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Performance Enhancement of Bulk Heterojunction Hybrid Solar Cell Using Macroporous Silicon

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Abstract

This article focuses on the work which deals with the effect of introducing macroporous silicon as the cathode of hybrid solar cell. This work shows that the photocurrent of bulk-heterojunction hybrid solar cell can be enhanced by using macroporous silicon (macro-PSi) as the cathode that provided increased effective contact surface area at the interface of organic-inorganic material. The organic compound (3-hexylthiophene) (P3HT) and phenyl-C61-butyric acid methyl ester (PCBM) polymer blend at 1:1 ratio was used to fabricate the solar cell. It was found that the pore-diameter of the porous silicon plays an important role on short-circuit current of the fabricated hybrid solar cell. Huge enhancement of short-circuit current density (~ 73 times) was obtained when the average pore diameter of macro-PSi was comparable to the photogenerated carrier transport length of the photoactive polymer. The annealing of the whole structure further enhanced the overall performance of the fabricated hybrid solar cell.

Keywords: Hybrid-solar-cell; Macroporous; Exciton; Bulk-heterojunction; Electrochemical-etching; Fill-factor; Conversion-efficiency.

INTRODUCTION

Hybrid solar cells are consisting of organic and inorganic materials. Hybrid solar cells hold promise for low cost energy production [1-3], but only if further developments in their design and manufacture make them practical for real-world applications. When light is absorbed in organic semiconductors, bound electron–hole pairs known as excitons are generated. The electrons and holes separate from each other at an interface between two semiconductors by electron transfer [4]. In conventional organic solar cell the photocurrent is limited due to the poor charge collection efficiency. The charge collection may drastically increase by producing porous cathode (charge collector) instead of flat collector. The average pore's size of the porous cathode should be comparable to the charge carrier diffusion length to enrich the charge transportation. In the conventional photoactive polymers like P3HT:PCBM the transport length is reported to be 180 nm [5]. Therefore, it is advantageous to form well-ordered nanostructures which will provide large interface with an organic semiconductor to inorganic semiconductor. As a result most of the excitons are expected to reach at the interface between the two semiconductors and all of the charge carriers have a pathway to the appropriate electrode. This will increase the charge collection efficiency of a solar cell [6]. In this work enhancement in the short-circuit current density was observed when Si nanowires (NW) were embedded in the bulk-heterojunction (made of P3HT:PCBM) solar cells [7, 8]. Similarly a porous matrix, particularly porous silicon (PSi) matrix [9-11], appears to be an ideal material for this purpose [12-15] as its pore-size, morphology and the conductivity of the PSi layers can be controlled [16, 17]. Further the efficiency of hybrid solar cell is bounded because of the limited contact surface area at the interface of organic and inorganic materials. The large effective contact surface area can be achieved by using porous cathode.

In addition PSi has also been investigated as an anti-reflection coating in the hybrid solar cells for extending and enhancing the absorption of hybrid solar cells [18-19]. So the PSi also works in light trapping.

In this work blend of P3HT (poly-3-hexylthiophene) and PCBM (Phenyl C61 butyric acid methylester) at 1:1 ration has been used as photo active material and macroporous silicon (macro-PSi) has been investigated as the cathode of the hybrid solar cell. The photocurrent of the fabricated solar cell was found to vary with pore size of PSi and heat-treatment of the whole structure. As was expected when the pore diameter was comparable to the transport length of photo-generated carriers the short-circuit current density enhanced drastically. The highest enhancement was about 73 times compared to conventional cathode (without porous structure). The experimental investigations indicate lots of promise for porous-cathode in the hybrid solar cells having higher performance.

EXPERIMENTAL

A. ***Wafer cleaning***

There are various techniques of silicon wafer cleaning. The choice of a particular technique depends on the nature of the substrate, the nature of the contamination and the degree of cleanliness required. In this work the silicon wafer ware cleaned as the following steps:

a) Initially the silicon wafers were dipped and vibrated in acetone by Ultrasonic Vibrator for 2-3 min to remove organic residues.
b) Then the wafers were cleaned with de-ionized (DI) water.
c) Again the wafers were dipped and sonicated in acetone by Ultrasonic Vibrator for 2-3 minutes.
d) Then again the wafers were cleaned with de-ionized water.
e) After that the wafers were dipped in 10% HF solution for 30 sec to remove the oxides from the silicon surface.
f) Then the wafers were again cleaned with de-ionized water properly.
   And finally the wafers were dried in hot air.

B. **Macroporous silicon (macro-PSi) preparation**

Porous silicon can be defined as the silicon crystal containing matrix of pores and having high surface area [20-22]. There are three types of pores observed in silicon: (a) Nanopores with pore diameters and pore distances are smaller than 2 nm, b) Mesopores with pore diameters and pore distances are between 2nm and 50nm c) Macropores having geometries of pore diameters are larger than 50nm [23]. In most cases, the PSi is formed by electrochemical etching of Si wafers in electrolytes including hydrofluoric acid (HF) and surfactants (conventionally ethanol) [24]. In this work macro-PSi having two-dimensional periodicity has been fabricated by electrochemical etching of p-type silicon substrate (Boron doped, Resistivity of 1-5 Ω-cm, (100) oriented) having Aluminium (Al) back contact, with 1HF:4DMSO (v/v) solution [25]. The Al back contact was made by electron beam evaporation technique. The DMSO was chosen because it offers a faster etching rate, allows larger range of etching current density and produces wide range of pore diameter [26]. Three different samples were prepared for three different etching times (Sample1: 10min, Sample2: 3min, Sample3: 1min) with constant etching current density of $30mA/cm^2$. After the etching process, the samples were rinsed in ethanol and blown dry in air.

The experimental set-up of electrochemical etching cell is shown in Figure 1. The Si wafer with aluminium back contact acts as the anode and the platinum acts as the cathode. As shown in Figure 1 under this fabrication process the wafer was attached to the side wall instead of conventionally placing it at the bottom [27] of etching chamber. That prevented deposition of the etched Si inside the already formed pores that might have helped produce pores of uniform size. It was observed that the fabricated porous silicon was nearly uniform. The porosity, i.e. the void fraction in the porous layer is well controlled by adjusting the anodization current density, composition electrolyte and concentration of the HF in the solution. The porosity is also highly dependent on the doping density of Si substrate.

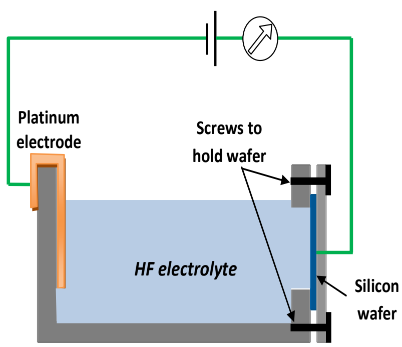

Fig. 1. Cross-sectional view of a two electrode electrochemical etching cell used to fabricate porous silicon.

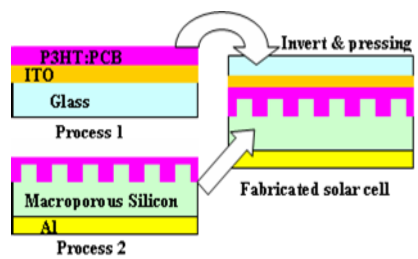

Fig. 2. Fabrication process of hybrid solar cell that uses macro-PSi as the cathode.

C. Device fabrication

A schematic of the fabrication process (with layered structure) of hybrid solar cells (fabricated using blend of P3HT (poly-3-hexylthiophene) and PCBM (Phenyl C61 butyric acid methylester) at 1:1 ration with porous silicon) is shown in Figure 2. The Fabrication of hybrid solar cells starts with an indium tin oxide (ITO) film coated on a glass substrate. Very thin layer of ITO (~ 100 nm) was deposited by electron-beam evaporation process with the chamber pressure of 2.5×10^{-5} mbar, Beam current of 30 mA and voltage of 2KV. After ITO deposition, the samples were annealed in a furnace (Carbolite CWF 12/13) in air at 600°C for 10 min.

The fabrication process begins with "Process-1" where a thin layer of photo-active material (P3HT:PCBM) was deposited onto the ITO coated glass substrate by spin coating process at 500 RPM for 40 sec. In a separate process (Process-2) a thin layer of P3HT:PCBM was deposited onto macro-PSi by drop casting method. Heat treatment of PSi/P3HT:PCBM at 45°C for 5 min was done to liquefy the P3HT:PCBM for penetration into the pores of PSi. Thus prepared PSi sample containing P3HT:PCBM layer was then pasted with the P3HT:PCBM layer on the ITO by pressing the two substrates (PSi and glass). The whole fabrication process is shown schematically in Figure 2. Afterwards the whole cell structure was covered with glass plate and then plastic and brought out connections from anode and cathode by using copper wire with silver paste. Finally, the whole structure [ITO/P3HT:PCBM/PSi/Al] was annealed at 120°C for 20 min to establish good contacts between two different P3HT:PCBM layers and also with PSi.

Electron-beam evaporated indium tin oxide (ITO) and Aluminum (Al) have been used as anode and back-contact of PSi respectively. The active area of our device was 0.7 cm^2. Fet Quanta Inspect S-50 scanning electron microscopy (SEM) imaging was used to study the surface morphology of the PSi samples. Photo-current density versus voltage (J-V) measurement was performed with a Keithley model 2400 source meter and a solar simulator system.

The energy relationship for the fabricated solar cell is shown in Figure 4. The value of work-function of macro-PSi is assumed to be around that of bulk silicon as the position of the conduction band of macro-PSi is not expected to be significantly different from the bulk. The 2eV optical band gap of macro-PSi allows proper bands alignment, carrier separation and transportation to the cathode. Also the dissociation of excitons that are generated under illumination is expected to be enhanced because of the electric field produced due to the different electron affinities and ionization potentials. Therefore according to the energy relationship efficient charge transfer and collection can be expected.

RESULTS AND DISCUSSION

The pore-diameter and nature of porosity is important for the enhancement of the performance of organic solar cell fabricated with the macro-PSi as the cathode. Formation of macro-pore in Si wafer was studied varying the anodizing current density (etching current density) and anodizing time (etching time) by SEM. Figure 4 shows the SEM image of the top of the PSi sample with anodization current density of 30mA/cm^2 and anodization time 10 min. The average pore diameter was about 790 nm and the porosity was about 66%. The pore diameter and porosity were calculated using SEM image processing. Different current densities such as, 10, 20 and 30 mA/cm^2, and etching time of 1, 3, 6, 10 and 15 min were investigated [28].

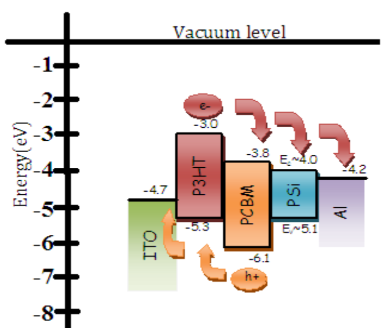

Fig. 3. The expected energy band diagrams of the fabricated hybrid solar cell.

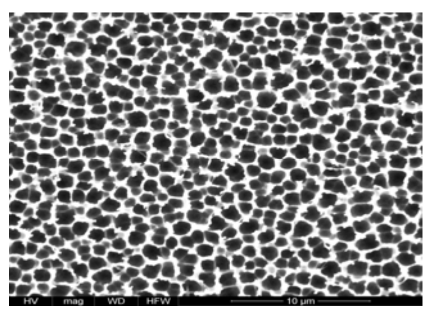

Fig. 4. The SEM image of porous silicon surface.

The pore diameter was found to vary linearly with the etching time upto 10 min. This gives us the tunability of the average pore diameter, which is shown in Figure 5. The pore inititiation time as found from Figure 5 was about 30 sec. This is due to use of DMSO as the electrolyte and high etching current density. DMSO has very good oxidizing property and forms silicon oxide very fast [26]. The similar linear relationship for pore-depth with etching time has been reported [29].

Figure 6 shows the cross-sectional view of the fabricated device at 150 times magnification that shows the various layers of the glass/ITO/P3HT:PCBM/PSi/AL heterostructure. The ITO layer is not clearly visible as the thickness of ITO layer was around 100 nm whereas the other layers (P3HT:PCBM, Al, bulk-Si, etc were in the range of micrometer). The thickness of the glass substrate is about 1mm and the thickness of the bulk-Si is about 260 µm. Figure 7 shows the cross-sectional view of the fabricated device at 500 times magnification, from this figure it is observed that the thickness of the Al is about 60 µm and the thickness of the P3HT:PCBM layer is about 13 µm.

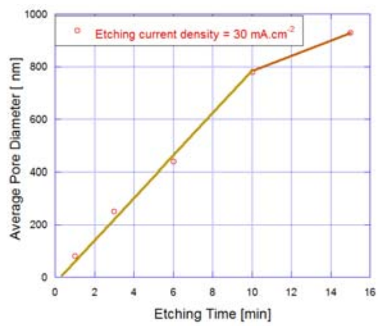

Fig. 5 Tunability of average pore diameter macro-Psi.

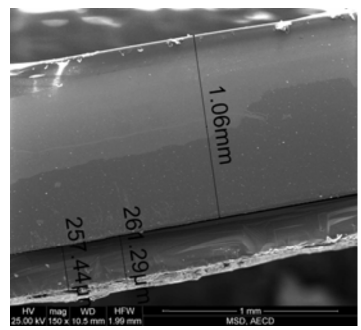

Fig. 6. SEM image of cross-section for full structure with 150 times magnification.

Huge influence of pore diameter on the forward and reverse current density (J) in dark as well as under 1.5 AM simulated light was observed. The J-V characteristic in dark is shown in Figure 8.

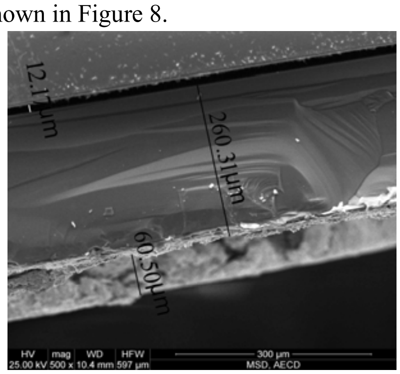

Fig. 8. SEM image of cross-section for full structure with 500 times magnification.

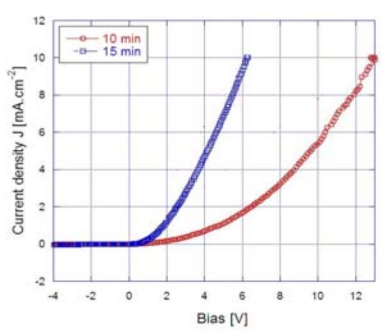

Fig. 7. J-V characteristics (in dark) of fabricated hybrid solar cells.

The dark J-V characteristic of the fabricated hybrid solar cell (Figure 8) shows a typical rectifying junction behavior with threshold voltage of 0.3V. The dark current was found to increase with the increased pore diameter. The diode factor as calculated from the ln(J)-V characteristic (Figure 9) of macro-PSi fabricated by 15 min etching was found to be 4.36. This very high value of diode ideality factor (4.36) indicates strong recombination at the heterojunction interface resulting from the barrier inhomogeneity, series resistance, image force lowering, existence of interfacial layers, recombination effect of charge-carrier drift and diffusion or tunneling currents through the barrier or a combination of these effects [30].

The diode ideality factor was found to increase with decreasing etching time and etching current corresponding to smaller average pore diameter. This increase in ideality factor may be due to improper pore filling and contact with the skeleton of the macro-PSi.

The reverse J-V characteristics under simulated light illumination were investigated to find the influence of pore diameter on the performance of fabricated hybrid solar cell. Figure 10 shows the J-V characteristics under 1.5 AM simulated light illumination for the solar cells fabricated with 1, 3, and 10 min etched porous silicon cathode. The corresponding average pore diameter was 90, 250 and 930 nm respectively. The short circuit current density (JS_C) was very low for larger pore diameter (930nm). The JS_C was also smaller for lower diameter (90nm). When the pore diameter was very high compared to the reported carrier transport length of the acceptor polymer the charge collection was poor. This is because of the recombination of photo generated charges before they are reached at collectors. Again for the smaller pores the pore filling cannot provide a sufficiently dense filling of P3HT:PCBM blend to be able to form a "conductive path" for electron transport to the cathode. But when the pore diameter was 250 nm (3 min etching time with current density of 30 $mA.cm^{-2}$) the observed JS_C was highest.

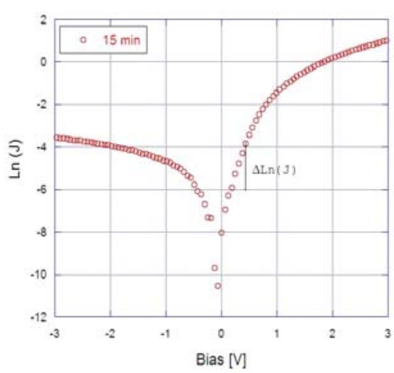

Fig. 10. Diode ideality factor at the low-bias region.

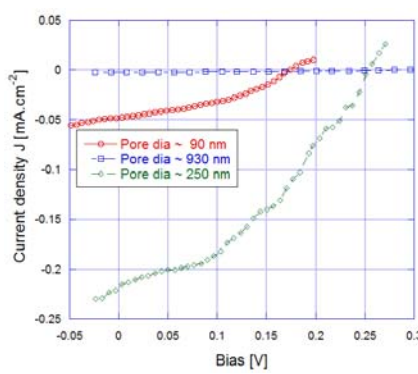

Fig. 9. J-V characteristics under 1.5 AM light of hybrid solar cell with macro-PSi cathode having various pore diameters.

The mentioned enhancement in JS_C is due to enhanced charge collection efficiency, in addition of having antireflection nature of porous silicon. Being situated with in the electron transport distance the cathode was able to collect electrons more efficiently.

Although we did not measure the depth of the etched pore but with the mentioned etching condition we can assume a shallow depth of couple of 100 nm as reported by Bettotti et al [31].

Similarly, assuming a partial filling of the pore by the P3HT:PCBM blend, the dissociated holes are also expected to be collected efficiently by the ITO anode. The blend of P3HT:PCBM formed bulk heterojunctions. The junctions in which the donor and acceptor phases exist as an interpenetrating network is called bulk heterojunction [32]. This hypothesis can be viewed schematically from Figure 11.

The influence of average pore diameter on the JS_C, open-circuit voltage (VO_C) and fill-factor (FF) has been summarized in Table-I. From the Table-I it is seen that the VO_C is decreasing with the smaller pore diameter. This is may be due to the insufficiently dense filling of pores.

TABLE I. INFLUENCE OF AVERAGE PORE DIAMETER ON THE SOLAR CELL PERFORMANCE.

Average pore diameter [nm]	***J_{SC} [µA.cm^{-2}]***	***V_{OC} [V]***	***FF [%]***
930	2.67	0.278	38.5
250	218	0.254	40.1
90	48	0.175	39.7

Heat treatment (annealing) of the whole structure had direct influence on the solar cell parameters. Figure 12 shows the influence of annealing of the whole structure at 120°C for 10 min on the J-V characteristics of the fabricated solar cell. From the Figure 12 it is seen that, by annealing at 120°C for 20 min the open-circuit voltage and short-circuit current density has been increased. The annealing of the fabricated final structure may have established good contacts between P3HT:PCBM and PSi.

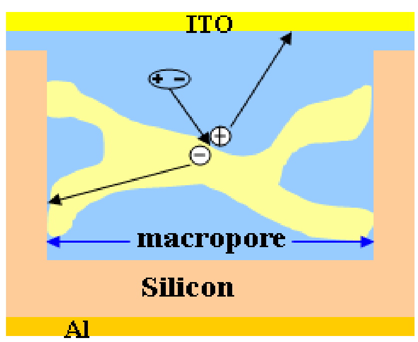

Fig. 12. Schematic of bulk heterojunction hybrid solar cell with macro-PSi as the cathode. The blue region is the P3HT and the yellow region is the PCBM.

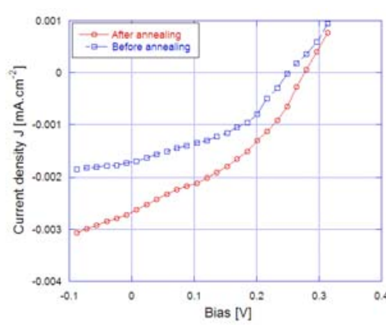

Fig. 11. Comparison the effect of annealing on V-J (Under 1.5AM light) characteristics for constructed hybrid solar-cell.

CONCLUSIONS

Macroporous silicon can be fabricated at low temperature from solution processing without any vacuum equipment or high-temperature processing. Compared to hybrid solar cells reported previously [33–36], the whole fabrication method (in this work) for preparing hybrid solar-cell is simple, cheap and fast. The performance parameters such as, JS_C, VO_C, FF and conversion efficiency was low for our fabricated cells. This we believe was due to fabrication process used to fabricate the solar cell, processing environment, quality of the polymer blend etc. The highest improvement in short-circuit current density was approximately 73 times compared to the normal structure (without PSi). As the mentioned processing factors were common during the fabrication of all the solar cells, the enhancement in performance can be attributed to the incorporation of porous cathode with pore-diameter comparable to photogenerated charge transport length. It ensures a highly efficient charge extraction by macro-PSi, as most of generated charges are very close to the electrodes, giving a high probability of being collected before recombining. Furthermore use of macroporous silicon might have also contributed in reduction of light reflection and increase in light trapping [37]. Altogether, it can be concluded that macroporous silicon having optimized pore-diameter and pore-depth that are comparable to the photogenerated charge transport length can be used in the hybrid solar cells for enhanced performance.

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