

# Enhanced performance of layer-evolved bulk-heterojunction solar cells with Ag nanoparticles by sequential deposition



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## ABSTRACT

Organic photovoltaic devices based on layer-evolved bulk-heterojunction (LE-BHJ) from poly{[4,4'-bis(2-ethylhexyl) dithieno(3,2-b:2',3'-d)silole]-2,6-diyl-alt-[4,7-bis(2-thienyl)-2,1,3-benzothiadiazole]-5,5'-diyl} (Si-PCPDTBT)/[6,6]-phenyl C<sub>71</sub> butyric acid methyl ester (PC<sub>71</sub>BM) are successfully fabricated by a sequential solution deposition process. During the process of a top layer being deposited from an appropriate cosolvent ratio control, the top layer of PC<sub>71</sub>BM vertically penetrates a pre-coated layer of Si-PCPDTBT, resulting in an inter-diffused nanostructure with an LE-BHJ surface morphology. The LE-BHJ cells exhibit improved nanoscale phase separation and comparable performance, in comparison with the conventional BHJ device. Furthermore, Ag nanoparticles (NPs) with optimized ratio embedded LE-BHJ exhibit significant enhanced efficiency in comparison with the conventional BHJ and pristine LE-BHJ devices, because the multiple scattering from the Ag NPs leads to longer optical paths, which give rise to effective absorptivity and reduced leakage current within the BHJ active film.

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## 1. Introduction

Organic photovoltaic (OPV) devices based on an active layer composed of a mixture of donor and acceptor materials have been broadly researched, because of meaningful advantages, such as the potential of large-area solar devices, flexibility in a variety of future industries, and roll-to-roll fabrication by a solution process [1–10]. In order to improve the performance of the OPV device, various fabrications and researches have been carried out, such as the thermal and solvent annealing processes [11–14], insertion of interlayers for efficient transport [15–18], and layer-evolved heterojunction (LE-BHJ) for spontaneous interdiffusion [19–21].

In 1987, C. W. Tang reported bilayer structured solar cells with the power conversion efficiency (PCE) of ~1%, which have a small interfacial contact area between the donor and the acceptor [22], and therefore give rise to reduced photo-generated charge carriers, and yield a poor short-circuit current ( $J_{sc}$ ), compared to the bulk heterojunction (BHJ) solar cells with nanoscale phase-separation, with efficient percolated structure [1–2]. According to several recent reports, promising bilayer structured solar cells have been fabricated by using inter-diffusion of acceptor materials of the top layer into the donor material based bottom layer, via different

solvent or thermal annealing [19–21]. However, while the bilayer structured solar cells demonstrate a better nanoscale morphology, and solve PCBM top layer aggregation by a sequential deposition process, the power conversion efficiency (PCE) (%) of the organic solar cells exhibits only similar or comparable efficiency, compared to the conventional BHJ and LE-BHJ.

One of the important approaches to increase the device PCE, that of embedding of metal nanoparticles in the active layer, is an efficient and widely used fabrication procedure in (OPV) devices. Metallic silver (Ag) nanoparticles (NPs) are effective in organic active thin film due to their scattering performance, which lead to increase of the light absorption within an active layer [23–32]. Thus, the insertion of Ag NPs with optimized ratio into an active layer of the devices can significantly enhance the absorption, and increase the photo-generated charge carriers.

In this study, we fabricated layer-evolved BHJ (LE-BHJ) using poly{[4,4'-bis(2-ethylhexyl) dithieno(3,2-b:2',3'-d)silole]-2,6-diyl-alt-[4,7-bis(2-thienyl)-2,1,3-benzothiadiazole]-5,5'-diyl} (Si-PCPDTBT) and [6,6]-phenyl C<sub>71</sub> butyric acid methyl-ester (PC<sub>71</sub>BM), by sequential spin-coating of Ag NPs embedded PC<sub>71</sub>BM top layer dissolved in the optimized cosolvent (0–30% volume ratio of DCB in DCM). The LE-BHJ solar cells were fabricated on top of the pre-deposited Si-PCPDTBT layer, by inter-diffusion of Ag NPs embedded (30 nm, 2 wt%) in PC<sub>71</sub>BM dissolved in a cosolvent, from a volume ratio of 0% to 30% dichlorobenzene (DCB) in dichloromethane (DCM). The newly designed metal NPs

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embedded LE-BHJ solar cells by solution process exhibit improved optical absorption, effective morphology, and reduced leakage current, and thus contribute to the increased PCE (%).

## 2. Experimental details

### 2.1. Device fabrication

Donor–acceptor organic solar cells based on Si-PCPDTBT/PC<sub>71</sub>BM bilayer-mixture, and layer-evolved bulk heterojunction (LE-BHJ) were fabricated on ITO/glass substrate. The substrate had to be washed by detergent, and then an ultra-sonication process followed in water, acetone, and isopropyl alcohol, to clean the ITO surface. The ITO was exposed to the UV for 15 min, to reform the surface. Then, the efficient hole transporting material PEDOT:PSS (Clevis PH) was spin-coated on top of the ITO (thickness of ~35 nm). The substrate was dried at 140 °C for 10 min in air, and then transferred to a glove box, to spin-coat active materials. The Si-PCPDTBT solution with a concentration of 10 mg/ml in DCB was spin-coated on the PEDOT:PSS layer at 2000 rpm for 40 s, and the PC<sub>71</sub>BM solution of 20 mg/ml without (bilayer mixture) and with cosolvent (up to 30% DCB in DCM: LE-BHJ) was directly spin-coated on the Si-PCPDTBT bottom layer at 2000 rpm for 20 s. Furthermore, the Ag NPs with various concentrations (1–5 wt%) are inserted to the LE-BHJ, and coated same condition of 2000 rpm for 20 s. The total thickness of the active layer was 150 nm, as confirmed by the SEM cross-sectional image of Fig. S1. All the active layers were dried at 120 °C for 10 min in glove box. For the reference cell of Si-PCPDTBT/PC<sub>71</sub>BM BHJ solar cells, a mixture solution of Si-PCPDTBT (10 mg): PC<sub>71</sub>BM (20 mg) in 1 mL of DCB was spin-coated onto PEDOT:PSS at 2000 rpm for 40 s with a thickness of ~150 nm, and then the active layer was annealed at 120 °C for 10 min in glove box. After that, metal cathode (Al) was deposited with a thickness of ~100 nm under a pressure of  $4.0 \times 10^{-6}$  Torr, using a thermal evaporator.

### 2.2. Characterization

The performances of the organic solar cells were measured by solar simulator, with Air Mass 1.5 Global (AM 1.5 G) at an intensity

of 100 mW/cm<sup>2</sup>; and the current density–voltage characteristics of the solar cells was measured by Keithley 2400 source meter. The total cell area was confirmed 15.2 mm<sup>2</sup> and an aperture with an area of 9.84 mm<sup>2</sup> was used on top of the cell, to correct the measurement. The surface morphology and roughness of the BHJ, bilayer mixture, and LE-BHJ were observed by atomic force microscopy (AFM) (Veeco, USA; D3100). IPCE spectra (PV measurement) for the solar cells was conducted, to prove the short-circuit current of  $J_{SC}$  related to the  $J$ – $V$  curves.

## 3. Results and discussion

Fig. 1(a) shows a schematic diagram of the device structures of a bilayer mixture (without cosolvent), pristine LE-BHJ, and LE-BHJ with Ag NPs solar cells, respectively, with a concentration graded interpenetration zone, using a sequential solution deposition process, with the help of an optimized cosolvent ratio. Fig. 1(b) and the inset of Fig. 1(a) also show the molecular structures of Si-PCPDTBT and PC<sub>71</sub>BM, and a band diagram of the component materials. Detailed fabrication of devices will be described in the following experimental section. We fabricated conventional Si-PCPDTBT:PC<sub>71</sub>BM BHJ solar cells, for comparison with the pristine LE-BHJ and LE-BHJ with Ag NPs devices.

Fig. 2(a) and Table 1 show that the pristine LE-BHJ devices with an optimized cosolvent ratio of 10% (which is composed of 10% DCB in 90% DCM (v/v)) achieve a device performance = 3.9%, with  $V_{OC} = 0.58$  V,  $J_{SC} = 12.46$  mA/cm<sup>2</sup>, and FF = 53, respectively, which is comparable with the efficiency of the reference device with BHJ of 4.2%. The pristine LE-BHJ solar cells fabricated from a cosolvent ratio of more than 30 wt% exhibit poorer device performance, owing to lower photo-current density, and reduced FF. Furthermore, the efficiency of the solar cells fabricated by spin-coating a PC<sub>71</sub>BM top layer dissolved in 100% DCM has the significantly reduced performance of 2.7%, because of only partial inter-diffusion, and unfavorable nanomorphology, with micron sized PCBM aggregated regions [21].

To increase and optimize the device performance based on LE-BHJ, we adapted Ag NPs with various concentrations

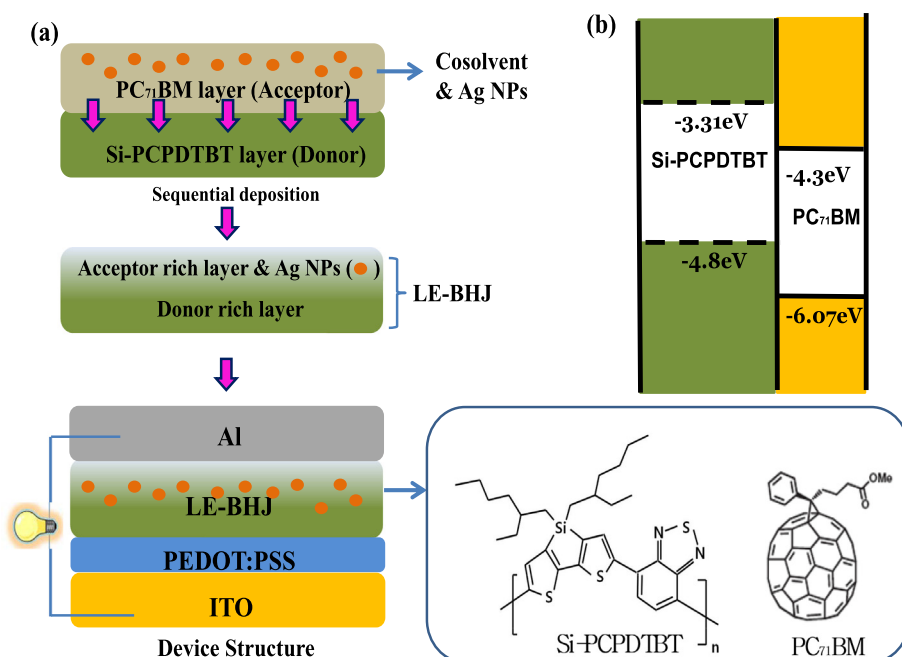


Fig. 1. (a) The molecular structure of Si-PCPDTBT and PC<sub>71</sub>BM, and proposed device sequential structures of bilayer mixture and LE-BHJ solar cells fabricated by interpenetration of the PC<sub>71</sub>BM top layer (dissolved in cosolvent and with Ag NPs) to the Si-PCPDTBT bottom layer. (b) Energy level diagram of the component materials.

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