ASI

Carbon – Science and Technology

ISSN 0974 – 0546

http://www.applied-science-innovations.com

LETTER Received : 01/11/2007, Accepted : 01/12/2007.

Single Crystals of Fullerene (C_{60}) makes Organic Thick Film Solar Cells and Self Supporting Organic Solar Cells Possible.

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Single crystals of Fullerene (SC-C_{60}) were synthesized by simple liquid/liquid interface precipitation method. Organic thick film solar cell (with an active layer thickness of approximately 20 microns thick) is demonstrated by combining SC-C_{60} with poly(3-octylthiophene). Our preliminary results indicate that organic thick film solar cells are possible; which were considered to be impossible due to low mobility and small exciton diffusion lengths in most of the organic materials including small organic molecules and conjugated conducting polymers. Further, SC-C_{60} seems to be promising materials for organic photovoltaics. Self supporting organic solar cell is also demonstrated using SC-C_{60}.

Organic thick film solar cells were considered to be impossible due to low mobility of charge carriers and small exciton diffusion lengths (typically between 20 nm to 60 nm) in most of the organic materials including small organic molecules and conjugated conducting polymers as compared with their inorganic counterparts. Inorganic thick film solar cells are existing. There are several advantages of thick film based solar cells over thin film ones such as easy fabrication and lower cost and hence are expected to be more suitable for industry. In fact, this is one of the biggest advantage that Gratzel solar cells (Popularly known as dye sensitized solar cells) have (in which the typical porous titania thick films are made using Dr. Blade technique or screen printing technique). In contrast, the organic solar cells demonstrated until now are all thin film devices having typical active layer thickness less than 300 nm. One of the common strategies used to increase the efficiency of such cells is to reduce the active layer thickness (i.e. to make it more thin). Very often, it again gives rise to problems such as pin-holes in the films and shorts. One way to avoid such pin-holes in the thin films and shorts in the devices is to fabricate the devices in clean room or in vacuum conductions or using special environments / conditions, which is again contrary to the main objectives of organic solar cells research. Due to these reasons, organic thick film solar cells which could be fabricated by as simple method as that in Gratzel cells are highly desirable. In the present work undertaken, our aim was to demonstrate an organic thick film solar cell and not towards high efficiency. We believe that once such thick film solar cell is demonstrated, efficiency could be improved in future.

Fullerene (C_{60}) has remained most preferred electron accepting and transporting material in most of the organic solar cells ever since the discovery of ultra fast photoinduced electron transfer from conducting polymer to fullerene (C_{60}) and invention of bulk heterojunction concept [1-3]. Generally, bulk heterojunction donor-acceptor type solar cells are fabricated by casting thin film from a solution containing mixture of conducting polymer and fullerene (C_{60}). Thermal annealing usually results in the useful phase separation of individual component materials with improved device performance. Such phase separation of donor and acceptor materials creates percolative paths for electrons and holes to be transported to the respective electrodes.

Theoretical predictions and experimental findings suggest that the crystals formed from fullerene (C_{60}) are n-type semiconductors with direct band gaps [4]. Closed packed face-centered-cubic (f.c.c) crystal structure of fullerene (C_{60}) molecules results in high electron mobility (~ 0.1 cm^{-2} V^{-1} s^{-1}) [5], which makes them attractive n-type materials for organic solar cell applications. In past, fullerene (C_{60}) fine particles were used in photoelectrochemical (PEC) solar cells by Kamat and co-workers. However, such solar cells are wet state solar cells (using liquid electrolyte) [6-13]. In the present work, we demonstrate a bulk heterojunction donor-acceptor type 3D solar cell using single crystals of fullerene (SC-C_{60}) as

electron acceptor and regioregular poly(3-octylthiophene) (P3OT) as electron donor. Our device is a solid state device. SC-C_{60} were synthesized from simple liquid/liquid interface precipitation method. We report open circuit voltage, short-circuit current density, fill factor and light conversion efficiency of about 0.153 V, 0.0148 mA/cm^2, 0.234 and 0.0005 %, respectively, for a device with 20 microns thick photoactive layer (P3OT+SC-C_{60}). Device is un-optimized for its performance and is fabricated by simple drop casting technique. Our preliminary results indicate that : (1) Organic thick film solar cells are possible. (2) SC-C_{60} are excellent electron acceptors and transporters with potential applications in organic solar cells and (3) Self supporting organic thick film (flexible) solar cells are possible.

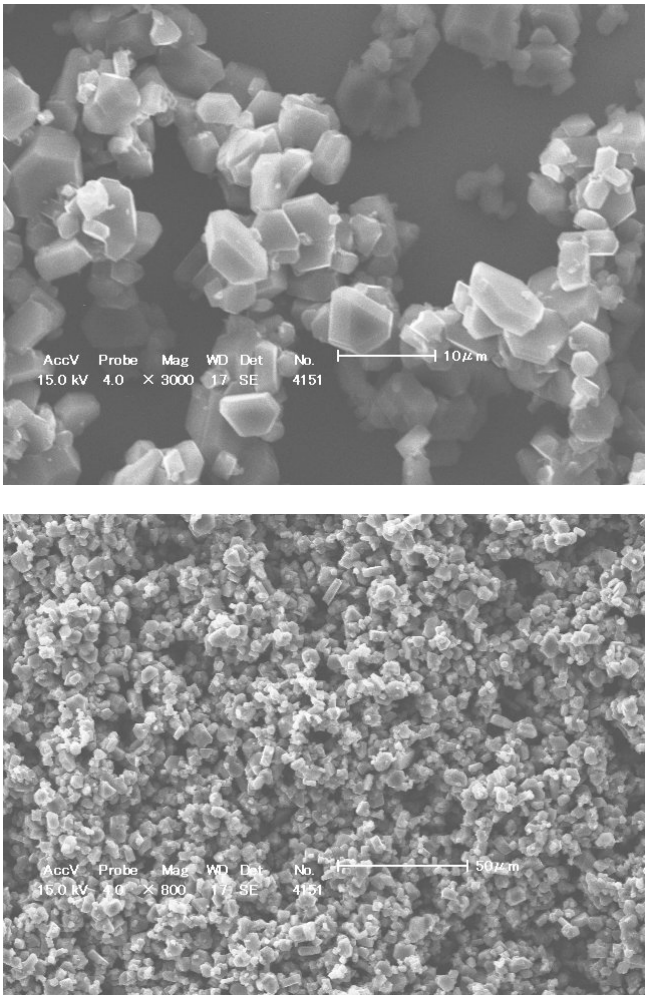

Figure (1) : SEM of the SC-C_{60} (a) and the porous thick film deposited using SC-C_{60} by drop casting method (b).

SC-C_{60} were synthesized by liquid/liquid interface precipitation method. Our method is a modified version of the method reported by Jin et. al [5] for synthesis of fullerene (C_{60}) single crystal nanorods. Fullerene (C_{60}) was purchased from Frontier Carbon Corporation, Japan and was used as received. In a typical experiment, 2.0 ml 0.30 wt. % C_{60}-toluene solution was dropped into 8.0 ml isopropyl alcohol (IPA) at room temperature (25 ^{o}C) using a syring pump. Addition was done drop by drop with stirring. The liquid instantly turned brown on addition of C_{60}-toluene to IPA. The suspension was allowed to settle and concentrated. This suspension contained SC-C_{60} which were used without further separation and purification steps. Bulk heterojunction donor-acceptor type 3D solar cells containing SC-C_{60} were fabricated by the following method. Highly doped n-Si substrates (2 X 2 cm^2) were cleaned in acetone and methanol by ultrasonication. Appropriately diluted HF treatment was given to n-Si substrates so as to remove the native oxide layer. A thick film of SC-C_{60} was deposited on n-Si by drop casting technique followed by drying in argon atmosphere at an appropriate temperature. The thick films of SC-C_{60} were observed to be significantly porous in nature – as indicated by scanning electron microscope (SEM). Such films were infiltrated with P3OT using a solution of P3OT (Aldrich, 8.5 mg/ml) in chloroform. SC-C_{60} films on n-Si were slightly heated (at approximately 100 ^{o}C) and P3OT is infiltrated in them in such hot condition in inert gas atmosphere. P3OT is observed to get solidify quickly and the chloroform solvent do not dissolves the SC-C_{60}. Films were dried in argon atmosphere so as to remove the traces of solvent, if any. Semitransparent thin film of gold (25 nm) was deposited on top so as to complete the cell configuration (i.e. n-Si/SC-C_{60}+P3OT/Au). Current-voltage characteristics were measured in dark and under AM 1.5 simulated solar radiation using JASCO SS-200 W solar simulator. n-Si makes direct contact with the conducting stage which was made of stainless steel. Cells were illuminated from the semitransparent gold electrode side. Thickness of the active layer (i.e. SC-C_{60}+P3OT) was measured by taking the cross section of the cell and observing by SEM.

Figure (1a) shows the SEM micrograph of the SC-C_{60}. Many nano-sized crystals of fullerene (C_{60}) with different shapes and size are observable. Most of these crystals have size ranging between 7 – 8 microns. However, crystals with larger or smaller size are occasionally observed. From the SEM micrographs, the crystals seem to be well separated and do not show aggregation. Disc like hexagonal crystals are also observed. TEM observations and the fast-fourier-transform (FFT) patterns indicate that they are F. C. C. crystals. Figure (1b) shows typical SEM micrograph of the SC-C_{60} thick film deposited by drop-casting method on n-Si substrate. Porous nature of the film is observable.

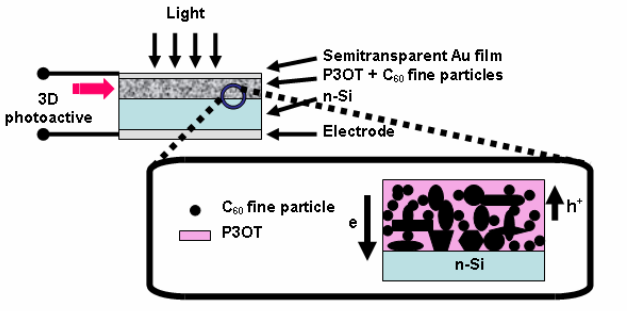

Figure (2) : Schematic of the SC-C_{60}-P3OT based solar Cell. Inset shows an artists view of the internal structure.

Figure (2) shows the schematic of the bulk heterojunction donor-acceptor type 3D organic thick film solar cell comprising the photoactive layer made from the

mixture of P3OT and SC-C_{60}. Inset shows an artist view of the bulk heterojunction formed between P3OT and SC-C_{60}. Thickness of the photoactive layer (SC-C_{60}+P3OT) was estimated to be approximately 20 microns by SEM.

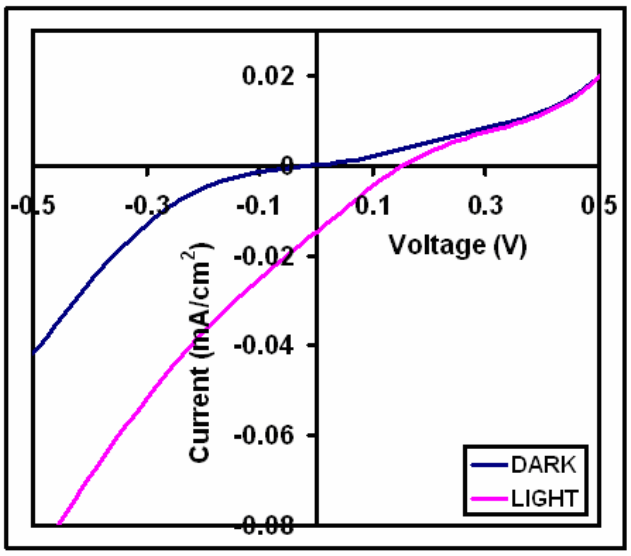

Figure (3) : Current-Voltage (I-V) characteristic of the solar cell in dark and under AM 1.5 simulated solar radiation. Cell is illuminated from the semitransparent Au electrode side.

Figure (3) shows the current-voltage (I-V) characteristics in dark and under AM 1.5 simulated solar radiation of the cell. Cell is illuminated from the semi-transparent gold electrode side. The open circuit voltage (V_{oc}), short circuit current density (I_{sc}), fill factor (FF) and light conversion efficiency (% η) observed are about 0.153 V, 0.0148 mA/cm^2, 0.234 and 0.0005 %, respectively. Photovoltaic action observed in such cells is solely due to the photoactive layer (i.e. SC-C_{60}+P3OT). Experimentally, it has been observed that the amount of light that might reach n-Si surface is almost nil (optical absorption measurements – not shown here). Hence, there is no contribution of n-Si in the photovoltaic effect observed in such cells. Role of n-Si is to collect the electrons given by SC-C_{60}. SC-C_{60} transports the electrons which are in turn donated by P3OT as a result of photo-induced electron transfer. P3OT acts as a donor while SC-C_{60} as acceptor. As stated earlier, except for the efficient electron collection and their transport to the electrode, n-Si plays no active role in the photovoltaic effect observed in these cells. Other suitable substrates can also be used. Lower efficiency observed in the present work is due to relatively thick photoactive layer. Device is un-optimized for its performance and there is ample scope for its improvement, particularly by optimizing the photoactive layer thickness. Further, the photoactive layer (SC-C_{60}+P3OT/Au) was easily peeled-off from the silicon substrate and pasted on other substrate (stainless steel). It has been observed that the device still works. This confirms that the photovoltaic action is from the composite layer and not from the silicon substrate used. Further, this indicates that self supporting organic solar cells are possible. It is to be noted here that all the organic solar cells demonstrated until now uses either glass or plastic substrates (coated with transparent conducting coatings). Our preliminary results indicate that such self supporting organic solar cells might also be flexible. In past, we have demonstrated similar organic thick film solar cell using Fullerene single crystal nanorods which also can be made self supporting in a similar way [14].

In all, we demonstrate an organic thick film solar cell and self supporting (flexible) organic thick film solar cell using SC-C_{60} as electron acceptor and P3OT as donor. The present demonstrated cell is bulk heterojunction donor-acceptor type 3D solar cell. Technique of synthesis and fabrication of SC-C_{60} and the present reported solar cell are simple and can easily be scaled up. Our initial results indicate that SC-C_{60} are excellent electron acceptors and promising materials for application in organic solar cells. Organic thick film solar cells are possible and might be more friendly with industry. We speculate that combination of carbon nanotubes, SC-C_{60} and conducting polymers may leads to efficiency organic thick film solar cells.

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