

Large-area, Flexible Polymer Solar Cell Based on Silver Nanowires as Transparent Electrode by Roll-to-Roll Printing*

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Abstract Conventional organic solar cell's (OSC) architectures, including rigid transparent substrate (Glass), conductive electrode (Indium tin oxide, ITO) and small working areas, are widely utilized in organic photovoltaic fields. However, such a structure as well as conventional spin-coating method obviously restrict their industrial application. In this article, we report the deposition of silver nanowires (AgNWs) on the flexible substrate by slot-die printing. The obtained AgNWs films exhibited a high transmittance and a low resistance, and were further used as the transparent conductive electrode of OSCs. A typical conjugated polymer, poly[(2,5-bis(2-hexyldecyloxy)phenylene)-*alt*-(5,6-difluoro-4,7-di(thiophen-2-yl)benzo[c][1,2,5]thiadiazole)] (PPDT2FBT), was used as the active material to fabricate large-area (7 cm²) solar cells by a slot-die coating process. The power conversion efficiency (PCE) could reach 1.87% initially and further increased to 3.04% by thermal annealing. Compared to the performance of reference cell on ITO substrate, the result indicated that the AgNWs could be developed as an alternative substitute of conductive electrode to fabricate the large-area flexible OSCs by roll-to-roll printing.

Keywords Polymer solar cell; Silver nanowires; Slot-die; Large area; Flexible substrate

INTRODUCTION

Organic solar cells (OSCs) have attracted much attention in the past decade for the multiple advantages, such as low-cost, environment-friendly, renewable resources and so on^[1–12]. Indium tin oxide (ITO) has been used widely as the material of transparent electrode due to its good conductivity and transmittance. However, the application of ITO in the preparation of large area devices remains various challenges, for instance, the high price and increased resistance with the working area^[13, 14]. In addition, ITO electrode deposited on polyethylene terephthalate (PET) substrates is not the best choice because of its brittle feature to cause fissures for the flexible OSC device upon bending^[15]. Therefore, there is an increasing interest for replacing the traditional ITO-based conductive materials by the low-cost transparent electrode. Recently, silver nanowires (AgNWs) have been vigorously investigated as the transparent electrode owing to its high transmittance and conductivity to fabricate the OSC devices^[16–22]. Moreover, the film fabrication process is also simple for AgNWs on the flexible substrates, such as slot-die coating, spray coating, bar coating and spin coating^[16, 23–25].

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Although a variety of requirements for the application of organic photovoltaics (OPVs) has been proposed in recent years, a large working area produced by a facile method is essential for the industrial application^[1, 26, 27]. Nowadays, the efficiency of bulk heterojunction (BHJ) OSCs has reached up to 9%–10% for single-junction cells^[28–30] and 10%–11% for ternary or tandem cells^[31–34] in the small-scale devices (< 1 cm²) on a rigid substrate. Spin-coating is the most traditional technique in laboratory, which cannot be scaled up for a larger area and in the mass-production^[2, 35, 36]. Slot-die printing is regarded as the most promising method for a fast preparation of OSCs. By this way, various layers of the OSCs can be printed by roll-to-roll (R2R) manner on a flexible substrate. Thus it can greatly enhance the possibility for practical application of OSCs in the future.

Herein, we report the large area AgNWs film produced successfully by slot-die printing with the crossed multilayer structures on PET substrate. The conducting flexible film showed a good transmittance and a high conductivity as compared with ITO electrode. The silver nanowires were deposited with different diameters (*i.e.*, 30, 50 and 70 nm), and the influences of the nanowire diameters and coating times on the conductivity and transmittance were investigated. AgNWs are the promising alternatives of ITO as a flexible transparent electrode. A typical conjugated polymer poly[(2,5-bis(2-hexyldecyloxy)phenylene)-*alt*-(5,6-difluoro-4,7-di(thiophen-2-yl)benzo[c][1,2,5]-thiadiazole)] (PPDT2FBT) was used as the active material to manufacture large-area flexible OSCs by slot-die coating, which showed a good photovoltaic performance compared with the reference solar cell with PET-ITO as the transparent electrode. The power conversion efficiency (PCE) of the flexible OSCs increased from 1.87% to 3.04% after thermal annealing (TA), and the change of active film morphologies was further investigated by atomic force microscopy (AFM). This study reports a useful strategy to prepare the large area OSC by R2R printing techniques, and will be helpful for their industrial application in the future.

EXPERIMENTAL

Materials

Silver nanowires initial solution (6 mg/mL) was purchased from Hyper Chemical Co., Ltd., China, and 5 mL was extracted into the tube before use. The conjugated polymer poly[(2,5-bis(2-hexyldecyloxy)phenylene)-*alt*-(5,6-difluoro-4,7-di(thiophen-2-yl)benzo[c][1,2,5]-thiadiazole)] (PPDT2FBT) was purchased from 1-Material Inc. as a donor. [6,6]-Phenyl-C71 butyric acid methylester (PC₇₁BM), was purchased from the American Dye Source, Inc. as an acceptor. The donor and acceptor blends (1:1.5, *W:W*), were dissolved in *o*-dichlorobenzene (*o*-DCB) at a concentration of 8 mg/mL, and 2% (*V:V*) diphenyl ether was added as the additive solvent before slot-die coating.

Slot-die Coating Process

A slot-die coating instrument (Extrusion Deposition System, nTact) was used to fabricate the film of AgNWs. The size of PET substrate was 200 mm × 200 mm. The solution of 0.5 mg/mL AgNWs was loaded into the equipment. After optimization of the parameters, the dispense rate of the solution was 200 mL/h, the coating velocity was 85 m/h, and the gap distance between the coating header and the substrate was 150 μm. In addition, the coating direction was changed by every 5 times to form the cross-network structure of AgNWs.

The R2R instrument (Mini Roll Coater, FOM Technologies) was used to fabricate the large-area flexible polymer solar cell, the ZnO sol-gel solution was synthesized successfully and dissolved in isopropanol following the reported protocol^[38].

Measurements

Atomic force microscopy (AFM) was measured by Dimension 3100 to investigate the film morphology of active layer. Scanning electron microscopy (SEM) was measured by Hitachi S4800. UV-VIS-NIR spectrophotometer (UV-3600) and 4-point probes resistivity measurement system (RTS-9) were used to measure transmittance and resistivity of AgNWs, respectively.

The *J-V* characteristics of devices were assessed under AM1.5G (100 mW/cm) with a Newport Thermal Oriel 91159A solar simulator. Light intensity was calibrated with a Newport oriel PN 91150V Si-based solar cell. *J-V* characteristics were recorded with a Keithley 2400 source meter unit.

RESULTS AND DISCUSSION

Washing and Diluting the AgNWs

Silver nanowires initial solution (6 mg/mL, diameter of silver nanowire was 50 nm, Hyper Chemical Co., Ltd.) was prepared and dispersed in 5 mL of ethanol (Fig. 1a). Then acetone was added into the solution with quickly shaking (Fig. 1b) to wash out poly(vinyl pyrrolidone) (PVP), which prevented the silver nanowires from gathering into cluster. After removing all the solvents, 60 mL of ethanol was added into the test tube to disperse the nanowires at a concentration of 0.5 mg/mL. Cleaning and washing the nanowires according to the above-mentioned methods for 2 or 3 times could get the pure AgNWs with few nanoparticles. The SEM images clearly showed the differences of AgNWs before and after washing. There were quite a few nanoparticles around the nanowires before washing, and almost no particles were observed after cleaning (Figs. 1c and 1d).

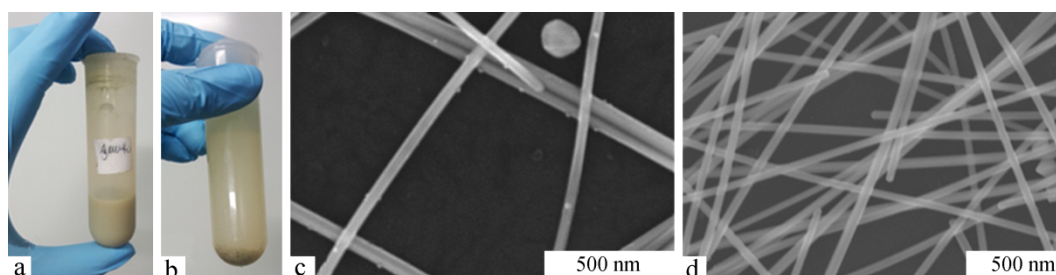


Fig. 1 Photographs of (a) the original solution of AgNWs dispersed in ethanol with concentration of 6 mg/mL and (b) after adding acetone into the original solution to wash the nanowires; SEM images of the AgNWs (c) without acetone-washing and (d) with acetone-washing on Si wafer

Coating AgNWs on Flexible Substrate

A slot-die coating instrument (Extrusion Deposition System, nTact) was used to fabricate the film of AgNWs. The size of PET substrate was 200 mm × 200 mm. The solution of 0.5 mg/mL AgNWs was loaded into the equipment. After optimization of the parameters, the dispense rate of the solution was 200 mL/h, the coating velocity was 85 m/h, and the gap distance between the coating header and the substrate was 150 μm. In addition, the coating direction was changed by every 5 times to form the cross-network structure of AgNWs (Fig. 2).

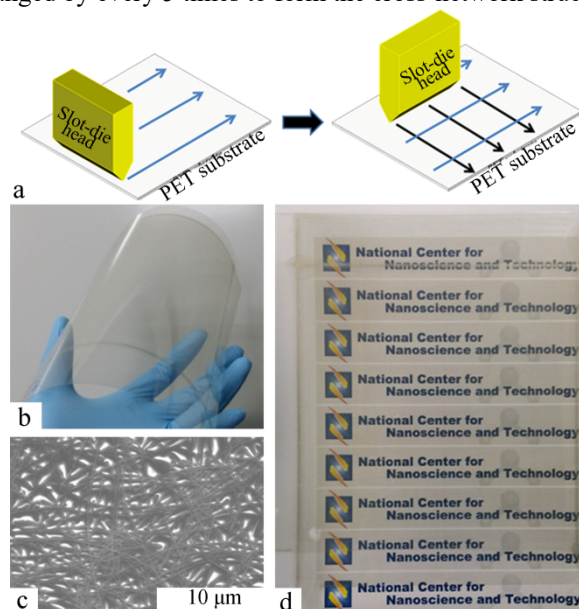


Fig. 2 (a) The schematic diagram of depositing AgNWs on PET substrate by slot-die coating; (b) AgNWs coated on the flexible substrate after slot-die coating for 30 times; (c) SEM of AgNWs coated on PET substrate; (d) the transmittance of PET-AgNWs

From the SEM (Fig. 2c) images, it could be seen that AgNWs were fabricated with the compact interpenetrating network structure after being coated on the PET substrate. The mesh of nanowires could provide high conductivity for using as the transparent electrodes^[37]. The transmittance of AgNWs achieved from 94.9% to 70.8% in the visible wavelength by regulating the slot-die coating times, and the resistance was changed from 73.9 $\Omega/\square$ to 6.4 $\Omega/\square$ as well (Fig. 3a). Overall, a high conductivity and a high transparency were gained by the way of multiple cycles of coating, which resulted from the AgNWs junctions and their mesh. The conductivity and transmittance can be well controlled by the times of slot-die coating^[18]. On the other hand, different diameters of AgNWs were also deposited as the transparent bottom electrodes. Diameters of 30 nm and 70 nm had no obvious influence on the properties of the films (Figs. 3c–3f), while the diameter of 50 nm showed a better balance in regard of transmittance and conductivity (Figs. 3a and 3b). Therefore, we select AgNW-based transparent films with 50 nm in size as a substrate for further investigation.

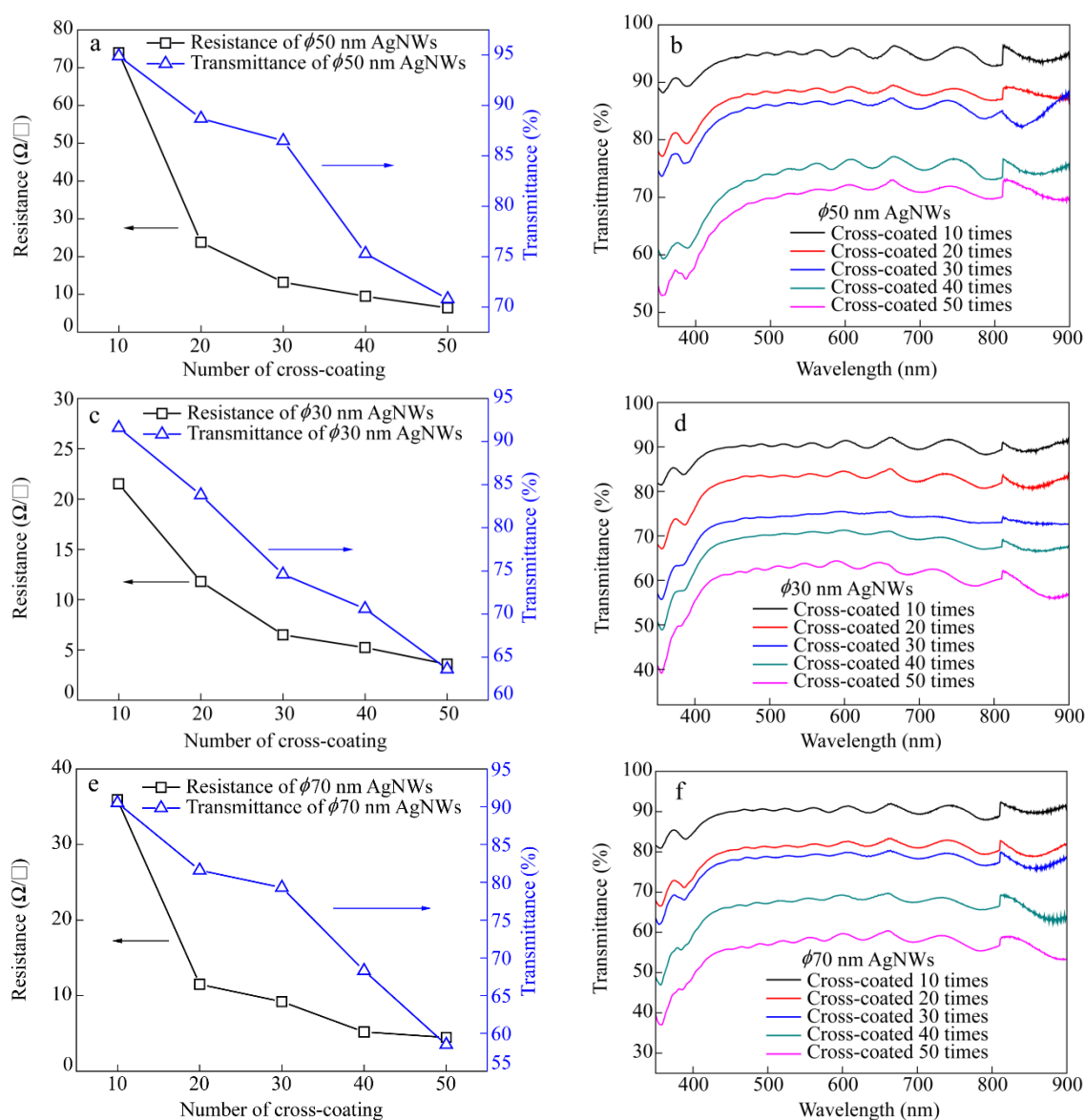


Fig. 3 The diagram of corresponding relations between transmittance (T) and resistance (R) by changing the diameter of AgNWs and the number of cross-coating: (a), (b) 50 nm; (c), (d) 30 nm; (e), (f) 70 nm (The online version is colorful.)

Fabrication of Large-area Flexible Polymer Solar Cell

The highly efficient OSCs with a small cell area (0.04 cm^2) on a rigid substrate based on PPDT2FBT have been reported by Nguyen *et al.*^[29]. The PCE could reach 9.21% with a conventional device architecture of glass-ITO/PEDOT:PSS/polymer:PC₇₁BM/Al (PEDOT, poly(3,4-ethylenedioxythiophene); PSS, polystyrene sulfonic acid; PC₇₁BM, [6,6]-phenyl-C₇₁ butyric acid methylester). In this study, large-area (7 cm^2) flexible OSCs were made through slot-die coating (Fig. 4a). The molecular structure of the polymer is shown in Fig. 4(b). The large-area solar cell had an architecture of PET-AgNWs/ZnO/polymer:PC₇₁BM/MoO_x/Ag, and PET-ITO/ZnO/polymer:PC₇₁BM/MoO_x/Ag structure was used as a reference cell.

In Fig. 4(a), the concentration of ZnO solution was 15 mg/mL in isopropanol^[38]. The donor and acceptor blends (1:1.5, *W:W*), were dissolved in *o*-dichlorobenzene (*o*-DCB) at a concentration of 8 mg/mL, and 2% (*V:V*) diphenyl ether was added as the additive solvent. During solar cell preparation, the dispense rate of ZnO solution was 5.46 mL/h, the coating speed was 60 m/h, and the thickness of ZnO layer was 70 nm. The active layer was dispensed at 2.92 mL/h and coated at 40 m/h (Fig. 4c); the thickness of active layer could reach 377 nm. Hence, after all the solution process with R2R fabrication at room temperature under ambient conditions, the sample was transferred to vacuum evaporation system to deposit the 10 nm MoO_x and 200 nm Ag. The active area of all the flexible polymer solar cells was 7 cm^2 (Fig. 4d). The *J-V* curves and performance data are shown in Fig. 4(e) and Table 1, respectively. The PCEs of the flexible polymer solar cell were 1.87% on the PET-AgNWs substrate and 2.17% on the PET-ITO substrate as the bottom electrode, respectively. Comparing to the photovoltaic performance of small area solar cell, the open circuit voltage (V_{oc}), the short circuit current (J_{sc}) and fill factor (FF) were decreased to 0.70 V, 9.81 mA/cm^2 and 31.4%. Obviously, the performance decreased due to an increase of resistance when the active area of cell was scaling up^[13, 39].

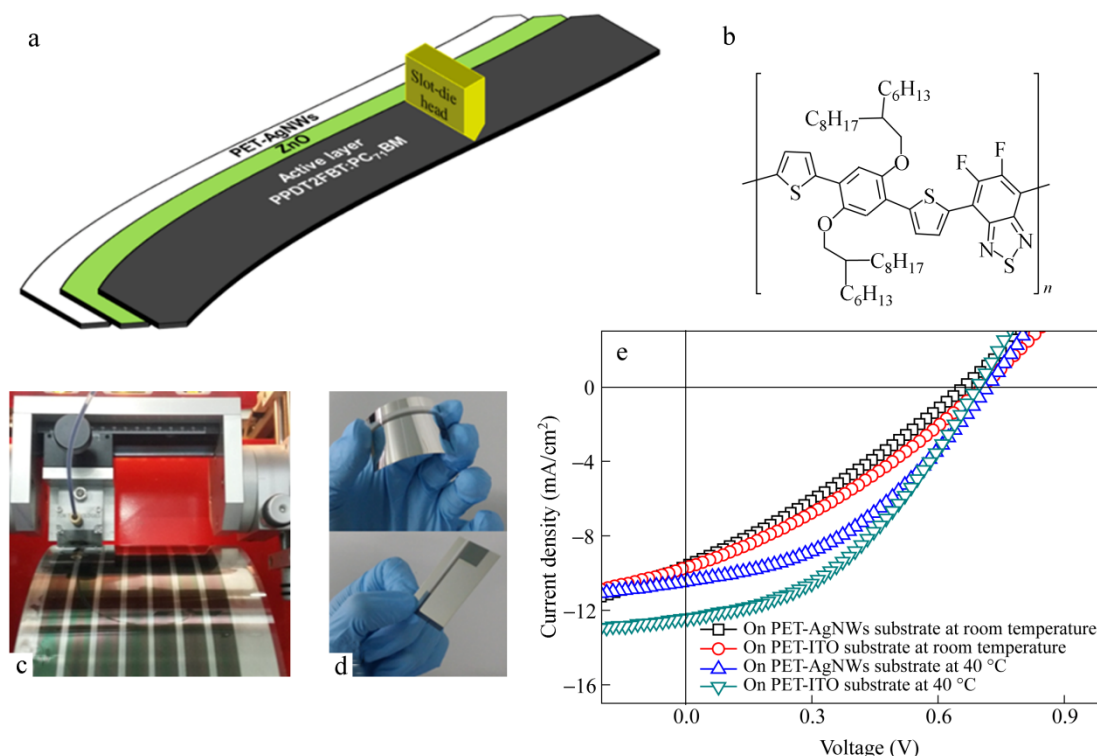


Fig. 4 (a) The architecture of large-area device with solution process; (b) The molecular formula of PPDT2FBT; (c) The fabrication of active layer in R2R process; (d) The flexible polymer solar cell with large-area (7 cm^2); (e) *J-V* curves of the performing polymer/PC₇₁BM-based flexible, large-area PSCs

Table 1 Photovoltaic performance of the flexible PSCs with different bottom electrodes under illumination of AM1.5G (100 mW/cm²)

V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)	Bottom electrode
0.66	9.69	29.3	1.87	PET-AgNWs ^b
0.70	9.81	31.4	2.17	PET-ITO ^a
0.71	10.45	40.6	3.04	PET-AgNWs (40 °C) ^c
0.69	12.44	41.2	3.54	PET-ITO (40 °C) ^c

All the active area of flexible PSCs was 7 cm².

^a Reference solar cell on the PET-ITO without thermal annealing; ^b Solar cell on the PET-AgNWs without thermal annealing; ^c Flexible solar cell on the two kinds of substrates with thermal annealing

To optimize the device performance, solution fabrication of R2R was also operated under the condition of thermal annealing during the deposition of active layer in the ambient atmosphere. The active layer was slot-die coated on the flexible substrate at 40 °C. Obviously, the performance of PSCs was improved after thermal annealing. FF was increased from 20.3% to 40.6% on PET-AgNWs electrode and such tendency was observed on the reference solar cell. Moreover, the same phenomenon was observed on J_{sc} , thus leading to a significant improvement of PCE.

Under different thermal annealing conditions during fabrication, different morphologies of the films were observed as shown in Fig. 5. It revealed distinct differences with temperature in the fabrication processing. Figures 5(a) and 5(b) show the height images of polymer/PC₇₁BM blend films, and the corresponding phase images are shown in Figs. 5(c) and 5(d). The root mean square (RMS) roughness decreased from 5.05 nm to 2.02 nm after thermal annealing. From the phase image, obvious aggregation was arisen without TA after the R2R fabrication of active layer. It caused large phase separation that it was not beneficial for the charge transport. Comparing with images of Fig. 5(a), the film morphology was changed a lot when the active layer was heated at 40 °C during the fabrication of solution processes. The film not only was uniform, but also displayed suitable phase separation. Therefore, it suggested that the thermal annealing treatment could provide better photovoltaic performance of large area flexible solar cell than that fabricated under the room temperature.

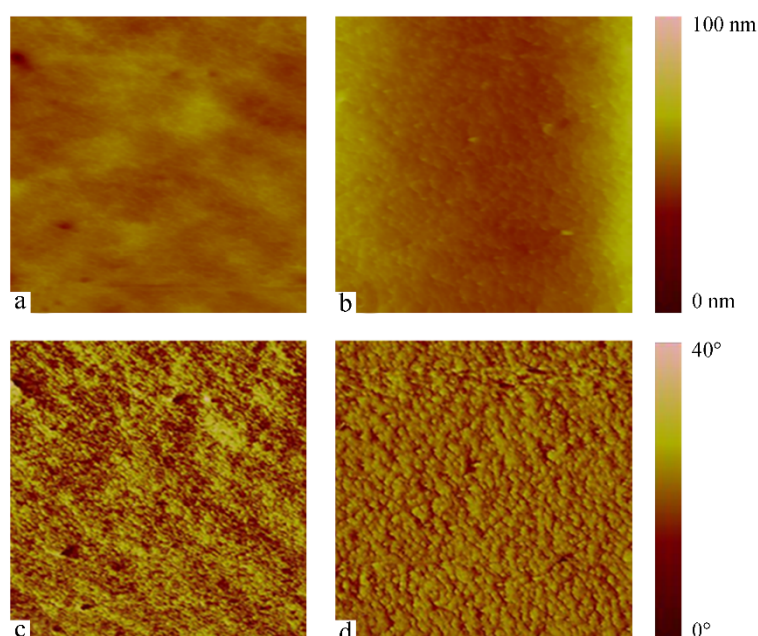


Fig. 5 AFM height (a, b) and phase (c, d) images: (a, c) polymer/PC₇₁BM blend film without thermal annealing, (b, d) polymer/PC₇₁BM blend film with thermal annealing (All the images were 1 μm × 1 μm.)

CONCLUSIONS

In summary, the AgNWs were deposited on flexible substrate as the transparent conductive bottom electrode by slot-die coating. Various AgNWs with different diameters (30, 50 and 70 nm) were coated on the PET substrate, and they exhibited good electrical conductivity and transmittance. Moreover, large area (7 cm²) flexible OSCs based on the conventional conductive substrate (PET-ITO) and the alternative conductive substrate (PET-AgNWs) were produced successfully through the slot-die printing process, respectively. The post treatment like thermal annealing was taken to optimize the performance of solar cell. The OSCs based on AgNWs substrate showed a similar performance as that the ITO-based substrate. Therefore, the AgNWs could be used as an alternative substitute for bottom electrode to fabricate the large-area flexible polymer solar cell.

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