

Organic Solar Cells - An Energy Source of the Future: A Review

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Abstract - Theoretically, solar energy is the only renewable source of energy, which has the potential to meet the projected enormous energy demand after a few decades. Although the photovoltaic technology has allowed us to harness the ubiquitous solar energy, high cost, complex manufacturing and scarcity of raw materials required have been deterring its universal growth. A new research area in the field of solar energy emerged in the year 1978 following a discovery that conductivity of conjugated polymers can be controlled to imitate the functions of a semiconductor, when doped with iodine. Over the years, this concept has been used to develop Organic Photovoltaics (OPV) or Organic Solar Cells (OSC) with a spate of combinations of polymers and other transparent conductors. Though easy manufacturing, flexibility and printability have been their selling points, their application has remained elusive; for their power conversion efficiency and stability has been quite subpar compared to their inorganic counterparts. This review paper describes the origin, working, structure, challenges, developments, and application of carbon nanotubes in OPVs. An attempt is also made to give a critical view on the future and applications of this emerging technology, which is still in its inception.

Keywords: Organic solar cells, plastic photovoltaics

I. HISTORY

The first research on organic photovoltaic dates back to 1959; much before the Noble prize winning discovery that conjugated polymers doped with iodine behave like semiconductors, was made by Alan J. Heeger, Alan MacDiarmid and Hideki Shirakawa in the year 1978 [1]. Kallman and Pope observed a photovoltaic effect when light was illuminated on one side of a single crystal of anthracene, sandwiched between two identical electrodes [2]. However, it could produce only 0.2 V open-circuit voltage with power conversion efficiency of just 2×10^{-4} %. They postulated that different exciton dissociation mechanisms must occur at the two electrodes. Twenty-seven years later, in 1986 Tang achieved a major breakthrough in his work, which introduced the concept of

donor-acceptor bilayer configuration [3]. The bi-layer configuration helped dissociation of a larger number of excitons into free charge carriers. Later with the advent of Bulk Heterojunction (BHJ) configuration in 1995 [4], power conversion efficiencies (PCE) of up to 4-5% were reported. Another configuration called Interdigitated Bulk Heterojunction, proposed by Guangyong Li et al [5] in 2011 promised a PCE of anything more than 12% in a simulation study. In the year 2013, a device with 10.1% PCE was developed using solution processed small-molecular materials [6]. Till date, advanced Organic Photovoltaics (OPV) have exhibited PCE of about 10-11% compared to 25-30% efficiency of conventional inorganic solar cells. A number of studies conducted by Alan MacDiarmid et al. in the early twenty-first century lead to the discovery that different polymers can be modified to make them electrically conductive. These discoveries gave a further fillip to the research in this budding technology.

II. OPERATION

Just like regular solar cells, polymer solar cells also convert light into electricity by converting a flux of light particles (or photons) into a flux of charged particles (or current). Different polymer and non-polymer materials with distinct electrical and optical characteristics are employed to make it possible.

The working of an Organic Solar cell (OSC) consists of the following four steps (Fig. 1) [7] [8] [9] [10]:

1. Absorption of light

Organic semiconductors forming the active layer are responsible for the absorption of light from the visible solar radiation. Those materials ought to have certain band gap energy (energetic separation between the valence electrons and the nearest free state) in their molecular structure.

2. Creation of exciton

Because of photo-excitation, the electrons in the valence band - Highest Occupied Molecular Orbital (HOMO) excite to a higher band - Lowest Unoccupied Molecular Orbital (LUMO), thus forming a quasi-particle of an electron-hole pair called exciton.

$$E_{\text{photon}} = \frac{c \cdot h}{\lambda_{\text{photon}}} \geq E_g \quad (1.1)$$

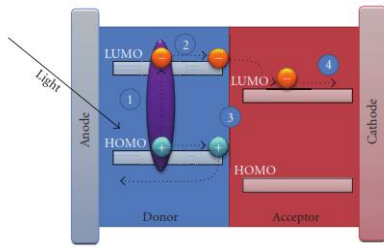


Fig. 1 Working of an OSC
(source: Intl. Journal of Photoenergy)

Where, E_{photon} is energy of the photon, c is speed of light, h is Planck's constant, and E_g is the band gap energy. Since E_{photon} is higher than E_g , the excess energy ($E_{\text{photon}} - E_g$) is wasted as heat (called as relaxation heat).

3. Diffusion and separation of excitons

To generate a current, the exciton must be broken and separated by overcoming its coulomb forces. This is done by introducing a secondary organic semiconductor, called acceptor, with lower energy LUMO level, so that the electron is effectively transferred.

For electron transfer, $E_{\text{donor, LUMO}} - E_{\text{acceptor, LUMO}} \geq E_{\text{ex-b}}$

For hole transfer, $E_{\text{donor, HOMO}} - E_{\text{acceptor, HOMO}} \geq E_{\text{ex-b}}$

Where, $E_{\text{ex-b}}$ is the exciton bond energy

4. Charge collection

The final step involves collection of electrons and holes at the opposite electrodes (i.e. holes at cathode and electrons at anode) and their recombination after being put to work in an external circuit.

III. STRUCTURE AND MATERIALS

Materials used in an OPV are generally prepared using polymers dissolved in organic solvents. These materials are arranged in layers in a certain way to form a solar cell device (Fig. 2). The layers constituting an OPV are: **active layer**, where photons are absorbed and electrons and holes are released by photoelectric effect; **selective charge transport layers**, which restricts electrons/holes from entering wrong electrode; **electrodes**, for extracting the charges; and a **substrate**, to support the solar cell stack.

In a single material active layer, a majority of excitons do not remain intact for long enough to reach an electrode and dissociate. By introducing a secondary semiconductor called the acceptor, with a lower lying LUMO level, the electrons from the original material called donor, can be favourably transferred [3]. This is called a bilayer configuration of the active layer (Fig. 3a). Since an excited electron has a very limited lifetime and tends to pair with the hole, the distance between the donor-acceptor interface must be of the order 5-10nm [11]. But for the excitation to occur, the photon needs to traverse about 100nm. Hence, a

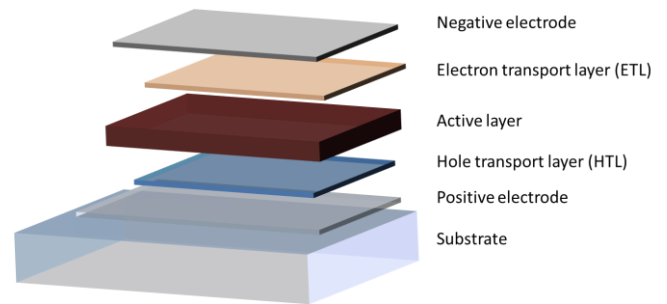


Fig. 2 Structure of a Normal geometry OPV
(source: plasticphotovoltaic.org)

simple bilayer structure is not optimal. The solution to this restriction is a 3-Dimensional nanoscale phase separation of the donor-acceptor (D/A) interface (Fig 3b). The closest configuration that could be achieved conventionally is called the Bulk Heterojunction (BHJ), where, an interpenetrating network morphology at nanoscale is obtained by dissolving and mixing donor and acceptor materials in a common solvent (Fig. 3c) [12] [13] [14] [15]. Such an arrangement effectively increases the D/A interface area, thus raising the number of electron-hole separations. Hence, a higher efficiency of 4-5% has been reported [16].

Poly(3-hexylthiophene), also called P3HT, is a popular material used in organic solar cells. It is a conjugated polymer (polymer with alternating single and double bonds in its backbone), where one electron per carbon atom is left unbounded in p_z orbital. The mutual overlap among these p_z orbitals results in the formation of π bonds along the conjugated backbone, thereby delocalizing the π electrons along the entire conjugation path. The delocalized π electrons fill up the whole band and therefore behave like semiconductors. P3HT can be synthesized through the methods Stille cross-coupling [17], Rieck [18], Direct arylation polymerization [19], etc.

Other important polymers used as donors in the active layer are low band gap polymers obtained by copolymerization of the monomers benzothiadiazole and pyridalthiadiazole with carbazole or cyclopentadithiophene-type units. Some examples of low band gap polymers are PSBTBT, PTB7, PDPP3T, PDTSTTz.

Fullerene is the most applied acceptor material. Fullerene was used as C60 in bilayer type of active layer. However, in a BHJ it is used as PC61BM (phenyl-C61-butyric acid methyl ester). PC71BM, ICBA (indene-bis-C61-adduct), perylene and pentacene are also used. Some polymers like cyanated polyphenylenevinylens (PPV) are also used [20]. The advantage of using polymers is that it is easy to tune LUMO levels (band gap), thus ensuring efficient charge transport between donor and acceptor.

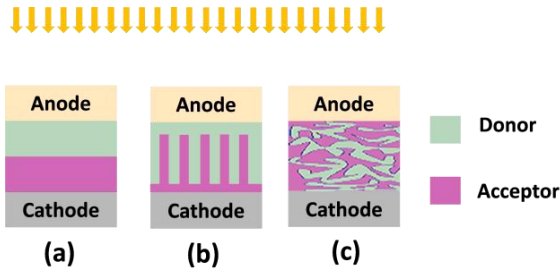


Fig. 3 Different configurations of active layer; (a) Bilayer junction (b) Ideal morphology (C) Bulk Heterojunction

Selective Transport Layers, namely, Electron Transport Layer (ETL) and Hole Transport Layer (HTL) facilitate transport of charge carriers to their respective electrodes. Some ETL materials are titanium oxide, zinc oxide, lithium fluoride. Some HTL materials are Poly(3,4-ethylenedioxythiophene)polystyrene sulfonate (PEDOT:PSS), Molybdenum oxide, Vanadium oxide.

Electrodes collect the charges of opposite polarities and connect them to the external circuit. In addition or alternative to HTL/ETL, to ensure that the electrons and holes get into the right electrodes only, materials having sufficiently different work functions are selected as electrodes. The most used electrode material is indium tin oxide (ITO), as it has high transparency and low resistivity (85% optical transmission and $<10 \Omega/\text{m}^2$). Evaporated metals like aluminum and silver, PEDOT:PSS, doped PEDOT:PSS are also used. One of the electrodes is of transparent material to allow light into the active layer.

Substrates used in OPVs can be classified into glass and plastics. They should essentially be transparent to allow light into other layers. Two most commonly used substrates are floated glass and Polyethylene terephthalate (PET, in flexible solar cells).

Different forms of OPVs can be made with a combination of different architectures and geometries among the aforementioned layers. Some of them are, normal geometry, inverted geometry, and tandem cell.

The one shown in Fig. 2 is that of a normal geometry OPV. Similar to it, an inverted geometry can be obtained by reversing the order of arrangement. Here, the transparent electrode at substrate is negative and ELT and HLT are switched around. It was invented to utilize certain properties of other electrode materials. Since cathode, which reacts with air is encapsulated in between the substrate and active layer, the overall stability of an inverted geometry becomes higher than the normal geometry.

Tandem solar cell is a combination of several junctions i.e. a cell with two or more active layers. The active layers are connected by a recombination layer or interlayer

(HTL+ETL) at their junctions. It has much higher efficiency and lower relaxation losses [21].

IV. CHALLENGES AND DEVELOPMENTS

Though this area of research was born quite a few decades ago, the product has not yet made it to the market. For, they were impeded by a number of challenges restricting their power conversion efficiency (PCE) and stability [5].

The major parameters affecting the performance and the relevant improvements made are discussed here:

A. Absorption efficiency or Spectral incongruence

Absorption efficiency, i.e., how much of the incident light is actually harvested, depends mainly on the materials used in the active layer. The extensively used conjugated π systems intrinsically absorb a broad spectrum of solar radiation. However, since the layer is made extremely thin, the absorption capability of the polymer is impaired [10]. For example, the absorption of P3HT is limited to wavelengths below 650 nm, and hence can collect only about 20% of the solar photons [22] [23].

Use of low-band gap polymers as donors is one solution. Unlike P3HT, they can absorb light with wavelengths of more than 650 nm, corresponding to a band gap of less than 2 eV [24]. Hence, they have better overlap with the solar spectrum and can absorb more photons, thereby giving an efficiency of 5-7% when blended with PC₇₁BM as acceptor [25].

Tandem solar cells are another solution. They exhibit better absorption as whole, as active layers of different absorption bands are employed. Fig. 4 shows the terrestrial solar irradiance and the absorption spectra of a high band gap polymer like P3HT and a low band gap polymer like MH306. Thus, when these two junctions are integrated into one cell, a broader part of the spectrum can be harnessed.

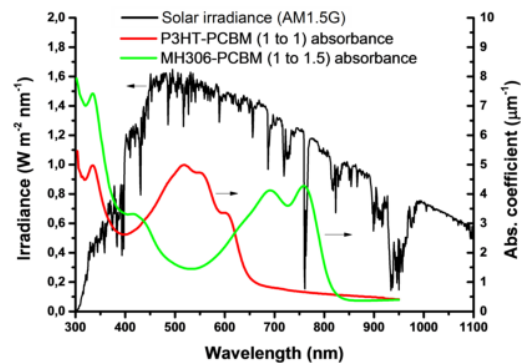


Fig. 4 Solar irradiance spectrum compared with the absorption characteristics of two active layer combinations (Shockley et. Al., Journal of Applied Physics, 1961)

B. High exciton binding energy

When excitons are generated, the electron-hole pairs are held by coulomb forces. Only a few pairs would break this high exciton energy and dissociate into the electrodes to cause current flow. The introduction of donor-acceptor concept, as discussed in section III, solved the problem [3].

C. Short exciton diffusion length

The excitons in the active layer break only at the D/A interface. Longer diffusion lengths correspond to a greater probability that the exciton reaches the D/A interface raising the electron-hole production. With the length of the exciton being just about 5-10 nm, a simple bilayer configuration with a limited D/A interface would be an under-utilization. This gave rise to the so-called Bulk Heterojunction [13] [16], where a huge surface area of D/A interface is created (Fig. 3c).

D. Charge transport

Though the BHJ enhanced the charge separation, it deteriorated charge transport. Unlike inorganic solar cells, BHJ's random orientation of domains cannot offer smooth conductivity of the charges into the electrodes and eventually a sizable quantity of charges recombines, leading to decreased cell efficiency.

To accomplish an ordered array of D/A, integrating nanorods and nanotubes into the active layer was successfully tried. Bang-Ying Yu et al [26] grew controlled vertical titania nanotubes arrays in a P3HT:PCBM active layer by anodization. OPVs were made using Single Wall Carbon Nano Tubes (SWCNT) as acceptor and poly(3-octylthiophene) (P3OT) as donor [27]. Y. A. M. Ismail et al, used a conjugated dye diphenylanthracene (DPA) in P3HT:PCBM active layer and found an increase of photocurrent by a factor of 3.7 [28].

E. Oxidation

Diffusion of water and oxygen into the organic layers is one of the biggest contributors to OPV degradation [29] [30]. Exposure to molecular oxygen with light destroys the π conjugation of P3HT, which induces a loss of absorption over time. Side chains (used to prepare solution during manufacturing) are prone to radical formation. Some electrodes, especially aluminum, are vulnerable to oxidation/corrosion from ambient air. PEDOT:PSS (as HTL) is hydrophilic and hence attracts moisture, affecting other layers.

Avoiding exocyclic double bonds in the main backbone (as in PPVs), using thermo-cleavable polymers as side chains and later cleaving them off, tuning electronic levels of the materials to resist oxidation, cross linking the polymer chains, capping electrodes with less reactive materials, providing proper encapsulation, etc. are some of the measures that can significantly delay degradation.

Chemical degradation still remains a humongous challenge for OPVs. Even with best of encapsulation

techniques the overall device degradation cannot be stopped [29]. It was found that water molecules enter the cell through the outer electrode too.

F. Flexibility and Delamination

In a flexible OPV, the ITO layer tends to crack at the micro level, leading to spike in resistance, thus decreasing performance and stability [31]. Delamination is another degradation mechanism caused due to poor adhesion at interface of different layers, owing to their varied properties.

ITO-free alternatives like thin-metal films, metal grids, nanowires, graphene, etc., have shown superior performance and flexibility. Delamination can be dealt with through the selection of right material combination.

G. Morphological instability

An optimized BHJ cannot be in a thermodynamic equilibrium state and causes it to evolve over time. E.g., Crystallization of PCBM acceptor leads to distortion of the original morphology. Some solutions to it are electron irradiation, annealing, incorporation of cross linkable polymer groups, cleaving of side chains etc.

V. CARBON NANOTUBES AND OPVs

Carbon nanotubes (CNT) find their place in multiple roles in the field of organic solar cells. They can be used as electrodes, replacing ITO to avoid vacuum sputtering and instead facilitate easy fabrication [32]. SWCNTs (Single Walled Carbon Nanotubes) can be used as acceptor material blended with 3-octylthiophene (P3OT) as donor to aide exciton dissociation and to provide ballistic pathway for carriers [27] [33]. The most important and successful application of CNTs have been as additives. When CNTs are blended in active layer, they provide ballistic pathways for the carriers [34] [35]. CNTs are also used as an interlayer in tandem solar cells [36]. One of the interesting applications of CNTs is in the Interdigitated Bulk Heterojunction structure proposed by Guangyong Li [5], which comes close to the ideal active layer configuration (Fig. 3b). When simulated, it exceeded the state-of-the-art performance, to about 12%.

VI. APPLICATIONS

Owing to poor efficiency of 5%-10% and low stability of just about 2-4 years (contrast with 20-30% efficiency and 25 years stability of inorganic PVs), organic photovoltaics have not yet been able to take the leap out of the labs. A 10% efficiency is considered as the threshold for commercialization. With researchers struggling to achieve 9-10% PCE with advanced techniques, OPVs do not seem to enter the market in the near future. The highest recorded PCE is 12%, developed by *Heliatek*, a German OPV company.

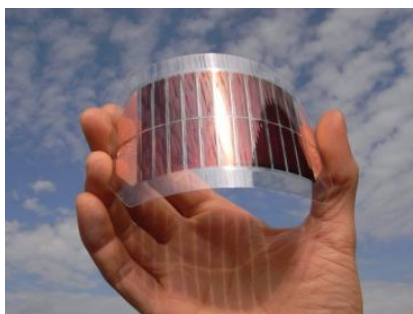


Fig. 5 A flexible organic solar cell module
(source: *exposolar.org*)

However, with flexibility, printability and low-cost manufacturing as their USP, OSCs have a great potential to compete with regular PVs over time. With such advantages, OSCs can be virtually applied anywhere. Maybe we can think of wind mills, with organic solar cells printed on their skins – harnessing both solar and wind power; solar powered mobile devices; power generating backpacks and hats; colourful sunshades at the beaches, lighting up the nearby streetlights in the night; solar cars with excellent aesthetic and aerodynamic design (unlike the present ones); energy scavenging window panes and skyscraper walls [37], etc. Given that OSCs can be printable, flexible and extremely thin, a time may come when every minor electromechanical system used outdoors is self-powered.

These apart, OPVs can obviously be used where inorganic PVs are presently used, at much cheaper costs.

With its current status, we suggest exploring commercialization of Roll-to-Roll produced [38] sheets of OSCs similar to tarpaulin sheets. They can be used to cover scaffolds of under construction buildings or tents used in trekking/housing and use the energy for passive lighting. The characteristics of low PCE, low durability and large surface area are apt for such an application.

VII. CONCLUSION

Though the efficiency of OSCs are considerably lower than other PV technologies, the cost and the energy consumed in its manufacturing is significantly favourable. The energy payback time (EPBT), i.e. duration taken by the cell to recover the amount of energy invested in it for manufacturing, is around 0.3 to 1 year for an OPV, whereas it is between 2 to 4 years for a Si based PV. The embodied energy per square meter of an OPV module was found to be half that of a crystalline silicon PV.

To better the cell performance in terms of PCE and stability, material scientists and chemists are searching for or low band gap materials with appropriate HOMO/LUMO levels, while physicists are putting efforts in matching the cell design with the available spectrum. Mechanical

engineers are trying new ways of easy and quick fabrication using nanotechnology.

The characteristics of organic solar cells seem to address almost all the drawbacks of silicon and other thin film photovoltaics, viz. cost, raw materials, weight, flexibility, manufacturability, etc. Its manufacturing is so easy that its layers can be *printed* in a roll-to-roll process just like news is printed in a printing press; contrast that with the energy-intensive vacuum deposition process involved in manufacturing of crystalline silicon PVs. With slow but steady progress being made in enhancing their efficiency and stability, one can easily expect them to conquer the renewable energy sector in the not too distant future and provide the world with clean and cheap energy.

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