Supporting Information

Spontaneously spreading film process to improve photovoltaic performance of organic solar cells with PHJ structure

Yetai Cheng,[a] Xiaodong Wang,[a] Zengliang Pei,[c] Ya-nan Chen,[a] Hao Lu,*,[b] Yahui Liu,*,[a] and Zhishan Bo*,[ac]

[a]College of Textiles & Clothing, State Key Laboratory of Bio-fibers and Eco-textiles, Qingdao University, Qingdao 266071, China. E-mail: liuyh@qdu.edu.cn, zsbo@bnu.edu.cn

[b]College of Materials Science and Engineering, Qingdao University, Qingdao, 266071 P. R. China. E-mail: luhao@qdu.edu.cn

[c]Beijing Key Laboratory of Energy Conversion and Storage Materials, College of Chemistry, Beijing Normal University, Beijing 100875, China.

1. Experimental section

1.1 Materials

ITO glass substrate is purchased from Shenzhen South China Xiangcheng Technology Co., Ltd. PDINN is purchased from Ruixun Optoelectronic Materials Technology Co., Ltd. Polymer PTB7-Th is purchased from 1-Material Reagent Company. IEICO-4F is purchased from Solarmer Materials Company. Chlorobenzene and methanol solvents are purchased from Sigma Aldrich Company. The chloroform reagent is purchased from Beijing Dongfang Shibo Fine Chemical Co., Ltd. All reagents are directly used without further purification.

1.2 Fabrication of OSCs

The ITO glass substrate (square resistance of 15 Ω/sq) is cleaned with deionized water, anhydrous ethanol, and isopropanol, dried with nitrogen gas, and then treated in a UV ozone activator for 35 minutes. The PEDOT: PSS solution is spin-coated at 4000 rpm for 30 s and annealed at 150 °C for 15 min to obtain an anodic interface layer of 10 nm thickness. Then, the active layers of different structures and processes are prepared.

1.2.1 Preparation of the active layer of BHJ structure by SC process

Dissolve PTB7-Th:IEICO-4F (1:1, w/w) in a chlorobenzene solvent with a

polymer concentration of 10 mg/mL, and use 1,8-diiodooctane (4%, v/v) as a solvent additive to prepare an about 150 nm active layer film by spin coating.

1.2.2 Preparation of the active layer of BHJ structure by SS process

Dissolve PTB7-Th:IEICO-4F (1:1, w/w) in a chlorobenzene solvent with a polymer concentration of 10 mg/mL, and use 1,8-diiodooctane (4%, v/v) as a solvent additive. 25 μL of the mixed solution is dropped onto the surface of room temperature water and transferred to the substrate after the solution spontaneously diffuses to form a film.

1.2.3 Preparation of the active layer of PHJ structure by SS process

Dissolve PTB7-Th:IEICO-4F (1.5:1 for the first layer and 1:1.5 for the second layer, w/w) in a chlorobenzene solvent with a polymer concentration of 7 mg/mL, and add 1,8-diiodooctane (4%, v/v) as a solvent additive. 25 μL of the mixed solution is dropped onto the surface of room temperature water and transferred to the substrate after the solution spontaneously diffuses to form a film.

All active layer films were annealed at 100 ℃ for 1 minute. PDINN (1 mg/mL, soluble in methanol) is spin-coated at 3000 rpm for 20 seconds to obtain a cathode interface layer. All treatments are performed in a nitrogen glove box. Finally, the 100 nm Ag electrode is vapor deposited under vacuum pressure at 3×10^{-6} Torr. An OSC area of 0.04 cm^2 is obtained.

1.3 Characterization of OSCs

The UV-visible absorption spectra are tested using a UV-vis spectrometer (PerkinElmer, Lambda 750). The morphology of the film is obtained by atomic force microscopy (Multimode Nanoscope IIIA) in tapping mode and transmission electron microscopy (HRTEM Talos F200X FEI) with a test voltage of 200kV.

After calibrating the light intensity using standard monocrystalline silicon photovoltaic cells, the external quantum efficiency was tested in the air using the solar cell spectral response measurement system (QER3011, Guangyan Technology Co., Ltd.).

1.4 Preparation of films for TEM test

The films with different active layers are prepared on a substrate with a

PEDOT:PSS layer and the substrate is floated on water to dissolve the water-soluble PEDOT:PSS layer, thus preserving the initial morphology of the active layer on the ITO surface

1.5 Characterization of the hole/electron-only devices

The mobility of holes and electrons is calculated, via the Mott-Gurney equation: $J = 9\varepsilon\varepsilon_0\mu/(8L^3)V^2$.

where J is the current density, ε is the dielectric constant of the film, ε_0 is the dielectric constant of the vacuum, μ is the zero-field mobility, and L is the thickness of the film. $V = V_{appl} - V_{bi}$, $V_{appl,}$ and V_{bi} are the voltage applied to the device and the built-in voltage.

2. Supporting Table

Table S1. Photovoltaic parameters of BHJ devices constructed by SC and SS processes

Process	Atmosphere	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
SC	N_2	0.717	26.40	57.55	10.91
SS	Air	0.687	27.09	58.24	10.84