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Bulk Heterojunction Tandem Photoelectric Cell Based on p-Si and Phthalocyanine

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An organic-inorganic (p-Si and phthalocyanine) hybrid tandem heterojunction Ag/p-Si/AlPc:H_2Pc/ITO photoelectric cell was fabricated by pressing technology using preliminary vapor deposited heterojunction films of mixed aluminum-phthalocyanine (AlPc) and metal free phthalocyanine (H_2Pc) on p-Si substrate and on ITO coated plastic substrate. By keeping organic films face to face both substrates were pressed and fixed together by adhesive at elevated temperature. Total thickness of the AlPc and H_2Pc films were equal to 300 nm. On the back side of p-Si substrate the Ag film was deposited. The device architecture was the following: Ag/p-Si/AlPc:H_2Pc/ITO. The morphology of the organic semiconductors film was investigated by AFM. The optical properties of the AlPc:H_2Pc film were studied by UV-visible spectroscope. Current–Voltage characteristics were measured in dark and also illumination conditions. Under illumination of 296 W/m^2 the values of V_{oc}, I_{SC}, *FF* and efficiency were equal to 0.5 V, 4 mA, 0.45 and 0.61 %, respectively. The *I-V* and *P-V* characteristics of the solar cell were simulated by using Shockley equation and its Newton Raphson solution, respectively for dark and illumination conditions. The obtained simulated results were in good agreement with the experimental results.

Keywords: pressing-technology; thermal evaporation; organic-inorganic; phthalocyanine; photoelectric cell

1. INTRODUCTION

Experts predict that to sustain the world economic growth about 30 TW energy will be needed in year 2050 [1]. Out of all energy sources, the role of electric power is very important in industrial as well as in domestic applications. It is expected that up to year 2020 about 4 % of the world's energy will be contributed by photovoltaic system. As sun is the free and eco-friendly source of energy, so it is very important that the solar cell (device to convert light energy into electricity) must be cost effective and environmental friendly as compared to conventional sources. It means that the low cost and eco-friendly materials and technology should be used for devices fabrication. For laboratory to commercial integration the crystalline silicon solar cells are considered successful and they cover 90 % of solar cells market but they are less cost effective. This problem can be solved by the use of less material and also by raising the efficiency (energy conversion) of the solar cells [2]. This task can be achieved by the development of organic-inorganic solar cell technologies and its implementation because the organic materials are low cost, easily available, flexible, light weight, easy to process and environmental friendly [3-5]. Moreover, the simple and low cost techniques are used for the deposition of organic thin films such as spin-coating deposition of organic semiconductors from solution, doctor blade, screen printing, vacuum deposition at lower temperatures (400-600 °C) and also the ink-jet printing technology [6-10]. The above mentioned fascinating features attracted the researchers to explore the potential of organic materials to replace or reduce their inorganic counterparts in semiconducting industry. The number of organic materials have been investigated from last few decades and organic materials based light emitting diodes (OLEDs) and various types of sensors have been commercialized [11, 12]. Being a no-toxic and stable p-type semiconducting materials the phthalocyanine and its derivatives are pertinent for optical and electronic devices like solar cells, OLEDs, OFETs, Batteries and sensors [13-16].

The 5 % efficiency of the OSCs was considered minimum required efficiency for its practical application [17]. Based on his calculations the Chamberlain predicted that in Schottky barrier SC up to 10 % efficiency can be realized [18, 19]. There are also forecast that higher than 20 % efficiency is achievable in the organic solar cell (single junction) [19, 20]. The organic solar cells were investigated in various designs which includes single layer, heterojunction or bi-layer and bulk heterojunction (BHJ) solar cells [21-25]. Amongst the solar cells the bulk heterojunction organic-inorganic solar cells are very talented. Being an inorganic semiconducting material single crystal or polycrystalline p-type or n-type silicon wafer can be used for organic-inorganic solar cells. One of the fabricated organic-on-inorganic cells (Ag/n-GaAs/p-CuPc/Ag) showed efficiency of around of 4 % [26]. Tandem approach for organic semiconductor solar cells allow to improve performance of the devices due to overlapping of absorption spectra of the several stacked cells [27, 28]. In this paper in continuation of our efforts for the fabrication and investigation of solar cells [10, 29, 30] we are describing the fabrication and investigation of organic-inorganic hybrid tandem heterojunction cell based on AlPc:H_2Pc bulk heterojunction and p-Si cells.

2. EXPERIMENTAL

The phthalocyanine, aluminium phthalocyanine, p-silicon, ITO coated plastic substrates and all other required materials were purchased from Sigma Aldrich (web. add). The chemical formula and the molar mass of the metal free phthalocyanine (H_2Pc) are $C_{32}H_{18}N_8$ and 514.55 g M^{-1}, respectively, while that of aluminium phthalocyanine (AlPc) are $C_{32}H_{18}AlN_8$ and 541.51 g M^{-1}, accordingly. The molecular structures of H_2Pc and AlPc are respectively shown in Fig.1 (a) and Fig.1 (b). The thickness of boron doped crystalline silicon (p-Si) wafer was 0.5 mm. Equal amounts (wt) of both (H_2Pc and AlPc) powders were mixed together homogenously by using mortar and pestle and then pressed in pellet form. This pellet was used in thermal evaporator to deposit H_2Pc-AlPc thin films.

The solar cell shown in Fig.2 was prepared by using thermal evaporator (Edwards AUTO 306) for the deposition of organic and metallic thin films. The organic film (H_2Pc-AlPc) of thickness 150 nm was deposited on the ITO plastic substrate as well as on one face of the p-Si wafer. On the other face of p-Si wafer 100 nm thick silver (Ag) film was deposited as an electrode. The thermal deposition was carried out under the vacuum higher than 10^{-6} Torr, while the rate of deposition was 0.1 nm/s. The film thickness was measured using FTM5 quartz crystal monitor. The sizes of the H_2Pc-AlPc films deposited on ITO and p-Si wafer were equal to 25:20 mm^2. The p-Si wafer and ITO were kept in a way that the organic (H_2Pc-AlPc) films of both the sample faced each other then the pressing was done at 60-80 $^{\circ}C$ by applying (4.7-5.7) x 10^{-2} kgf/cm^2 pressure for 20 to 30 min. The adhesive was applied to fix the pressed samples. Figure 2 shows the samples before (a) and after (b) pressing. The device architecture was the following: ITO/AlPc:H_2Pc/p-Si/Ag. The active area of the device was 500 mm^2.

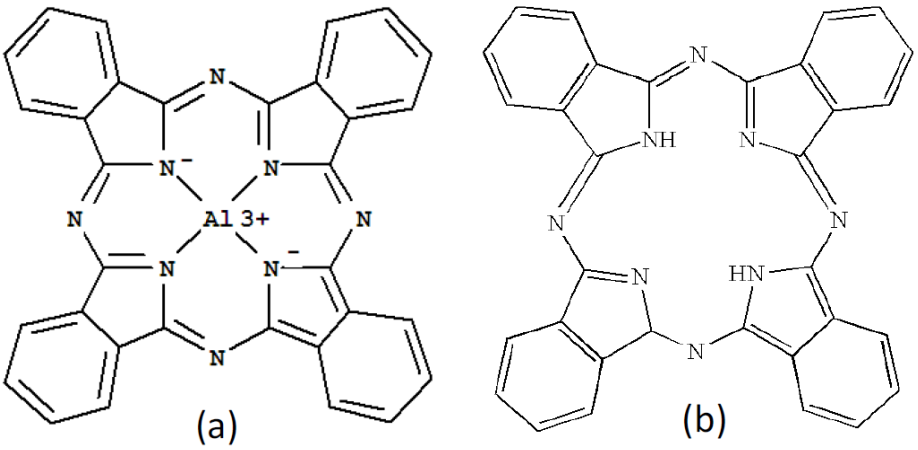

Figure 1. Molecular structures of (a) aluminum phthalocyanine (AlPc) and (b) metal free phthalocyanine (H_2Pc), respectively [29].

As a light source the filament bulb was used for the testing of fabricated cells. For the measurements of voltage and current the digital multimeter HIOKI DT 4253 was used, while the temperature was measured using digital multimeter FLUKE 87.

Materials which were used for fabrication of the photoelectric cell have the following work functions: 4.7 eV (ITO), 3.8 eV (AlPc), 4.04 eV (H_2Pc), 4.8 eV (p-Si) and 4.3 eV (Ag). It can be

considered that AlPc and H_2Pc form bulk heterojunction cell, p-Si forms second cell where top electrode is formed respectively by AlPc and H_2Pc film. Accordingly p-Si plays a role of bottom electrode of AlPc and H_2Pc bulk heterojunction photoelectric cell.

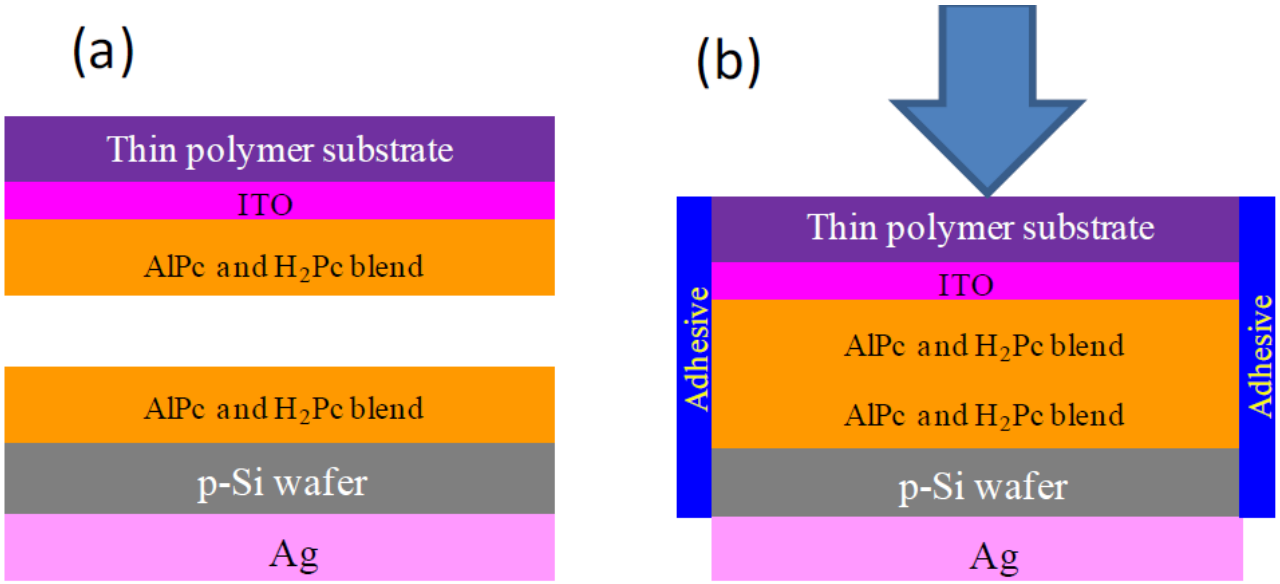

Figure 2. Schematic diagram of Ag/p-Si/AlPc:H_2Pc/ITO photoelectric cell before (a) and after (b) pressing

3. RESULTS AND DISCUSSION

The Fig. 3a shows the H_2Pc-AlPc film's atomic force microscope (AFM). It is evident from the surface morphology (Fig. 3a) that the deposited H_2Pc-AlPc film is well developed and pore-less. These properties are imperative to get the high yield of the incident light. The H_2Pc-AlPc mixture was characterized optically using UV-Visible spectroscopy. The absorption spectrum of the H_2Pc-AlPc mixture is shown in Fig. 3b. It is evident from the Fig. 3b that the strong absorption takes place at 715 nm, which can be regarded to the transition from valance band to conduction band.

The current-voltage (I-V) characteristics of bulk heterojunction Ag/p-Si/AlPc:H_2Pc/ITO photoelectric cell in dark and room temperature conditions are shown in Fig. 4a. The positive potential (forward bias) was applied on the ITO, while the negative potential was applied on silver (Ag) electrode. In Fig. 4a existence of rectification behavior can be observed. The rectification ratio (RR) determine as I_F/I_R, where I_R and I_F are reverse and forward bias currents at voltage equal to 0.8 V and the calculated value of RR is equal to 2.5. In the organic semiconductors this type of I-V characteristics are very common [11, 22]. The Fig. 4b shows I-V characteristics of the fabricated solar cell under illuminations of 296 W/m^2 and 256 W/m^2. The estimated efficiency of the solar cell was equal to 0.61 %. There is also similarity among the I-V characteristics of the fabricated cell and the characteristics of already investigated organic semiconductors [11, 22].

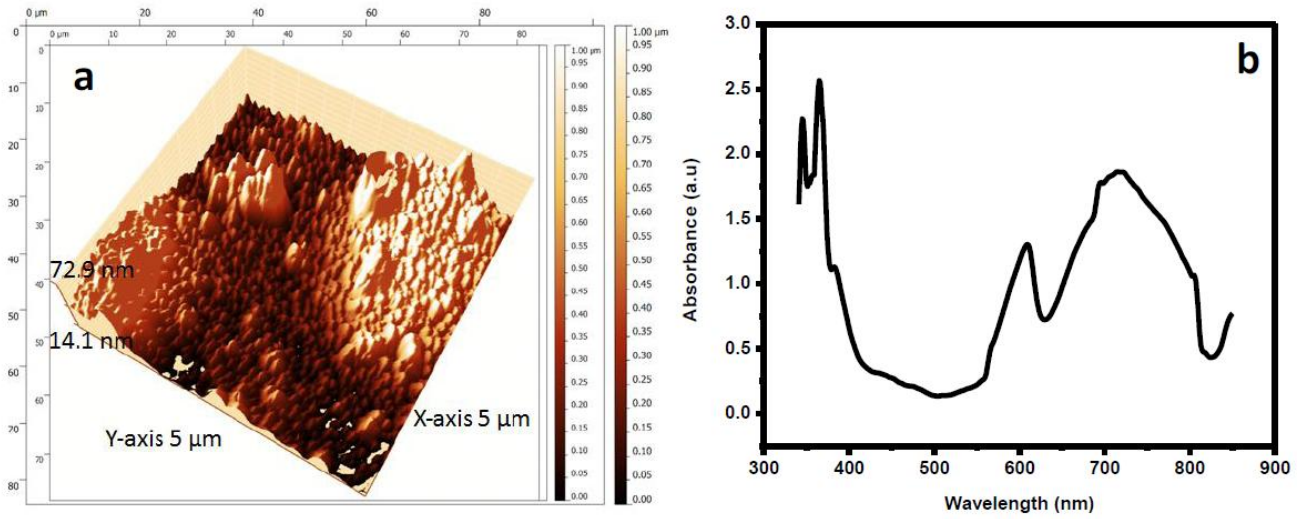

Figure 3. AFM image showing the surface morphology of H_2Pc-AlPc film (a) and the absorption spectrum of H_2Pc-AlPc mixture (b) [29].

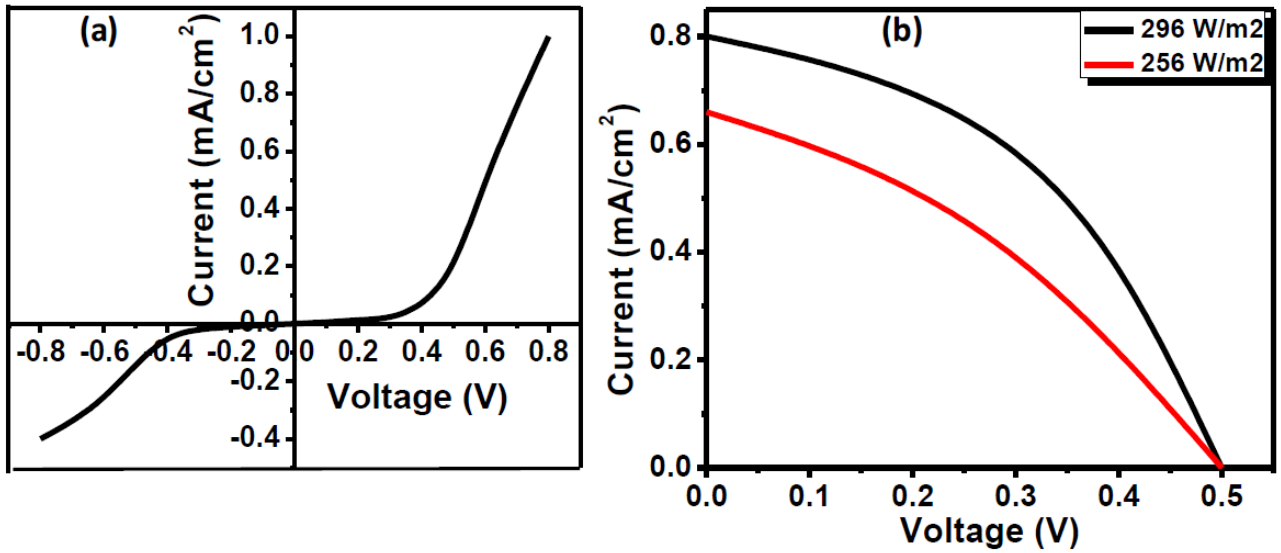

Figure 4. I-V characteristics of the Ag/p-Si/AlPc:H_2Pc/ITO solar cell in dark condition (a) and under illumination (b).

The maximum power point (MPP) is the imperative parameter of the I-V characteristics measured under illumination (Fig. 4b). To determine the MPP of a solar cell various methods were explained by Freeman et al. in ref. [31]. For modules the MPP tracking is described in [32], while the MPP tracking-control techniques have been discussed in [33]. For current work we adopted the method that was used by Faranda et al. in ref. [34]. The 76 % of the V_{oc} was selected as an operating point optimum voltage (V) and consequently the current density (J) at MPP was obtained. For the I-V characteristics shown in Fig. 4b the V (optimum voltage) and the J (current density) were obtained for the illuminations of 296 W/m^2 (V_1 = 0.38 V and J_1 = 0.417 mA/cm^2) and 256 W/m^2 (V_1 = 0.38 V and J_1

= 0.253 mA/cm^2). For the estimation of the coordinates of MPP from the I-V curve a method given in [49] can be used, which is simple as compared to methods given in [31-33].

For photoelectric cells the stability is very essential parameter. It is already reported that phthalocyanine based inverted structured photoelectric cells are more stable than the conventional structured cells (ITO/(ITO/PEDOT:PSS/Organic layer/Al)/Organic layer/Al) [35]. The reason behind the lower stability of the conventional structured cells is the oxidation of aluminium (Al) layer at Al/organic interface which causes to reduce the conductivity of Al electrode and also creates the recombination sites in semiconductor layer due to the diffusion of Al. Moreover the ITO gets crude due to the acidic nature of PEDOT:PSS which also causes to destabilize the cell [35, 36]. In the inverted cell (ITO/TiO2/organic layer/PEDOT:PSS/Au) the PEDOT:PSS is not in direct contact with ITO and the Al is not used. So, the inverted structured photoelectric cells showed the stable low efficiency (η= 0.059 %).

Bulk heterojunction tandem photoelectric cell shown in Fig. 2 has the organic semiconductor (AlPc:H2Pc) film in contact only with p-Si wafer and indium tin oxide (ITO), that may provide a longer life to the Ag/p-Si/AlPc:H_2Pc/ITO photoelectric cell. These cells showed a stable behavior after one month of fabrication. The detailed stability study of the Ag/p-Si/AlPc:H_2Pc/ITO cells will be conducted in near future, where the effects of environmental condition and annealing temperature and time will be investigated. Earlier, degradation study of Ag/n-GaAs/p-CuPc/Ag photoelectric cell showed that being a fresh cell (in year 2000) it had 4 % efficiency (power conversion) but this efficiency degraded exponentially to 0.6 % in next five years which was then stable for further 10 years (until 2015) [26].

As the Ag/p-Si/AlPc:H_2Pc/ITO cells have been fabricated using pressing technology in which the two bulk heterojunction films of the same composition deposited on different electrodes were pressed together. This technology makes the cell's fabrication process more reliable and its properties more predictable. Unlike to tandem or stacked cells [37, 38] the Ag/p-Si/AlPc:H_2Pc/ITO cells have no connecting layer (conductive) between two cells as implemented in the heterojunction structure [39]. In short, the Ag/p-Si/AlPc:H_2Pc/ITO cell has combination structural features of both tandem and heterojunction cells.

The I-V characteristics comparable to that of Ag/p-Si/AlPc:H_2Pc/ITO solar cell (Fig. 4a) were experienced in other studies on copper phthalocyanine (CuPc) or orange dye Schottky or bi-layer semiconducting devices [40]. The Fig. 5a shows the relationship of rectification ratio and applied voltage (RR-V) of the Ag/p-Si/AlPc:H_2Pc/ITO solar cell: it is evident that the rectification ratio depends upon the voltage. For the organic semiconductor devices in the narrow potential (0-0.5V) range the modified Shockley equation may be used to evaluate the dark I-V characteristics [29, 41, 42]:

$$I = I_o \, [exp \, \{q(V\text{-}IR_s)/nkT\} - 1] + (V - IR_s) / R_{sh} \qquad (1)$$

where I is the current, V is the terminal voltage, R_s series resistance, R_{sh} shunt resistance of the device, while T, n and k are the absolute temperature, diode quality or ideality factor and Boltzmann constant, respectively.

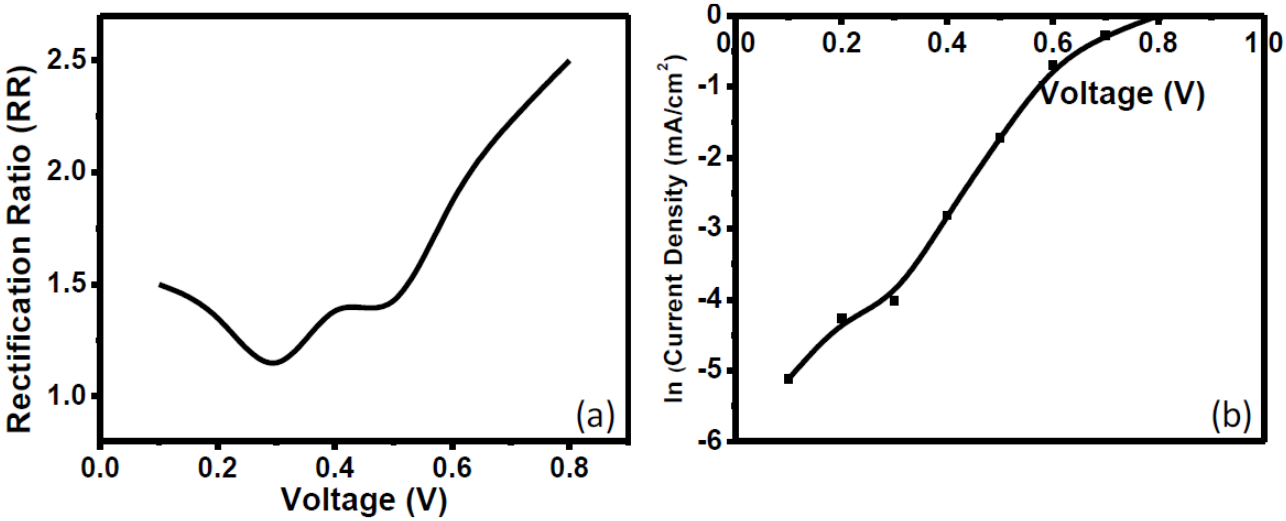

Figure 5. (a) Rectification ratio versus voltage (RR-V) and (b) ln(current density) versus voltage (ln(I)-V) relationships of Ag/p-Si/AlPc:H_2Pc/ITO solar cell

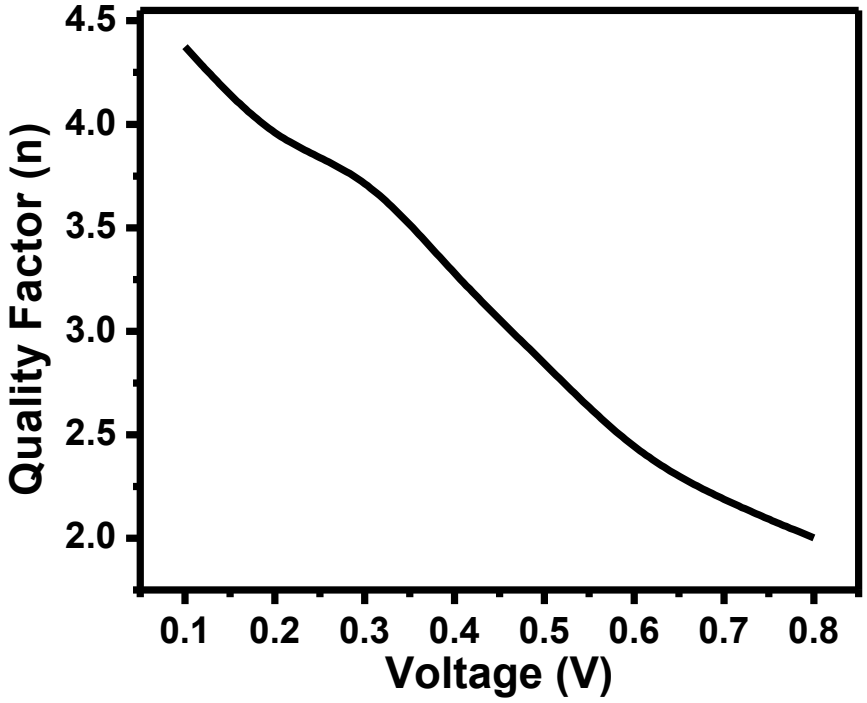

Figure 6. Quality factor-Voltage relationship of Ag/p-Si/AlPc:H_2Pc/ITO solar cell

The I_o represents the reverse saturation current as given in ref. [41]:

$$I_o = A^* T^2 \exp(- q\Phi / kT) \quad (2)$$

where A^* and $q\Phi$ are the Richardson constant and Shottky barrier [39]/height of contact barrier, respectively. The R_s and R_{sh} given in Eq. 1 were calculated from *I-V* characteristics shown in Fig. 4a in forward bias (lowest resistance) and in reverse bias (highest resistant), respectively. The values of R_s and R_{sh} are 160 Ω and 400 Ω, accordingly. The data of *I-V* curve shown in Fig. 4a is also used to find the reverse saturation current (I_o) by using the method devised in [39] and applied in [42]. So, the

intersection of *ln(I)* versus *V* curve (Fig. 5b) at $V = 0$ gives the value of I_o, which is 0.0124 mA (0.0025 mA/cm^2 (current density)).

For the *I-V* characteristics shown in Fig. 4a the effect of applied voltage on the quality/ideality factor (*n*) of the cell can be determined by Using Eq. 1. The reliance of the quality factor (*n*) of Ag/p-Si/AlPc:H_2Pc/ITO solar cell is shown in Fig. 6. From the Fig. 8 it can be clearly seen that the quality factor depends upon the voltage and it decreases with rising voltage. Upon changing voltage from 0.1 V to 0.8 V the quality factor decreases from 4.37 to 2.00. Moreover, the results shown in Fig. 4b can also be simulated by using the Shockley equation for only the light induced current as carried out in [43].

The I-V characteristics of the Ag/p-Si/AlPc:H_2Pc/ITO solar cell shown in Fig. 4b have been simulated using the modified Shockley equation [44]:

$$I = I_{pv} - I_o\left[\exp\left(\frac{V+IR_s}{aV_t}\right) - 1\right] - \frac{V+IR_s}{R_p} \quad (3)$$

In Eq. 3 I_{pv} *is the* photovoltaic current, I_o is reverse saturation current, R_s and R_p are the series and parallel resistances of the solar cell and a is the diode quality or ideality factor, while the V_t is called the thermal voltage represented as ($V_t = N_sKT/q$), where N_s, K, T and q denote the number of cells connected in series, Boltzmann constant (1.3806503x10^{-23}), temperature in Kelvin and charge of electron (1.60217646x10^{-19} C), respectively. The values of R_s and R_p were determined by the *I-V* curve as shown in Fig. 7 and given in Table-1. The value of I_{pv} was calculated by the following equation [43]:

$$I_{pv} = \left(\frac{R_p+R_s}{R_p}\right)I_{sc} \quad (4)$$

Where R_p, Rs and I_{sc} are the parallel resistance, series resistance and the short circuit current (Fig. 4b), respectively.

Moreover, the value of reverse saturation current (I_o) was found by the following equation:

$$I_o = \frac{I_{pv} - (V_{oc}/R_p)}{\exp\left(\frac{V_{oc}}{aV_t}\right) - 1} \quad (5)$$

To solve the modified Shockley equation (Eq. 3) the *f(I) = 0* was found and then differentiated with respect to *I(f′(I))* and finally substituted in the following expression:

$$I_{n+1} = I_n - \frac{f(I_n)}{f'(I_n)} \quad (6)$$

The final solution also called Newton Raphson solution of the modified Shockley equation is the following:

$$I_{n+1} = I_n - \frac{I_n - I_{pv} - I_o + \left(\frac{V+R_sI_n}{R_p}\right) + I_o \exp\left(\frac{V+I_nR_s}{aV_t}\right)}{1 + \frac{R_s}{R_p} + I_o\left(\frac{R_s}{aV_t}\right)\exp\left(\frac{V+I_nR_s}{aV_t}\right)} \quad (7)$$

The parameters used in Eq. 7 are experimentally measured or calculated from the experimental data. For simulation the coding is done in MATLAB and its algorithm is given in Fig.7. The arbitrary value is used to initiate ideality factor. As an initial estimate I_n is taken equal to I_{sc} ($I_n = I_{sc}$). The diode ideality factor (a) act as a fitting parameter. The "For" loop continues in the range of $0 \leq V \leq V_{oc}$ to find the new values of I_{n+1} (current). Power is calculated for every iteration and for each loop the newly found value of current (I_{n+1}) is assigned to I_n. The loop continues until the simulated maximum power is approximately equal to experimental maximum power ($P_{m\text{-}sim} = P_{m\text{-}exp}$). The results of the simulation

of *I-V* and *P-V* characteristics of the Ag/p-Si/AlPc:H_2Pc/ITO solar cell under 256 W/cm^2 and 296 W/cm^{2}are shown in Fig. 8 and Fig. 9, respectively. It can be seen from Figs. 8 and 9 that the simulated results are well matched with the experimental results.

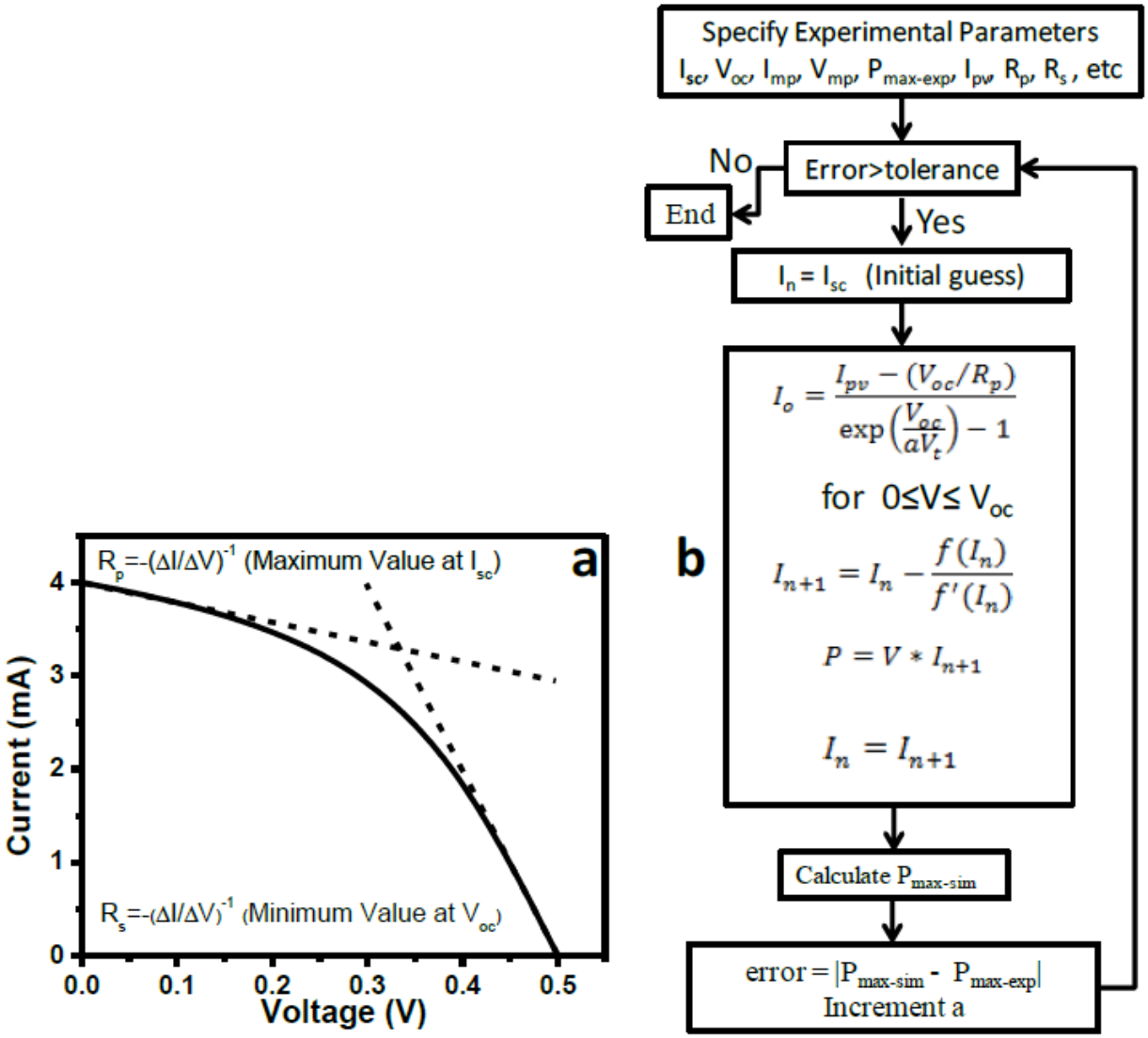

Figure 7. (a) Calculation of R_s and R_p from the *I-V* curve and (b) the algorithm used for the simulation of Ag/p-Si/AlPc:H_2Pc/ITO solar cell.

Table 1. Various parameters of the Ag/p-Si/AlPc:H_2Pc/ITO solar cell

Illumination G (W/m^2)	Temperature (°C)	Parallel resistance R_p (Ω)	Series resistance R_s (Ω)	Photovoltaic current I_{pv} (mA)	Reverse saturation current I_o (mA)
256	25	347	89.9	4.43	2.03x10^{-10}
296	31	458	49.4	4.15	4.3x10^{-9}

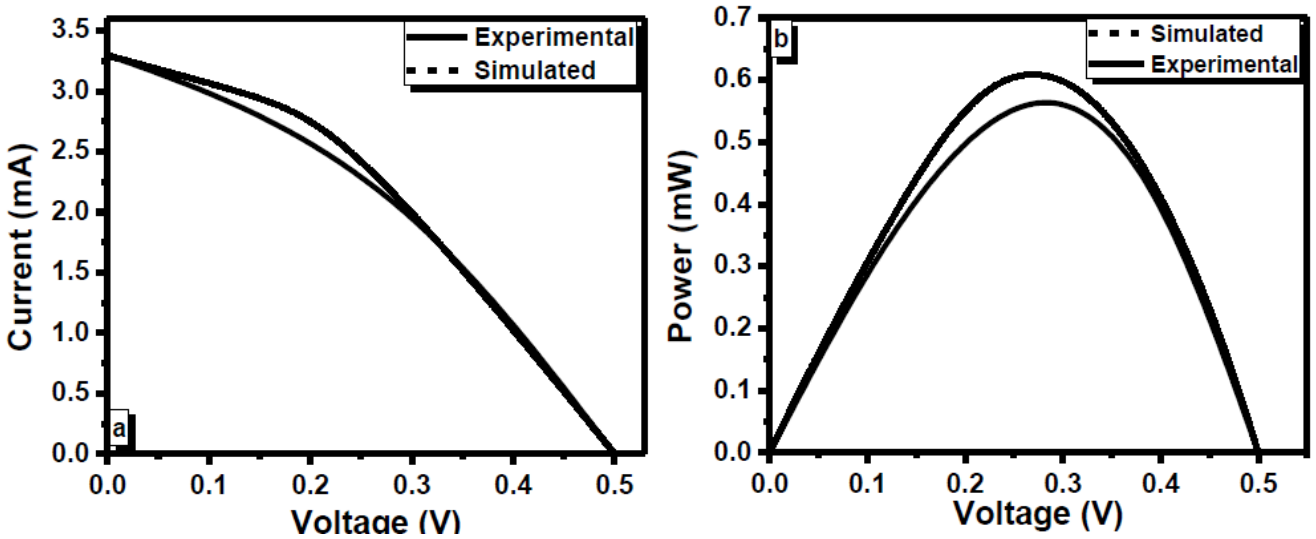

Figure 8. The comparison of experimental and simulated *I-V* and *P-V* characteristics of Ag/p-Si/AlPc:H_2Pc/ITO solar cell under illumination of 256 W/cm^2.

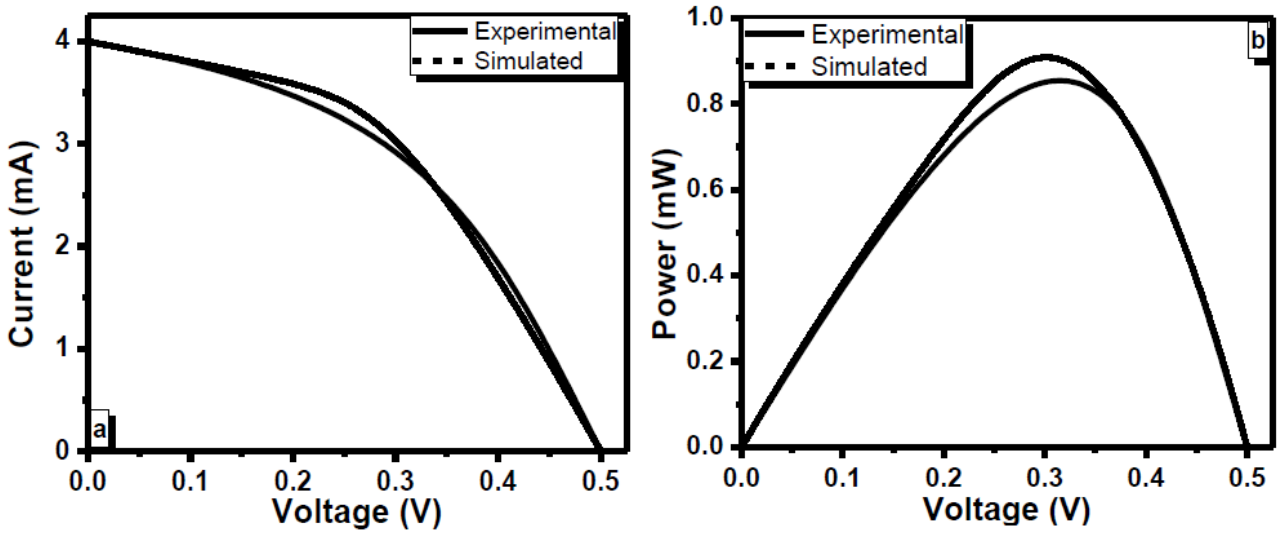

Figure 9. The comparison of experimental and simulated *I-V* and *P-V* characteristics of Ag/p-Si/AlPc:H_2Pc/ITO solar cell under illumination of 296 W/cm^2.

4. CONCLUSION

Bulk heterojunction tandem photoelectric cell based on p-Si and phthalocyanine (ITO/AlPc:H_2Pc/p-Si/Ag) was fabricated using thermal evaporation and pressing technique. The cell's current-voltage (*I-V*) behavior was investigated in dark and also in illumination conditions. The obtained experimental results were simulated using modified Shockley equation and its Newton Raphson solution. By the simulation of the experimental results various cell parameters were calculated. The simulated results of *I-P* and *I-V* characteristics were in good agreement with experimental results. The fabrication technique used in this work can also be adopted for the

development of solar cell technology. Additionally, the easiness of preparation makes the fabricated cells apposite to be used as a teaching aid in the educational institutes.

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