



Engineering vertical morphology with nanoparticulate organic photovoltaic devices



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ABSTRACT

Sequential deposition of monolayers, composed of nanoparticles with varied donor-acceptor concentration ratios, has allowed the fabrication of organic photovoltaic (OPV) active layers with engineered vertical morphology. The performance of polymer-polymer poly(9,9-dioctylfluorene-co-bis-*N,N*-phenyl-1,4-phenylenediamine):poly[(9,9-di-*n*-octylfluorenyl-2,7-diyl)-alt-(benzo[2,1,3]thiadiazol-4,8-diyl)] (PFB:F8BT) and polymer-fullerene poly(3-hexylthiophene):phenyl C61 butyric acid methyl ester (P3HT:PCBM) nanoparticulate (NP), graded nanoparticulate (GNP) and bulk heterojunction (BHJ) OPV devices have been studied. For both material systems the highest device V_{OC} is observed for the graded structure. Furthermore, thermal treatments can be used to alleviate parasitic series resistance in the GNP devices, thus improving device J_{SC} and efficiency. Overall, this work shows that the nanoparticle approach provides a new experimental lever for morphology control in OPV devices.

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1. Introduction

The fabrication of an optimal donor-acceptor OPV device sets a fascinating challenge for the researcher. Fundamentally contradictory requirements for active layer morphology must be balanced to allow for efficient charge separation between the materials, whilst still maintaining charge transport pathways which minimise bimolecular recombination [1]. Exacerbating this task is the limited number of experimental levers available to the researcher to influence active layer morphology [2]. In general, the experimental parameters which can be utilised to optimise device morphology and performance are limited to temperature, solvent and material choice (Fig. 1). Consequently, researchers have attempted, with varying degrees of success, methods to affect active layer morphology such as: thermal and solvent annealing [3,4], the addition of additives [5] and the application of new materials such as block-co-polymers [6]. Indeed, one of the main reasons for the continued interest in the ubiquitous P3HT:PCBM material system [7], despite extensive research and development into alternative

materials, is that significant optimisation of the active layer morphology can be achieved by simple fine-tuning of the temperature and solvent parameters [8].

Whilst active layer nanomorphology and material domain size have been recognised as key factors in optimising device performance for some time [4], it is only relatively recently that vertical stratification and phase segregation in devices has been considered and examined [9]. Previous work has shown that vertical phase segregation is an important optimisation parameter in OPV fabrication [17]. In particular, having incorrect phases at the electrode interfaces reduces device performance through increased recombination, whereas when the concentration gradient is aligned correctly improved performance is observed [18]. As such, given the carrier selectivity of the electrodes, having vertically graded donor and acceptor concentrations in OPV active layers is potentially an elegant way both to reduce recombination and increase device charge collection [10]. Consequently, recent years have seen a steady increase in the number of publications reporting attempts to induce and characterise graded active layers in OPV devices using the aforementioned experimental levers [11–16]. However, the methodologies available to systematically control vertical phase segregation are limited.

The use of graded heterostructures is well established in small-

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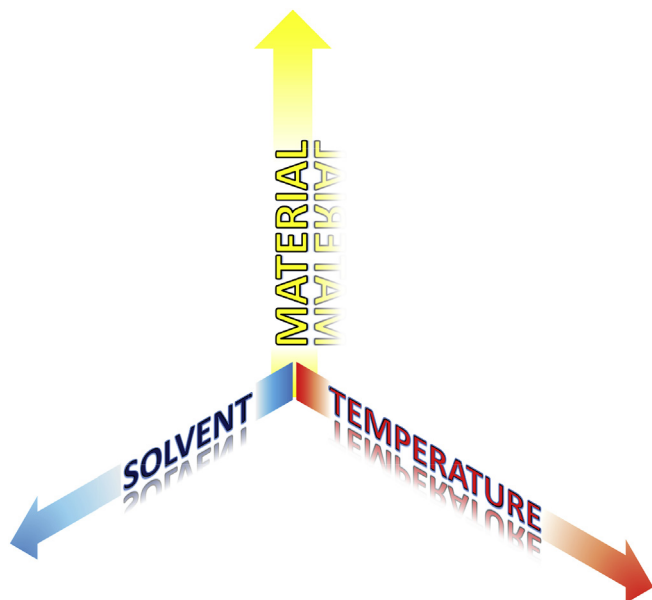


Fig. 1. The parameter space available for active layer morphology optimisation in OPV.

molecule OPV systems fabricated via vapour phase deposition where the composition of each layer can be readily controlled [19]. Enhanced concentration gradients are known to lead to both increased Voc and Jsc, which can be attributed to reduced recombination (associated with reduced charge build-up within the device) and an increased charge collection efficiency due to the composition gradient driving charges to the appropriate electrodes, respectively [20–22]. However, to date, proven examples of graded heterostructures in all-solution processed devices have been rare due to the limited experimental levers available for controlling layer composition [23]. Consequently, the fabrication approaches have been limited to manipulating the surface energy of the component polymers/molecules relative to the electrode interfaces via thermal treatment [24,25], direct manipulation of surface energy [16], electric fields [26] or the use of additives [27].

The application of nanoparticulate materials to OPV active layers provides a unique opportunity to control directly the vertical morphology at length scales appropriate for efficient charge separation within the active layer. We have previously shown that, by thermally modifying the nanoparticulate active layer surface, sequential deposition of nanoparticle monolayers can be used to systematically increase active layer thickness [28]. In this report, we demonstrate fabrication of vertically graded active layers of P8BT:PFB polymer:polymer and P3HT:PCBM polymer:fullerene blends via the sequential deposition of nanoparticle monolayers. Moreover, we show that engineering these vertical donor-acceptor concentration gradients across the device provides a level of control of device parameters hitherto only achieved in vapour deposited OPVs.

2. Experimental

2.1. Materials

All of the water required for experiments was treated via a reverse osmosis system. When ultra clean water was required, the reverse osmosis water was further treated with a Milli-Q[®] plus system in which the conductivity of the water was kept below $10 \mu\text{S cm}^{-1}$. All of the chloroform used was distilled, in house, prior to use. Pre-patterned indium tin oxide (ITO) substrates were

purchased from Kintec Company, Hong Kong. Sodium dodecyl sulphate (SDS) used during the miniemulsion process was purchased from BDH Lab and Sigma-Aldrich at a purity of 90–99% and used as received. The aqueous suspension of PEDOT:PSS used during this investigation was obtained from H.C Stark, U.S and used as received. The semiconducting polymers used were obtained from a range of suppliers; F8BT and PFB were obtained from American Dye Source, Canada, with molecular weights of 15,000–200,000 g/mol for F8BT and 15,000–75,000 g/mol for PFB quoted by the manufacturer and as measured by GPC against polystyrene standards. P3HT and PCBM were supplied by Reike, Canada and Solenne B.V, Netherlands, respectively.

2.2. Nanoparticle synthesis

Fabrication of the aqueous polymer dispersions of all material weight ratios required were performed as described previously. The semiconducting materials (a total of 30 mg) were initially dissolved in chloroform (0.8 g) and introduced to an aqueous SDS solution (52 mM in 2.8 mL). The mixture was stirred at 1200 RPM for 1 h to form a macroemulsion. The emulsion was then sonicated using a Branson 450 analogue sonifier operating at 60% of maximum amplitude for 2 min. After sonication, the miniemulsion samples were heated at 60°C and stirred at 1200 RPM for a minimum of 3 h in order to evaporate the chloroform from the suspension. Upon evaporation of the chloroform an aqueous suspension of polymer nanoparticles remained. The resulting particle suspensions were then dialysed to remove excess surfactant until the surface tension of the filtrate reached $38 \pm 2 \text{ mN/m}$.

2.3. Nanoparticle characterisation

A NanoSeries ZetaSizer Nano-ZS (Malvern Instruments, UK) equipped with a helium-neon laser source (wavelength 633 nm; power 4.0 mW) was used for measuring the hydrodynamic diameter of the semiconducting polymer colloids produced. Each sample

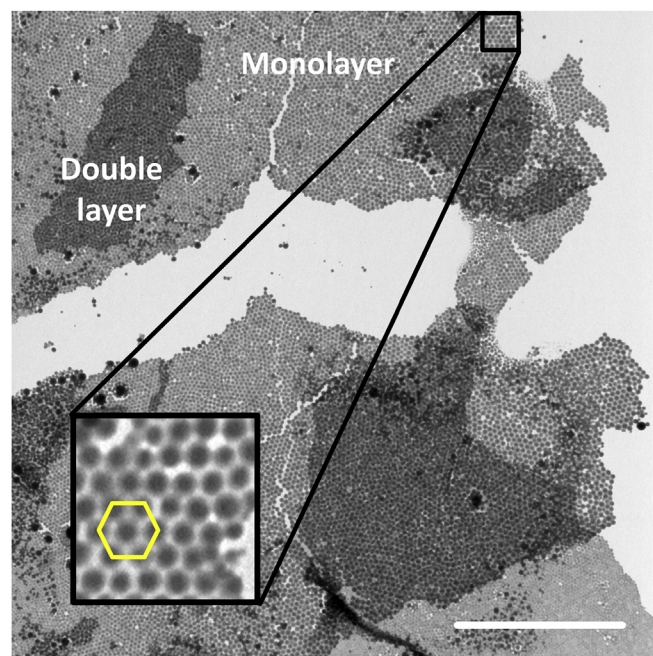


Fig. 2. Scanning electron microscopy image of P3HT:PCBM 1:1 nanoparticulate single and double layer structures. The inset shows a magnified image of the monolayer region emphasizing the hexagonal close-packing of the particles.

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