

The effect of MWCNT-PEDOT:PSS layer in organic photovoltaic fiber device

I. BORAZAN*, A. BEDELOGLU^a, A. DEMIR

Department of Textile Engineering, Faculty of Textile Technologies and Design, Istanbul Technical University, Gumussuyu, 34437, Turkey

^aDepartment of Fiber and Polymer Engineering, Natural Sciences, Architecture and Engineering Faculty, Bursa Technical University 16190, Bursa, Turkey

In recent years, studies, investigations to develop photovoltaic fibers have increased, since fiber shaped and flexible photovoltaics present possible advantages for wide variety of applications. This paper presents the development of indium-tin-oxide (ITO)-free photovoltaic fiber using polymer-based organic materials. Non-transparent and non-conductive conventional textile fiber was employed as flexible substrate. Detailed investigation for photoelectrical characterization and surface morphology was performed. MWCNT added conductive polymer-based electrodes definitely improved the power conversion efficiency of photovoltaic fibers.

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1. Introduction

Today, energy is a crucial issue in both daily and industrial life related to countries' economic policies. Renewable energy resources attract great attention due to concerns about global warming and its detrimental effects on earth and living organisms, and decreasing level of fossil based energy resources, in recent years. Therefore, researchers began to investigate and develop technologies to use renewable energy sources. Solar energy, which is abundant, free and clean, possesses a unique place among the other renewable energies [1].

Solar cells are the best way to harvest solar energy by converting sunlight directly into electrical energy. Polymer-based organic solar cells have many advantages such as being ease of processing and large scale applicability compared to silicon-based solar cells. The polymer-based organic solar cells consist of blends of different materials which are responsible for generating electron-hole pairs under illumination. Fabrication of organic solar cells is performed using several techniques such as thermal deposition, solution coating, printing, electrospinning and etc. [2]. A conventional organic solar cell structure (see Fig. 1) consists of the following layers: a transparent bottom electrode like ITO, a light absorbing photoactive layer sandwiched between two electrodes, and an upper metal electrode like Al which is about 100 nm. A PEDOT:PSS conductive polymer is used as a hole injection layer between ITO and the light absorbing layer [3]. Other great benefits of organic solar cell devices can be considered being flexible, lightweight and semi-transparent. Generally, conventional organic solar cells are constructed on rigid, heavy, brittle substrates like glass. Many indium-free materials are investigated to substitute indium tin oxide which is the high-cost and brittle

transparent electrode. The recent studies are focused on indium-free transparent conducting oxides, conducting polymers, thin metal layers, silver nanowires, carbon nanotubes, and graphenes [4-17]. In many studies, thin metal layers were also investigated as semi-transparent top electrodes [8-9, 18-25] for top illuminated devices. Carbon nanotubes (CNT) have attracted great attention due to their flexibility, chemical stability, cheap and continuous production methods, high conductivity and transparency advantages so that they are used in organic light emitting diodes and organic solar cells as an electrode [26-30]. Since multi walled carbon nanotubes (MWCNT) has a work function of 4,9 eV [31] which is appropriate to substitute ITO in solar cells, MWCNT was chosen as electrode in this study about development of photovoltaic fiber device.

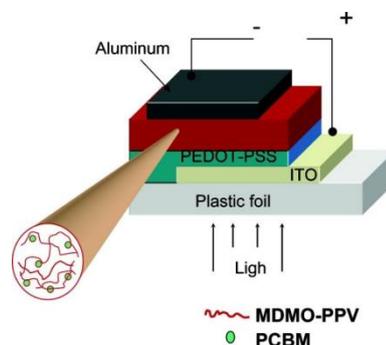


Fig. 1. Bulk heterojunction configuration in organic solar cells © [Rightsholder]. [Reprinted with permission from { Serap Günes, Helmut Neugebauer, and, and Niyazi Serdar Sariciftci, *Conjugated Polymer-Based Organic Solar Cells*, Chem. Rev., 2007, 107 (4), pp 1324– 1338}. Copyright {2013} American Chemical Society [3].

In last few years, polymer based substrates such as foils, wires, fibers are considered to develop portable, flexible, scalable and lightweight photovoltaics [1]. Besides, photovoltaic fibers (Fig. 2) which are capable to be converted into fabric form are promising candidates for generating electricity everywhere. Photovoltaic fibers, and fabrics produced from these fibers and garments will generate electricity for electrical devices requiring small amount of power in primarily use. Such photovoltaic textiles will be useful for especially far from grids, military applications, camping etc. They have several advantages such as lightweight, more flexible, graded transparency.

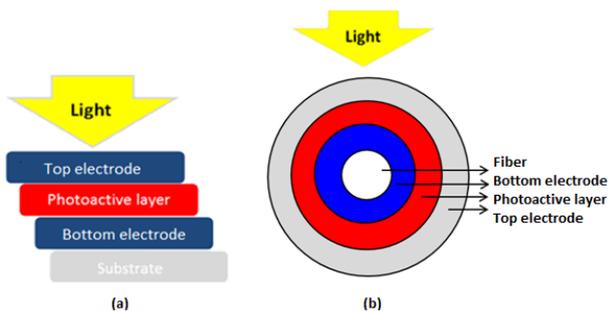


Fig. 2. Schematic demonstration of fabricated photovoltaic fibers and reference solar cells while measuring photoelectrical performances.

There are some studies about developing photovoltaic fibers using organic solar cell materials. Different substrates including PP fibers [18-22], SS wires [32], optical fibers [33] were used for fiber shaped photovoltaic devices. In 2003, Forrest et al. patented a photovoltaic fiber which has a core fiber surrounded by primary electrode, transparent secondary electrode enclosing the photoactive layer and having an electrical contact with this layer. A counter electrode has a contact with transparent electrode and a protective layer on the outer surface is encapsulating the structure [34]. Liu et al., in 2007, coated organic photoactive materials on optical fibers with a diameter of 0.6-1.4 mm and obtained 0.6-1% efficiency, respectively. This photovoltaic fiber was illuminated from inside by the help of optical fiber in the core [34]. Lee et al. developed wire-based photovoltaic device of which efficiency values ranged from 2.79% to 3.27% by eliminating the use of transparent oxide semiconductors [34]. Bedeloglu et al, developed a flexible and ITO-free photovoltaic device with a non-transparent and non-conductive PP tapes and fiber substrates. The device structures were appropriate to be illuminated from outer surface having a very thin semi-transparent LiF/Al and Al/Ag top electrode combinations [24,25,32,34]. Another approach used stainless steel wire and carbon nanotubes as primary and counter electrodes in wire based photovoltaic fiber, respectively. High power conversion efficiencies as 2.31% and 2.11% were obtained from carbon nanotubes applied as a thin layer and a yarn, respectively [35].

As comparing planar solar cells, controlling the coating or deposition process of solutions of polymer and other materials or adjusting the twisting process of counter electrode on fiber or wire surfaces is very difficult. Optical fibers are not appropriate to be used in textile applications, as they are illuminated from inner surface. Utilizing a metal wire electrode, which is quite rigid and limits the flexibility of device, could easily damage the thin polymer-based layers. Twisting a counter electrode on fiber device very closely is also difficult process which could cause a short circuit. Therefore, the choice of substrate, electrode materials and device design is very important for developing a practical photovoltaic fiber used in future possible applications. Polymer-based materials are compatible with the manufacturing processes and functionality of photovoltaic fibers, and the textile fibers are the most suitable materials for being the substrates of photovoltaic fiber devices. Polyamide fibre (Figure 3), which is very durable, light and fine, and also easy to wash and quick-drying with shape retention and can be processed by thermoplastic methods, was chosen. It has some advantages including high elasticity, low coefficient of friction, good abrasion, temperature and solvent resistance and etc. It has such an excellent energy absorbing properties that it is used in production of climbing ropes, parachute fabrics and spinnaker sails etc. [36].

The aim of this study is to develop flexible, thin and lightweight polymer-based organic photovoltaic fibers using MWCNT-polymer-based conductive layer. Well-known conventional fabrication and measurement techniques for solar cells were modified as previously mentioned [18] to apply on fiber structure. A thin Al metal was chosen as semi-transparent top electrode to take the sun light from outer surface into the device. The effect of MWCNT with conductive polymer layer in photovoltaic devices to substitute ITO was investigated in detail.

2. Experimental

2.1. Materials

The substrate for PV fiber, which was a polyamide (PA) monofilament, non-transparent and non-conductive, with 0.9 mm diameter, was employed to carry photovoltaic structure. Therefore, due to its features, for substrate polyamide monofilament was chosen and cut in 25 mm length. Then, the fibers gently cleaned from any industrial dirt in distilled water, acetone and isopropanol, respectively and dried with nitrogen flow. Highly conductive PEDOT:PSS (purchased from Sigma-Aldrich) solution which gives the best conductivity with a mixture of Dimethylsulfoxide (DMSO) (purchased from Sigma-Aldrich) (v:v, 95:5) [19] was prepared to obtain polymer-based electrode (anode) on fiber substrates. A small amount of non-ionic surfactant (Triton X-100, purchased from Sigma-Aldrich) was added into PEDOT:PSS solution to be able to coat it onto textile surface, and stirred at least 4 hours on magnetic stirrer. A MWCNT (purchased from

Sigma-Aldrich) solution were prepared according to a study in the literature [37]. MWCNT were dispersed in DMSO (0,1 mg/ml) and bath sonicated for 2 h, and then centrifuged at 9000 rpm for 30 min, centrifuge action was repeated. The supernatant of MWCNT-DMSO solution was mixed with PEDOT:PSS (9:1, v/v) and bath sonicated for 30 min. A blend of nano materials which consist of P3HT (Sigma-Aldrich) (electron donor and p-type semiconductor) and PCBM (electron acceptor and n-type semiconductor) materials with a weight ratio of 5:4 dissolved in chlorobenzene (Sigma-Aldrich) and stirred 24 h on magnetic stirrer at room temperature. Chemical structures of PEDOT:PSS, P3HT, PCBM and MWCNT are shown in Fig. 3(a-d), respectively.

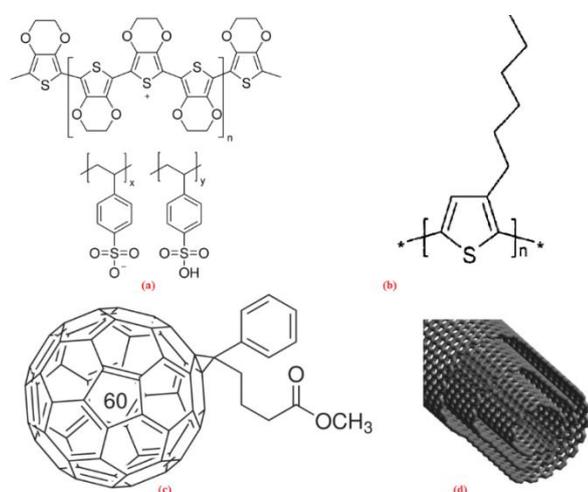


Fig. 3. Chemical structures of PEDOT:PSS (a), P3HT (b), and PCBM (c) [15], and MWCNT (d) [38].

2.2 Preparation of photovoltaic fibers and reference cells

For preparation of photovoltaic fiber devices, MWCNT- PEDOT:PSS mixture was dip-coated on a group of the polyamide fibers which are cleaned in isopropanol and distilled water, respectively, and then heating process was applied in hanging position at 50 °C for 30 min. PEDOT:PSS was deposited on both naked polyamide fibers and MWCNT- PEDOT:PSS-coated polyamide fibers by dipping and coated fibers were dried in hanged position in an oven at 50 °C for 30 min., again. Afterwards, P3HT:PCBM solution was dip-coated on polymer anode layer and subsequently heated at 50 °C for 30 min. For the last step of fabrication of a photovoltaic fiber, thin Al metal layer was deposited by using a mask in thermal evaporation machine in vacuum conditions of about 10^{-6} mbar. The mask with rectangular shape was used for fiber-based solar cells. For the fiber-based devices, evaporation was carried out half of the circular structure (180 degree radially) (Fig. 3-4). Then, a small drop of silver paste was applied onto the electrodes on the

photovoltaic structure in order to improve conductivity properties of the contacts.

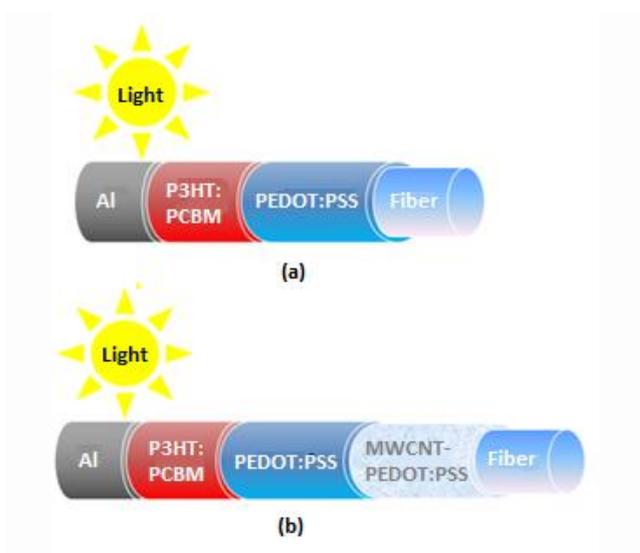


Fig. 4. Schematic representation of photovoltaic devices produced.

2.3 Characterization of the photovoltaic fibers

The photoelectrical characterization of the devices was performed by measuring current (I) and voltage (V) parameters using a sourcemeter (Keithley 2400), in the dark and under the illumination with a calibrated solar simulator (Lot Oriel) having AM1.5 global solar conditions at an intensity of 100 mW/cm² [ASTM standards, E892]. In this study, the illumination was provided through the semi-transparent cathode to compare results in the same conditions of all fabricated devices. Active area for fibers were 0.94 mm².

Performances of solar cells are calculated by using Equation 1 [39] given below:

$$\eta = \frac{P_{out}}{P_{in}} = \frac{J_{sc} \times V_{oc} \times FF}{P_{in}} \quad (1)$$

where P_{out} is the output electrical power of the device under illumination, J_{sc} (Ampere/meter², A/m²) is the short-circuit current density, V_{oc} (Volt, V) is the open-circuit voltage, FF is the fill factor, P_{in} (Watt/meter², W/m²) is the light intensity incident on the device as measured by a calibrated reference cell and η (%) is the power conversion efficiency of the photovoltaic devices measured with a solar simulator.

Thickness of each layers were evaluated by using a profilometer (Tencor Alpha Step 500). Light transmittance and absorption spectra (for light absorbing layer) were provided by a Spectrophotometer (PE Lambda 900 UV (Ultraviolet)). P3HT:PCBM blend in chlorobenzene solution and PEDOT:PSS solution were spin coated and Al was thermally evaporated in vacuum onto microscope

glass slides for these measurements. Surface studies of layers were performed by atomic force microscopy (AFM, Quesant Q-Scope) and scanning electron microscopy (SEM, Zeiss Evo MA10).

3. Results

The obtained photovoltaic devices characteristics for different configurations are summarized in Table 1 and I-V graphs of manufactured photovoltaic devices are presented in Fig. 5. 10 samples were studied and results are averaged. The efficiency of photovoltaic devices is strongly dependent on conditions in which measurements are performed. Therefore, standard conditions in terms of incident light intensity, cell temperature and solar spectrum, are specified to perform reproducible measurement way for the solar cells [36]. Thin metal cathode (ca.15 nm) of which evaporation is very related to machine limitations and influence the device efficiency caused moderate efficiencies in all photovoltaic devices. Photovoltaic fibers gave high photoelectrical results in the present experiments; this situation is most probably due to efficient coating methods used. Use of a MWCNT added conductive polymer-based electrode as the bottom electrode of photovoltaic fiber definitely improved the short circuit current density and fill factor of photovoltaic fibers resulting higher power conversion efficiency.

Table 1. Photoelectrical characteristics of manufactured photovoltaic devices.

Construction	V_{oc} (V)	J_{sc} (mA/c m^2)	FF (%)	η (%)
Fiber/ PEDOT:PSS /P3HT:PCBM/Al	0.784	0.436	83	0.3
Fiber/MWCNT:PEDOT:PSS/ PEDOT:PSS/P3HT:PCBM/Al	0.582	1	83	0.48

The absorption spectra for thin films of P3HT:PCBM (after annealing), Al on glass and reference glass samples in the spectrum (280-1000) are presented in Fig. 6. When solar irradiation is absorbed efficiently, solar cells can work more efficiently. The absorption edge of donor material (semi crystalline regioregular poly(3-hexylthiophene) (P3HT)), which is around 650 nm in P3HT:PCBM solution with optimized blend ratio (5:4), covers the sun's maximum photon flux. In polymer-based organic solar cell, light coming into device generates electron-hole pairs and then, they split into free carriers at the interface between the donor and the acceptor material to go to the selective electrodes. Therefore, the absorption of light in the photoactive layer can be improved by increasing the active layer thickness. However, increasing the active layer thickness causes a decrease in electrical

performance since the distance, which charges will move to be extracted, and thus, recombination possibility will increase. In the spectra, P3HT:PCBM blend shows an efficient absorption at near UV part and the longest absorption wavelength lays around 700 nm after annealing. The broader absorption spectrum of the solar cell is beneficial for the short-circuit current since the solar cell absorbs more photon in the solar spectrum. Annealing process is required and improves the open-circuit voltage V_{oc} , crystallinity and length of P3HT nanowires and also FF in polymer-based organic solar cells. Generally, open-circuit voltage V_{oc} is considered as a characteristic of a solar cell which is independent from light absorbing layer morphology [40-44]. Al is a common material used for top electrode and appropriate material to avoid short circuiting in organic solar cells. However, Al metal layer has the highest absorbance and also, transmission of Al layer is below 10% which is lower than results of our previous studies based on LiF/Al (ca.10 nm) [25]. Therefore, most of the coming light is absorbed mainly in semi-transparent metal cathode and moderate efficiencies were obtained since transmitted light through cell is too low to generate charges. Al has a low transmittance so it is not appropriate for top electrode but it should be substituted with another material.

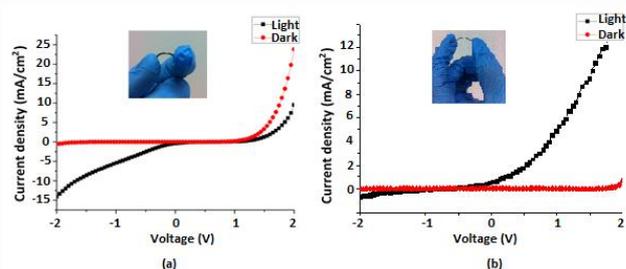


Fig. 5. I-V graphs of devices on glass (a), PET foil (b), fiber (c)-based substrates and fiber (c)-based substrates with MWCNT-PEDOT:PSS electrode.

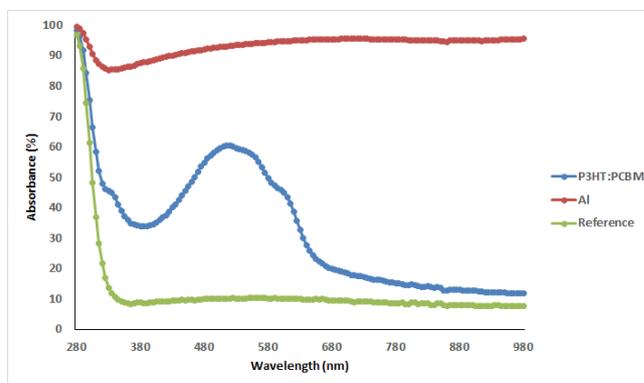


Fig. 6. Absorption spectra for P3HT:PCBM and Al layers and glass.

AFM study for photoactive (P3HT:PCBM) and PEDOT:PSS layers coated on glass was performed to determine surface characteristics of fabricated photovoltaic structures and presented in Fig. 7a and b, respectively. SEM images of MWCNT-PEDOT:PSS coated surfaces are also given in Fig. 8. Surface smoothness is a very important parameter since it affects the quality of solar cell and photoelectrical characteristics. Dipping procedure smoothens the defects on the rough surface of fibers and increases the efficiency of photovoltaic fibers [25]. Environmental parameters, especially dusts in the air, have also significant effect on manufacturing the solar cells since it causes short circuiting due to its micrometer size. The defects on PEDOT:PSS layer seen from AFM images of samples, are the possible reason for low photovoltaic performance. As can be seen from Fig. 8, CNTs with PEDOT:PSS are coated uniformly separated onto surfaces.

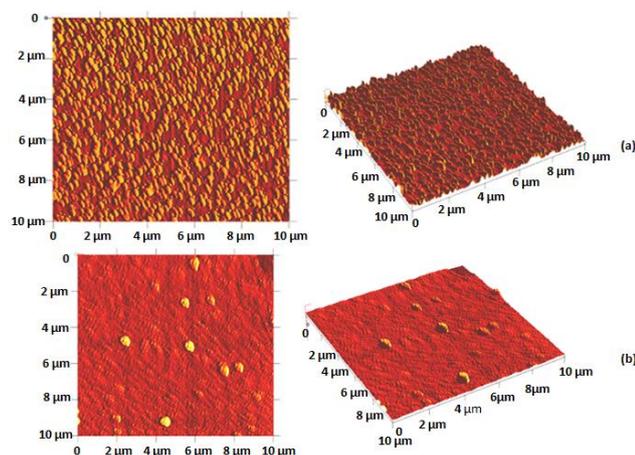


Fig. 7. AFM images of P3HT:PCBM (a) and PEDOT:PSS (b) layers.

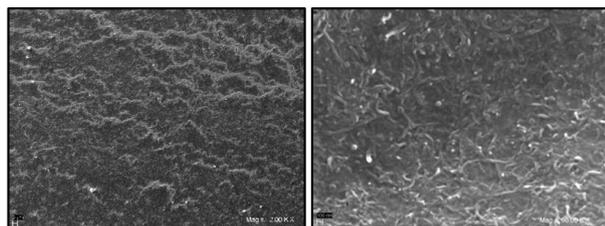


Fig. 8. SEM images of samples coated with CNT/PEDOT:PSS mixture on glass (left) and fiber (right).

4. Discussion

In this paper, possible reproducible manufacturing and measurement techniques for ITO-free photovoltaic fiber using highly conductive MWCNT/PEDOT:PSS electrodes were presented. MWCNT-PEDOT:PSS layer increased the conductivity of the bottom electrode resulting higher

efficiency. Obtained photovoltaic fibers are flexible, lightweight and thin enough to be used in possible different textile structures. In order to prevent photovoltaic fiber structure from short circuiting, Al thin metal layer was used as semi-transparent top electrode. However, in further studies, to improve the power conversion efficiency, novel materials and top electrodes will be investigated since transmitted light through cell is not satisfied in the case of Al electrode. Produced flexible photovoltaic fibers may be woven, knitted or integrated into textiles in order to give photovoltaic effect in conventional textile-like garments to use in off-grid places for electricity need, after further improvements in the future works.

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*Corresponding author: iborazan@itu.edu.tr