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Contributed Paper

Simple ITO Surface Treatments Induced Better Performance for Low Cost Organic Solar Cells

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ABSTRACT

The objective of this research is to study effects of different indium tin oxide (ITO) surface treatments on the performances of organic solar cells. The organic solar cells (ITO/PEDOT:PSS/PCDTBT:PC70BM/TiO_x/Al) was fabricated using a cost-effective method called Rapid Deposition Convective (RDC)technique, whichuses small amount of solution to create uniform thin film on a substrate. The film thickness can easily be controlled by deposition speed and volume. Two surface treatment methods, UV radiation method and oxygen plasma method, were compared. The efficiency of solar cells under UV radiation treatment and oxygen plasma treatment are 6.30 and 7.24%, respectively, while the efficiency of untreated surface is 4.93%. AFM topography images also revealed that the surface prepared by oxygen plasma is smoother than the surface prepared by UV radiation. Both surface treatment methods not only effectively increase the work function of ITO substrate but also can improve the conductivity and the hydrophilic properties for the compatible of ITO and PEDOT:PSS layer.

Keywords: surface treatment, rapid deposition convective technique (RDC), organic solar cells, oxygen plasma, UV radiation

1. INTRODUCTION

Organic solar cells (OSCs) are being established owing to its benefits such as light weight, flexible substrates, low-cost ability and large scale fabrication process [1-7]. Nevertheless, for conventional organic solar cells, the cathode with a low work function is easily oxidized after the exposure in the air [8], which blocks the wide application of OSCs. Additionally, the indium tin oxide (ITO) as the anode is widely used as a transparent electrode in optical devices. ITO can be corroded by hole transport layer (HTL), like poly (3,4ethylenedioxylenethiophene): poly(styrene sulfonic acid) (PEDOT:PSS) which shortens the lifetime of the device [9-11]. PEDOT:PSS has been generally used as a HTL layer because of its effective electron blocking, solution process ability, and hole transportation qualities [12-13]. Despite such benefits, it is known that PEDOT:PSS corrodes ITO due to its extremely acidic (pH 1) aqueous suspension, and can additionally bring in water to the photoactive layer, leading to the deterioration of the device performance [14-16]. Two different approaches have been suggested to replace PEDOT:PSS. These include the incorporation of transition metal oxides with high work functions (MoO_3, V_2O_5, WO_3, and NiO) and surface treatments of ITO [17-22]. Especially, the work function of ITO is sensitive to surface treatment. The surface treatment of ITO is particularly interesting because it is very simple and cost-effective, and does not require any additional materials. ITO surfaces treated by various methods, such as mechanical polishing, O_2 plasma treatment, ultraviolet-ozone (UV-ozone) treatment, has been widely studied in recent years. Among such surface treatments, O_2 plasma and UV-ozone treatment are known as the most effective techniques to increase the work function of ITO. It was suggested that the change of In, Sn, O, surface carbon and the formation of a type of oxygen species explain to increase in ITO work function. Raising the ITO work function is useful as it reduces the interfacial barrier and incresing the hole injection efficiency from ITO to organic layer, hence improving organic solar cells operation. [23-26].

Nowaday for organic solar cells have various surfaces coating technique to deposit layer materials. The potential technological applications of micro- and nanoparticles coatings require the development of rapid, inexpensive, and easily controlled deposition procedures. Processes have been developed to facilitate particle deposition including spin coating [27], epitaxy [28-29], optical tweezers [30], electrophoretic assembly [31], and convective deposition [32-34]. Rapid Convective Deposition (RCD) offers the greatest promise in well-ordered, rapid, and controllable deposition on industrially relevant scales [35]. The mechanism of self-assembly of colloidal particle suspensions in thin evaporating films has been identified as “convective assembly” [36-37]. Continuous convective assembly is a prime candidate for achieving a desired surface morphology due its scalability. RCD is the alternative technique of thin film coating for organic solar cells for the improvement of fast, cheap, and readily controlled deposition procedures.

In this work, we studied the effect of surface treatment techniques, namely oxygen plasma and UV radiation techniques, with varied time treatment from 0-120 s and 0-30 min, respectively on the morphology and the performance of organic solar cells. In active layer comprised of a 1:4 (w/w) conjugated copolymer of Poly[N-9′-heptadecanyl-2,7-carbazole-alt-5,5-(4′,7′-di-2-thienyl-2′,1′,3′-benzothiadiazole)] (PCDTBT), as electron donor, and [6,6]-phenyl C_{70}-butyric acid methyl ester (PCBM), as electron acceptor. Examination on the sheet resistance, contact angle measurement, thickness of thin films, crystal structure, surface morphologies of ITO films and the performance of organic solar cells with Rapid Convective Deposition technique (RCD) were carried out.

2. MATERIALS AND METHODS

2.1 Materials

In active layer materials were comprise of Poly[N-9′-heptadecanyl-2, 7-carbazole-alt-5,5-(4′,7′-di-2-thienyl-2′,1′,3′-benzothiadiazole)] (PCDTBT) and PCBM ([6,6]-Phenyl C_{70} butyric acid methyl ester) were purchased from Sigma-Aldrich at 1:4 ratio by weight. For PEDOT:PSS hole transport layer from Clevios PH 1000 commercial available aqueous. The ITO glass substrates at sheet resistivity = 15 W/sq from Semiconductor wafer Inc.

2.2 Photovoltaic Devices

Conventional organic photovoltaic with structure of ITO/PEDOT:PSS/PCDTBT: PC70BM/TiO_x/Al were fabricated. The ITO surface was used as the base substrate. The ITO surface was cleaned using detergent and then ultrasonication for 20 minutes in de-ionized water and iso-propanol, followed by drying with N_2 gas. Then, ITO substrates were separated in two surface treatments; oxygen plasma and UV light radiation. The characteristic of oxygen plasma equipment was power of 100 watt and oxygen plasma pressure of 0.2 mbar, while UV radiation treatment was carried out at wavelength of 395 nm (UV-A). Treatment time was varied in both methods. For oxygen plasma, treatment times of 15, 30, 60, 90 and 120 s were investigated where as for UV radiation techniques time treatment of 10, 15, 20, 25 and 30 min were used. The results were compared with untreated surface treatment. After treating surface, PEDOT:PSS, electron transporting layer, was coated on ITO glass substrate with Rapid Convective Deposition. Active layer was mixed from Poly[N-9′-heptadecanyl-2,7-carbazole-alt-5,5-(4′,7′-di-2-thienyl-2′,1′,3′-benzothiadiazole)] (PCDTBT) and phenyl-C_{70}-butyric acid methyl ester ($PC_{70}BM$) at 1:4 ratio by weight [38-39] in dichlorobenzene solvent by rapid convective deposition technique in ambient atmosphere. Finally, all samples were then transferred to the vacuum chamber of the thermal evaporator to make electron transporting layer and Al electrodes. The Al electrodes with a thickness of 200 nm were deposited on TiO_x is electron transporting layer of the photovoltaic devices as active area of 0.1 cm^2.

2.3 Characterization and Measurements

The surface roughness morphological features of the films were examined using the Seiko Instruments SPA-400 Atomic Force Microscope (AFM) system. The thin films thickness was tested by Avantes Ava-Thin Film. The sheet resistance of thin films was examined by Sheet Resistance Meter (SRM100) Four Point Probe Tester. The external quantum efficiency (EQE) was measured using an incident-photon-to-current efficiency measurement system by quantum efficiency model QEPVSIP-b. The current density-voltage (J-V) characteristics of the solar cell devices were measured by I-V measurements were conducted using source measurement unit (PXI-4130, National Instrument Inc.) Semiconductor Characterization System under an illumination from the Science Tech SS150 Solar Simulator at 100 mW/cm^2 with the AM 1.5 filter under ambient conditions. The contact angle measurement was characterized using DataPhysics PSL 250 socal. Conductivity of ITO films under different treatments was examined using conductive atomic force microscope (C-AFM, JPK NanoScience). A bias of 0.1V was applied to the sample. Gold tip silicon tip (TipsNano CSG01/Au, k ≈ 0.03N/m) was used. Crystal structure of surface treatment was measured by the Bruker D8 advance X-ray diffraction (XRD) technique with λ =1.5406 °A.

3. RESULTS AND DISCUSSION

3.1 Photovoltaic Characterization

Devices characteristic of conventional

cell structure, ITO/PEDOT:PSS/PCDTBT: PCBM/TiO_x/Al, were studied by varying the surface treatment techniques; oxygen plasma and UV radiation; on the performance of solar cells. Figure 1a-b show the J-V characteristics of the cells under illumination with oxygen plasma, UV radiation and compared with the best 2 techniques surface treatment with various times. The parameters extracted from the I-V curves are summarized in Table 1a-b. The reference cell with untreated film shows an open circuit voltage (V_{OC}), short-circuit current density (J_{SC}), fill factor (FF), and %PCE of 0.81 V, 13.33 mA/cm^2, 0.46, and 4.93%, respectively. The best performance under oxygen plasma and UV radiation, the optimize condition gave a short circuit current density of 15.93, 14.06 mA/cm^2 and open circuit voltage of 0.86, 0.85 V. were achieved with a fill factor of 0.52, 0.53, resulting in a power conversion efficiency (%PCE) of 7.24, 6.30% at 15 s in oxygen plasma and 20 min in UV radiation, respectively, when compared with untreated cell 4.93%. Consequently, a %PCE of oxygen plasma and UV radiation techniques was increased by 48, 28%, respectively compared to the untreated surface. Figure 1c showed EQE curves of the best cells. The external quantum efficiency (EQE) spectrum of devices, defined as the number of photogenerated carrier per incident photon, with and without surface treatment. The enhancement in EQE in range of 350-500 nm with oxygen plasma and UV radiation compared to untreated surface were observed. Clearly, after surface treatment, device performance was greatly improved, which was mainly ascribed to the improvement of the V_{OC} and FF. As a result, devices based on surface treatment showed higher performance than that based on untreated. These results demonstrate that surface treatment; oxygen plasma and UV radiation; is beneficial for improve ITO surface, and this will consequently lead to higher device performance. However, the oxygen plasma gave higher performance than UV radiation because of oxygen plasma has effected for removing organic residues from ITO surface Therefore, the enhanced uniformity of the electrical response of the oxygen plasma treated ITO is attributed to reduction of carbon contamination [40-42].

3.2 Sheet Resistance of PEDOT:PSS on ITO Substrates

The sheet resistance of surface treatment films was characterized by 4 point probe technique that coated PEDOT:PSS films on ITO substrates. From Figure 3 (a and b) show the sheet resistance graph in each surface treatment technique and can evaluate to the conductivity of thin films. These graphs were shown that the lowest sheet resistance gives the highest conductivity of 15 s in oxygen plasma treatment and 20 min in UV radiation, respectively. From these results supported that surface treatment in oxygen plasma technique can more improve the conductivity of PEDOT:PSS film on ITO substrates than UV radiation because of Helander et al [43] and Nagata et al [44] have found that the main role of UV light is to decompose the chemical bonds in PEDOT:PSS film, resulting in a decrease of conductivity, while in oxygen plasma the atomic oxygen are absorbed and oxidize the surface, leading to an increase of work function. Thus the oxygen plasma treatment is capable of controlling the work function and conductivity of PEDOT:PSS film, hence enable them to be modified to solar cell application.

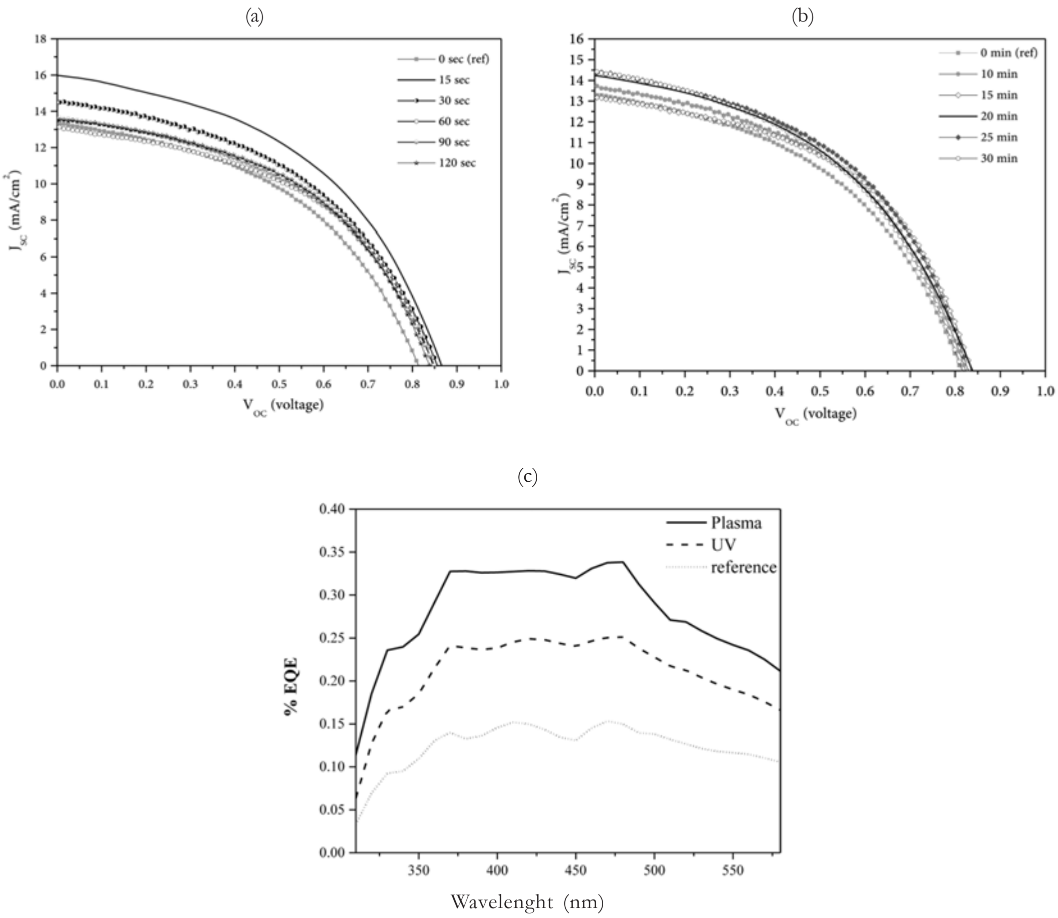

Figure 1. J-V characteristics of solar cells of (a) various oxygen plasma treatment time, (b) various UV radiation treatment time and (c) comparative external quantum efficiencies of different treatment types.

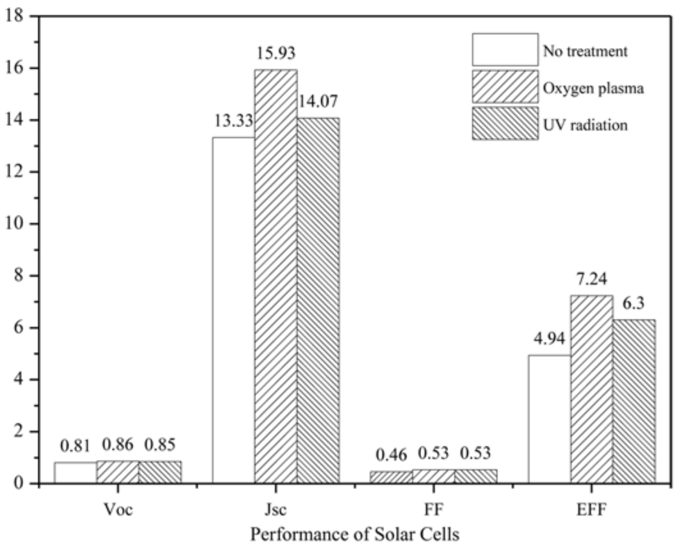

Figure 2. The characteristic parameters performance of solar cells.

Table 1a. Device parameters for the cells of PCDTBT:PCBM with the oxygen plasma surface treatment technique.

Time (sec)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	EFF (%)
Ref (untreated)	0.81 ± 0.005	13.33 ± 0.499	0.46 ± 0.011	4.94 ± 0.098
15 sec	0.86 ± 0.029	15.93 ± 1.284	0.53 ± 0.285	7.24 ± 0.769
30 sec	0.86 ± 0.009	14.54 ± 1.448	0.46 ± 2.481	5.67 ± 0.352
60 sec	0.85 ± 0.007	13.13 ± 0.287	0.48 ± 0.611	5.30 ± 0.141
90 sec	0.85 ± 0.001	13.64 ± 1.420	0.46 ± 1.801	5.37 ± 0.374
120 sec	0.84 ± 0.010	13.57 ± 1.827	0.48 ± 1.779	5.44 ± 0.947

Table 1b. Device parameters for the cells of PCDTBT:$PC_{70}BM$ with the UV radiation surface treatment technique.

Time (minute)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	EFF (%)
Ref (untreated)	0.81 ± 0.005	13.33 ± 0.499	0.46 ± 0.011	4.94 ± 0.098
10 sec	0.82 ± 0.020	13.70 ± 2.892	0.47 ± 0.845	5.34 ± 1.283
15 sec	0.84 ± 0.015	13.20 ± 0.791	0.49 ± 1.207	5.43 ± 0.534
20 sec	0.85 ± 0.005	14.07 ± 0.464	0.53 ± 0.932	6.30 ± 0.142
25 sec	0.83 ± 0.005	14.42 ± 1.653	0.47 ± 1.616	5.67 ± 0.513
30 sec	0.82 ± 0.011	14.43 ± 1.488	0.46 ± 1.910	5.37 ± 0.297

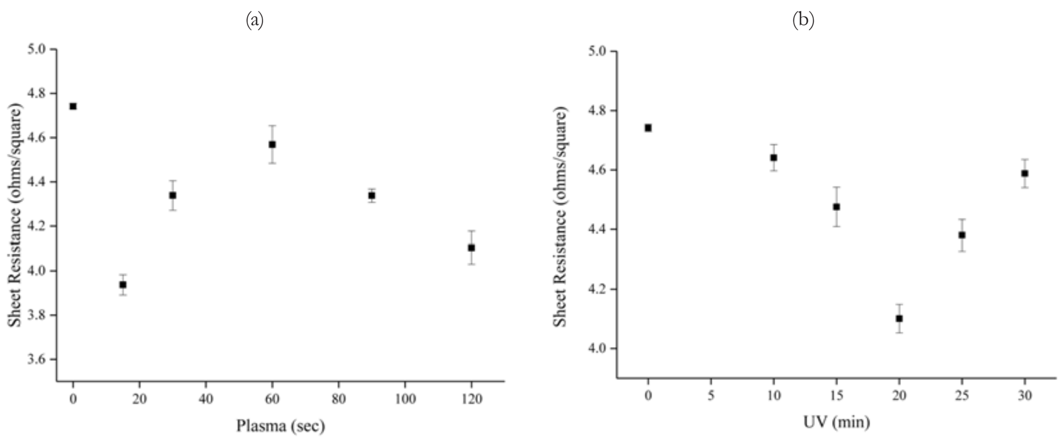

Figure 3. Sheet resistance of the PEDOT:PSS on ITO substrate after (a) oxygen plasma treatment (b) UV radiation treatment.

3.3 Contact Angle Measurement

The contact angle of surface treatment films was examined by contact angle machine that coated PEDOT:PSS films on ITO substrates. From Figure 4 a and b show the contact angle graph in two surface treatment techniques and can evaluate contact angle to the hydrophobic and hydrophilic properties of thin films. Comparing the PEDOT:PSS films on ITO substrates between untreated and treated surface was found that contact angle of the best surface treatment techniques; 15 s for oxygen plasma is 62° and 20 min for UV radiation is 64°; show the maximum contact angle when compared with untreated surface is 51°, respectively. It is well known that surface of PEDOT:PSS films on ITO substrate should be extremely hydrophobic after cleaning surface and PEDOT:PSS solvent can well dissolved in water so the treating PEDOT:PSS films on ITO substrates with plasma and UV radiation can made the surface to hydrophobic properties. By observing the contact angle was found that the contact angle of treated surface is greater than untreated surface. The rather enhancement in contact angle of PEDOT:PSS films on ITO substrate from 51° to 62° and 64° after treatment indicates that surface energy of ITO has increased. This result is similar to Kim et al [45-46] where surface energy was found to increase from an enlarged polar component after oxygen plasma treatment to improve wetting characteristic of PEDOT:PSS films on ITO substrate.

Also, oxygen plasma and UV radiation treatment surface not only can increase the ITO surface work function but also make it better hole injector [26]. From these results supported two surface treatment techniques; oxygen plasma and UV radiation; can appropriate to be the method to modify ITO surface of organic solar cells.

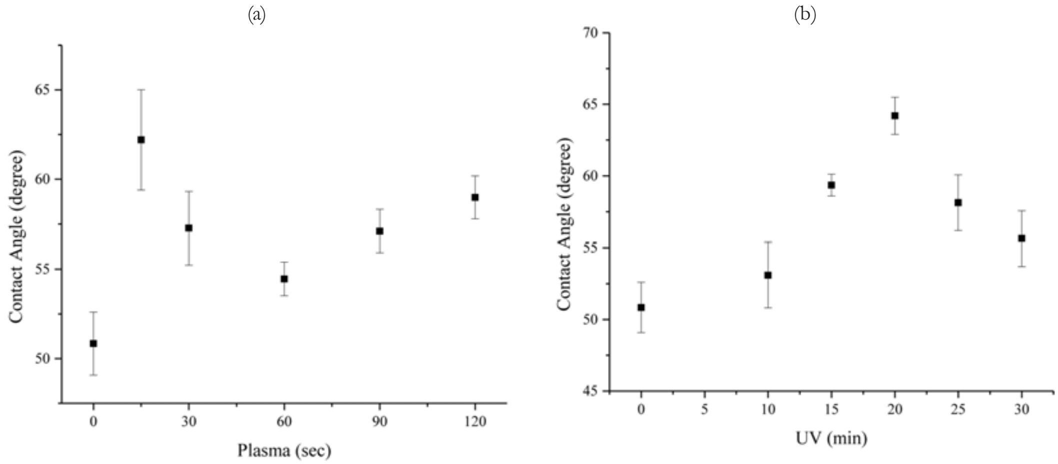

Figure 4. Contact angle of PEDOT:PSS films on ITO substrates after (a) oxygen plasma and (b) UV radiation treatments.

3.4 Thickness of the Thin Films

Figure 5 show the thickness of PEDOT:PSS films on ITO substrates by two treatment surface techniques; oxygen plasma and UV radiation. The graph tendency of two surface treatment techniques was shown the same variation. The thickness data present oxygen plasma and UV radiation in range of 115-119, 116-123 nanometers, respectively compared with untreated surface is 112 nm. With consideration of the best performance solar cells from two surface treatment techniques was found that at 15 s for oxygen plasma gives the lowest thickness but 20 min from UV radiation presents the highest thickness. The thin layers of PEDOT:PSS film can help charge to fast transfer to electrode [47]. These results support the oxygen plasma is an optimal technique to treatment surface for improving the efficiency of ITO substrate in organic solar cell.

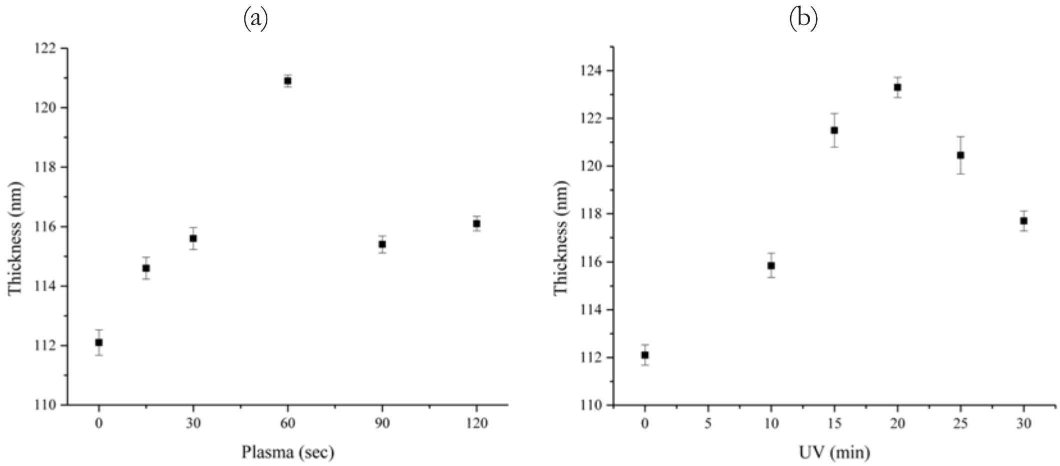

Figure 5. Thickness of PEDOT:PSS films on ITO substrates after (a) oxygen plasma and (b) UV radiation treatments.

3.5 Surface Roughness and Local Conductivity

Figure 6 a-c show the topography of PEDOT:PSS films on ITO substrates with treated and untreated surface. The root mean squared surface roughness (RMS) values have been extracted from the topography images of the films prepared with treated and untreated surface. The film surface handled with oxygen plasma and untreated surface appears quite smooth with the RMS roughness of 5.62 and 5.63 nm respectively, while the RMS roughness of UV radiation is 5.83 nm. The results are summarized in Table 2. From data indicates that the oxygen plasma treatment has little effect on the morphology of PEDOT:PSS films. In general, a smooth surface morphology of the films should lead to a better contact with the metal electrodes [44]. From such effect improves the interface contact between PEDOT:PSS and active layer, which increases the extraction efficiency of the photogenerated holes and decreases the recombination likelihood of charge in the active organic layers [48]

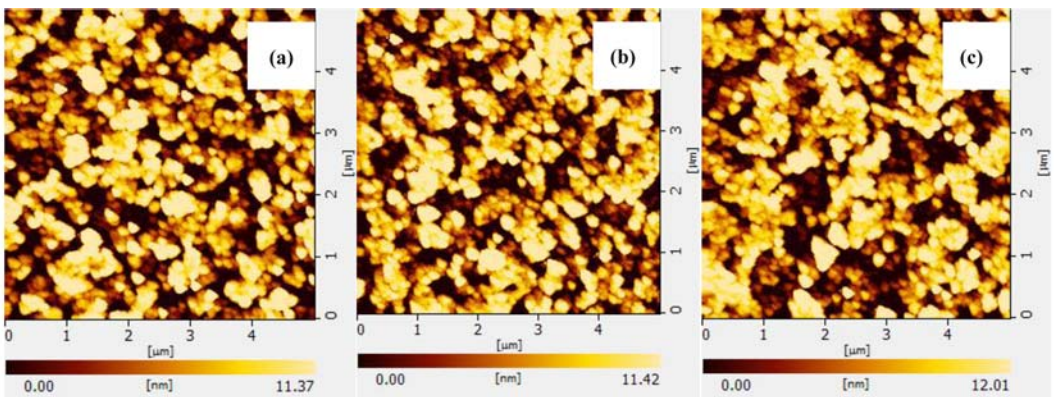

Figure 6. AFM topographic images at 5×5 μm^{2}of PEDOT:PSS films on ITO substrates with (a) untreated, (b) oxygen plasma and (c) UV radiation.

Table 2. The root mean square roughness (RMS) of PEDOT:PSS films on ITO substrates at various treatments.

Treatment	RMS roughness (nm)
Untreated surface	5.63
Oxygen plasma	5.62
UV radiation	5.83

Figure 7 shows representative topographical images and their corresponding current maps of ITO substrates with and without surface treatments. Noted that five areas were randomly selected to characterize and similar results were obtained. Topographical images show similar surface features on untreated and treated substrates. The root mean square roughness values of untreated, UV-treated and O_2 plasma treated substrates respectively are 6.55, 7.0 and 7.5 nm, indicating that ITO substrate becomes slightly rougher after both surface treatments. Current images clearly reveal that the ITO becomes more conductive after surface treatments. Average currents of untreated, UV-treated and O_2 plasma treated substrates are 1.7, 23.5 and 40.8 nA, respectively. Conductive-AFM data confirm that both surface treatments improve conductive of ITO substrates significantly, facilitating better charge transport at contacts which is in good agreement with the improvement in the overall solar cell performances.

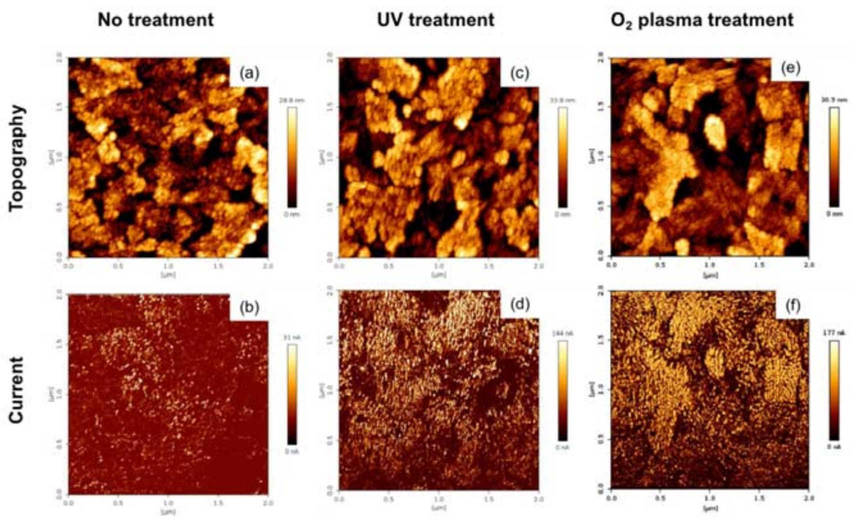

Figure 7. AFM micrographs (2 × 2 μm^2) showing topography and corresponding current images of ITO substrates of (a, b) untreated, (c,d) UV treatment and (e, f) oxygen plasma treatment.

3.6 X-ray Diffraction

Figure 8 shows the XRD patterns of PEDOT:PSS films with a different surface treatment compared with untreated surface on silicon substrate. From Figure 8b displays XRD peaks of Si substrate at 33° and 34°, respectively when compared with Figure 8a was shown that there are a widely peaks from 15°-45° in all three films. The broadness of peaks reflects the semi-crystalline nature of PEDOT:PSS. This suggests that the crystallization behavior of surface treatment, O_2 plasma and UV radiation, on PEDOT:PSS film is unchangeable.

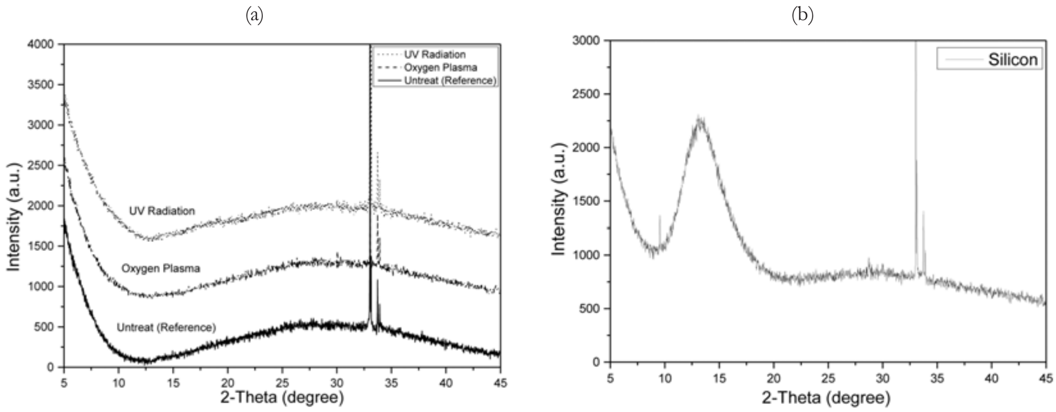

Figure 8. X-ray diffraction of PEDOT:PSS on ITO substrates after (a) oxygen plasma, UV radiation and untreated (b) Silicon reference.

4. Conclusion

We have demonstrated that surface treatment techniques, namely oxygen plasma and UV radiation techniques, can improve conductivity of ITO substrates of organic solar cells. The optimal time for surface treatment of oxygen plasma and UV radiation is 15 s and 20 min, respectively. From results revealed that the efficiency of solar cells in oxygen plasma and UV radiation method are 7.24 and 6.30%, respectively, while the efficiency of untreated surface is 4.97%. EQE described the achievement of the solar cells efficiency after surface treatment techniques. Both surface treatment methods effectively increase the work function of ITO substrate and can improve the hydrophobic properties of PEDOT:PSS layer on ITO substrate from contact angle measurement. Conductive-AFM data and local 4 point probe technique confirmed that both surface treatments improve conductive of ITO substrates significantly. AFM technique showed that roughness and topography of all films were similar.XRD results indicated the unchangeable of crystal structure under surface treatments. From these results supported two surface treatment techniques; oxygen plasma and UV radiation; can suitable to be the method to qualify ITO surface of organic solar cells.

In this research chose a conventional and simple method to treat ITO substrate by O_2 plasma and UV radiation methods with Rapid Convective Deposition (RCD) that is a specific technique. The advantages of RCD technique are create uniform films, use a small amount in microliter solution for research scale, can controllable film thickness by handle machine speed, can fabrication in ambient atmosphere and its scalability. The prominent points of RCD is cost reduction therefore it's a suitable technique to fabricate low cost organic solar cells.

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GOV-59-29.

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