this subject [2,4,7–10] and, as such, it will not be discussed in depth in this review; however, we would like to provide an overview of some of these techniques as, in terms of active layer morphology, the performance of the solar cell depends on parameters which span several length scales from the molecular level to the mesoscopic level.

The main techniques used in active layer characterisation are those that are able to provide structural information on the nanoscale. Various X-ray, electron or neutron-based techniques have been used to study properties from polymer crystal structures. Table 1 summarises some of the techniques reported in the literature for the characterisation of

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