

Organic materials for photovoltaic applications: Review and mechanism



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ARTICLE INFO

Article history:

Received 18 November 2013

Received in revised form 20 January 2014

Accepted 25 January 2014

Available online 21 February 2014

Keywords:

Organic materials

Photovoltaic applications

Energy

ABSTRACT

Recently Heliatek have reported the 12% certified efficiency of organic photovoltaic (OPVs). This rapid progress suggests that the commercialization of OPVs will be realized soon. In addition to the achievement of such high efficiency in OPVs, yet there is a wide need of improvements e.g. the need of electron-acceptors materials other than fullerene, better understanding of charge-transport mechanism in organic materials, requirement of the material compatible with the flexible substrate, durability of the organic materials based devices etc. The aim of this paper is to review the recent developments in OPVs and the potentials of organic photovoltaic, which has caught the attention of many researchers working in the field of optoelectronics. In this article, the organic solar cell mechanism, the basic design, the recent developments and the efficient organic materials for OPVs are reviewed. The authors have reviewed the recent articles to understand the mechanisms of photocurrent generation and sketched out the search for alternative materials for these devices.

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1. Introduction

Over the years, the world's industries and research organization have shown their interest in the renewable energy resources due to the limited fossil fuel reserves [1,2]. The efficient use of sunlight to produce the domestic electricity by means of solar cell devices is the perfect way to deal with the limitation of the reserve fossil fuel. The solar cell industry has been growing by over 30%/year, albeit admittedly from a very low base, and thus still represents a tiny fraction of the world energy supply (much below 1%) [3]. The conventional inorganic materials based solar cells, such as silicon solar cells and heterojunction solar cells which currently dominate the photovoltaic (PV) market, are relatively mature technologies, and the power conversion efficiency of these devices is approaching record limits of about 24.7% for crystalline silicon solar cells [4] and greater than 42.3% for certain multijunction solar cells [5] exposed to more than 400 suns (all reported efficiencies are under AM1.5 illumination conditions). Regardless of the efficient silicon based technology, their fabrication processes are complex, which involves a number of steps that make solar panels expensive and the energy they produce uncompetitive compared to traditional energy sources (e.g. coal, natural gas, hydropower, etc.) Moreover, silicon solar cells are rigid and cannot be fabricated industrially in large sizes due to the limitation of the silicon wafer processing technology. These disadvantages of silicon PVs and their relative limited ability to provide cost effective energy are some of the reasons that have led many researchers to explore alternative materials for solar energy generation. Several different materials have investigated as an alternative for high cost silicon PVs and some of them are currently dominating the PV market. Among them the most promising alternative candidates of the high cost silicon PVs are the organic photovoltaic cell (OPV) which expected to have a major impact in terms of reduction of production costs [6–8]. An organic solar cell or organic photovoltaic (OPV) cell is a photovoltaic cell that uses organic electronics – a branch of electronics that deals with thin film of π -conjugated semiconducting organic molecules, oligomers or polymers for light absorption and charge transport. They are fabricated by very simple and cost-efficient techniques, such as spin coating, spray deposition, and printing. Although OPV efficiencies are not yet competitive with more traditional technologies, a very rapid improvement has been observed in recent years and laboratory certified cells now reach 12% [9]. OPVs are commonly fabricated in thin film form of electron-donor (D) and -acceptor (A) materials with suitable energy levels matching, sandwiched between electrodes. Out of the two, one electrode must be transparent which acts as a window to the incident light. Due to the low dielectric constant in organic components (~ 3) photoexcitation leads to a strongly bound exciton, which needs to be dissociated into free carriers [10]. This dissociation can take place at the D–A interface. Then, free carriers need to be transported to the corresponding electrodes via drift and diffusion processes, where they are collected, giving rise to an electric current. The main reason behind the improper dissociation and transport of the charge carrier is the small diffusion length (10–20 nm) of the excitons [11,12].

The first report on an organic (excitonic) PV cell came as early as 1959, when Kallmann and Pope studied anthracene single crystal. The resulting cell exhibited an extremely low efficiency [13]. Till now, the resulting efficiency of the OPV cell with single active organic layer remained below 0.1% due to the formation of strongly bound excitons which need to be split to produce an external current. The pioneer work in this field has been done by the Tang when he fabricated the novel photovoltaic cell based on a two-layer structure of organic thin films and he achieved a power conversion efficiency of about 1% under simulated AM2 illumination [14]. He takes two organic semiconductors with offset energy bands (i.e.

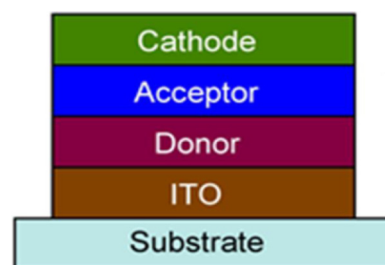


Fig. 1. Bilayer organic solar cell structure.

with different electron affinities: an electron donor and an electron acceptor) and demonstrated that the interface region is primarily responsible for the photogeneration of charges. Although the Tang work was remarkable but the limitation in the Tang's device is due to the low exciton diffusion length in conducting polymers (typically <10 nm), the only light absorbed very close to either side of the interface is effectively active to generate free carriers. Due to this device efficiencies were limited to around 1% for many years.

From the last ten years a large work has been devoted to understanding the basic transport mechanism of organic molecules for the improvement in the efficiency of the cell [15–18]. With the help of these studies and the introduction of new materials a remarkable improvement is observed in the field of OPV. Many simple and useful methods of optimization have been successfully performed in the last decade [19]. The choice of proper solvents [20] as well as the thermal treatment of the solution-processed polymer: fullerene solar cells [21] both lead to a more favorable inner structure in view of the dissociation of bounds electron–hole pairs and the subsequent charge transport. Thus, the power conversion efficiency was increased in the case of annealing from a bare half percent to above 3%. Indeed, optimization by novel routes is an ongoing process, and within the last five years, further steps in improving the power conversion efficiency have been made. Using a concept of planar-mixed heterojunction, coevaporated copper phthalocyanine/fullerene solar cells have reached 5.0% efficiency [22]. The solution-processed polythiophene: fullerene cells achieved between 6% and 8% efficiency by the use of novel materials as well as additives optimizing the phase separation [23–25]. The threshold efficiency for commercial applications of OPV is 10% and the researchers have strived to reach this goal. The remarkable effort to achieving this goal has been done by Zhicai He et al. group. The group demonstrates highly efficient polymer-based organic photovoltaic solar cells with a certified efficiency of 9.2% using an inverted structure [26]. Till now the highest reported efficiency of the OPV cell is 12% which is recently achieved by Heliatek Company [9].

In this article we present an organic solar cell mechanism and review of efficient organic materials. The basic photovoltaic characteristics, OPV device structure, materials for OPV and parameters affecting the efficiency of the solar cell have been described shortly. The different architectures of organic solar cell and their influence on the device performance are also included. We tried to include all the recent work on the OPV cell which have been done by the majority of researchers around the world over the years and request our apology if any contribution has left from this review.

2. Organic solar cell mechanism

To explain the mechanism of organic solar cell, let us take an example of bilayer organic solar cell (Fig. 1). In a bilayer devices two active organic materials namely donor (D) and acceptor (A) are sandwiched between the two electrodes. Out of the two electrodes at least one of the electrodes must be transparent to allow

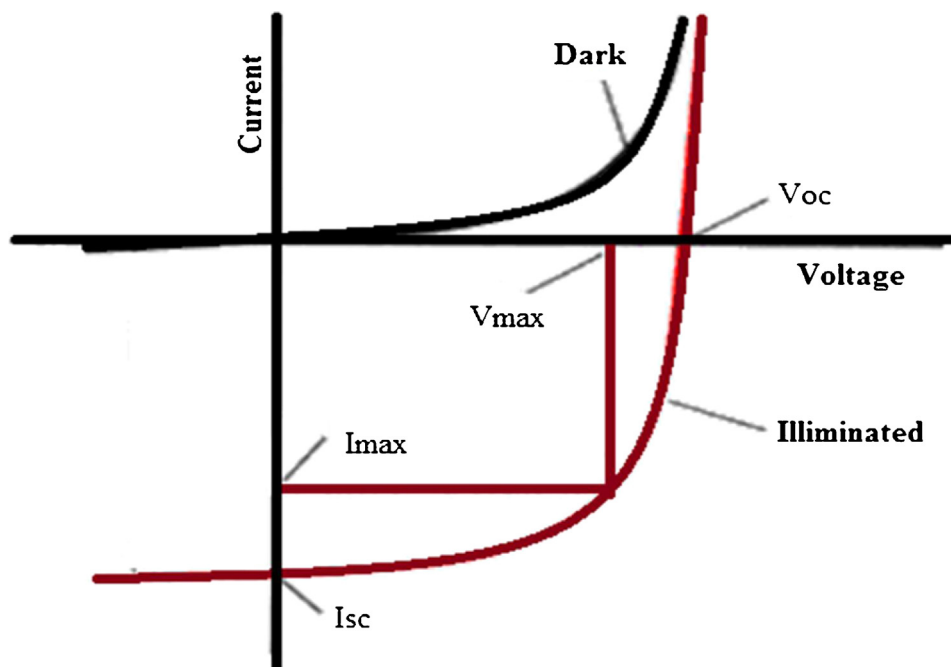


Fig. 2. Current–voltage characteristics of an OPV cell in the dark and under illumination.

photo-excitation of the active materials. The most commonly used transparent conducting electrode is indium doped tin oxide (ITO). The whole structure was made on a supporting substrate (glass, plastic, metal foil etc.). The general mechanism of the energy conversion process has at least five fundamental steps [27,28].

2.1. Absorption of light

The first step in the PV process involves the absorption of the light. In this process, the incident photon while travelling through the transparent electrodes is absorbed by the active organic material (D/A). This absorption of the incident photon leads to the generation of exciton (electron–hole pair). Due to the high absorption coefficients (e.g. $>10^5 \text{ cm}^{-1}$) of organic semiconductors, a layer thickness of a few hundred nanometers is sufficient to absorb the incident light. Besides the large absorption coefficient, still a small portion of the incident light is absorbed because the bandgap is too high.

2.2. Exciton diffusion and charge separation

After the generation of exciton, they diffuse through the donor (acceptor) phase and decay (radiatively or non-radiatively), or dissociate into free charges at D/A interface. Ideally, all photoexcited excitons should reach a dissociation site. But due to the small exciton diffusion length ranging between 1 and 20 nm [29,30], the decay or recombination routes are in competition with a charge carrier generation which limits the efficiency of the cell.

2.3. Charge transport

After the dissociation of exciton at the D/A interface, the electron is found in the acceptor phase whereas hole remains in the donor phase. Subsequently, the electrons and holes must be transported from the D/A interface towards the respective electrodes to produce photocurrent.

The transport of charges is affected by recombination during the journey to the electrodes. Also, interaction with atoms or other

charges may slow down the travel speed and thereby limit the current.

2.4. Charge collection

An efficient collection of charges at their respective electrodes requires that the charge must overcome the potential barrier of organic/metal interface. The charge extraction is a much more complex problem, due to the morphological and chemical nature of the organic/electrode interface. Ion or metal diffusion into the organic layer [31], interfacial dipoles at the contact [32], band bending [33] and chemical reaction [34] can all affect the energetics at the interface and influence the injection process.

3. Organic solar cell characteristics

Fig. 2 shows the typical current–voltage characteristics of a solar cell with dark and illuminated conditions. The power of a cell can be identified by the area enclosed by the fourth quadrant of the curve under illumination. The V_{oc} is the open-circuit voltage which is determined by the potential difference between the two terminals of solar cells under illumination when there is no current flowing through the terminals. The I_{sc} is the short-circuit current defined as the current produced by solar cells under illumination without applying any external potential. The I_{max} and V_{max} are respectively the current and voltage at the maximum power point. To determine the performance of the solar cell the V_{oc} , I_{sc} , fill-factor (FF) and the power conversion efficiency (PCE) are the important parameters. The fill-factor FF is the ratio of the maximum power to the external short and open circuit values and is given by:

$$FF = P_{max}/(V_{oc} \cdot I_{sc}) = (V_{max} \cdot I_{max})/(V_{oc} \cdot I_{sc}) \quad (1)$$

The power conversion efficiency (PCE), is the ratio of the maximum power generated by the solar cell to the incident radiant energy (E) times the surface area (s) of the cell.

$$PCE = P_{max}/(E \cdot s) \quad (2)$$

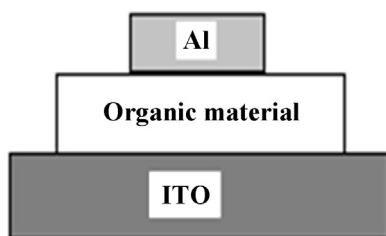


Fig. 3. Single layer OPV cell.

Some other important parameters in solar cell characterization are spectral responsivity (R) and quantum efficiency (QE). Spectral responsivity is defined as the quotient of the current coming out of the cell and the incoming light beam of a given power and wavelength. Quantum efficiency is determined by the number of electron-hole pairs collected per incident photon.

4. Organic solar cell device architecture

Over the year, different OPV device architectures have been used to improve the performance of the cell. The followings are the basic OPV device structures.

4.1. Single active-layer device

Single active-layer OPV cells have the simplest structure compared to the other forms of organic photovoltaic cells. In this cell, thin-film of organic semiconductor has sandwich between the two electrodes (Fig. 3). Typically a layer of indium tin oxide (ITO) with high work function and a layer of low work function metal such as Ag, Al, Mg or Ca are used as a electrodes. The structure of the cell is shown in Fig. 3. In these devices the difference of work function between the two electrode sets up an electric field in the organic layer and this field helps to separate the exciton pairs by pulling the electrons to the positive electrode and holes to the negative electrode. Practically, these devices did not work well and they have very low efficiency. Usually a single organic layer is not enough to absorb all the visible light.

4.2. Double active-layer device

As discussed earlier in Section 2, in the double layer or bilayer cells (Fig. 1), the two active organic materials one acting as a donor (D) and other acts as an acceptor (A) is sandwiched between the two electrodes. These types of devices are more efficient than the single-layer devices because in the later the excitons break-up is

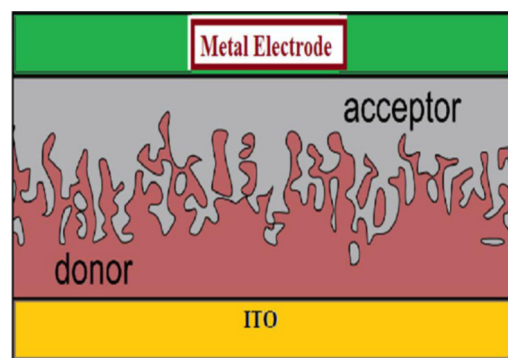


Fig. 4. Bulk heterojunction OPV cell.

more efficient than the former. The double layer OPV cells are also known as planer donor–acceptor heterojunction [35–37].

4.3. Bulk heterojunction photovoltaic cell

In this type of cell, the donor and the acceptor materials are mixed together to form the polymer blend. Fig. 4 shows the structure of the bulk heterojunction cell. In this type of cell, if the length scales of the polymer blend and the exciton diffusion length are comparable, then most of the excitons generated in either material may reach the interface, where they break efficiently. After the exciton break-up, the charge carriers (electrons and holes) are efficiently collected at their respective electrodes while traveling through the blend.

5. Potential materials for OPV applications

In an Organic photovoltaic (OPV) cell a thin film of organic semiconductor (OS) materials (polymer or oligomer) acts as an active layer is sandwiched between a transparent electrode and a metal electrode. Typically the organic semiconductors are either having an electron donating or electron accepting properties. Those OS which has electron donating properties are termed as electron-donor materials and which have an electron accepting properties are electron-acceptors. Typical example of electron-donor and electron accepting materials are copper phthalocyanine (CuPc) and fullerene respectively. The active layer of OPV device is composed of both electron donor and acceptor OS. So far many OS materials (electron donor and acceptors) as an active layer are incorporated into the OPV [38–45]. Now we are going to discuss about those

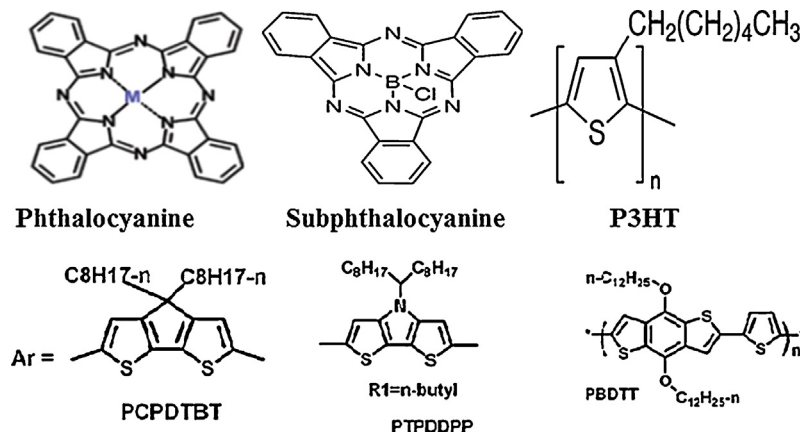


Fig. 5. Structure of some important electron-donor materials.

Table 1
Device structures and their efficiency.

References	Device configuration	Fill-factor	Efficiency (%)
[9] (Heliatek)	N/A	N/A	12 (certified)
[52]	ITO/CuPc/CuPc:C ₆₀ /C ₆₀ /PTCBI/Ag/m-MTDATA/CuPc/CuPc:C ₆₀ /C ₆₀ /BCP/Ag	0.54	5.5
[82]	ITO/PEDOT:PSS/DCAO3TSi:PC61BM/LiF/Al	0.64	5.8
[55]	ITO/SubPc/C ₆₀ /BCP/Al	0.57	2.1
[40]	ITO/MoO ₃ /P3HT:IC ₇₀ BA/Ca/Al	0.74	6.7
[78]	ITO/PFN/PTB7:PC ₇₁ BM/MoO ₃ /Al	0.70	9.2
[79]	ITO/TiPD/PBDTTTC:PC ₇₁ BM/MoO ₃ /Ag	0.65	7.4
[80]	ITO/PEDOT:PSS/PBnDTDTffBT:PC ₆₁ BM/Ca/Al	0.61	7.2
[81]	ITO/ZnO-PVP/PDTS-TPD:PC ₇₁ BM/MoO ₃ /Ag	0.67	8.5
[83]	ITO/PEDOT:PSS/PIDT-phanQ:PC ₇₁ BM/C ₆₀ -bis/Ag	0.61	6.2
[75]	ITO/MoO ₃ /SubPc/FSubPc/C ₆₀ /BCP/Ag	0.54	4
[76]	ITO/PEDOT:PSS/P3HT:99'BF/PBD/BHJ/Al	0.40	2
[77]	ITO/PEDOT:PSS/POPTor GRIMP3HT/EV-BT/LiF/Al	0.40	1.4

materials that have been extensively used and found potential application as an active layer for OPV.

5.1. Electron-donor materials

Phthalocyanines and their homologues are one of the OS materials that are most widely studied in the field of organic electronics. The tremendous photophysical and electrochemical properties have led them to the application as electron-donor or electron-acceptor moieties in multicomponent systems for energy and charge transfer processes. Due to their outstanding light-harvesting properties phthalocyanines are most commonly used as the donor units in such photoactive molecules. Phthalocyanines (Pcs, Fig. 5) [46–49], are two-dimensional 18 π -electron aromatic porphyrin (Por) synthetic analogues [50], consisting of four isoindole subunits linked together through nitrogen atoms. Among the metal-phthalocyanine, copper phthalocyanine (CuPc) and zinc phthalocyanine (ZnPc) are the most popular hole transporting materials which usually serve as the donor and dominant absorption material in OPV devices. Xue et al. reported efficiencies about 4% under 4 suns simulated AM1.5G illumination in a double-heterojunction (CuPc)/C₆₀ thin film cell with Ag as the metal cathode [51]. Their further work on (CuPc)/C₆₀ cell with hybrid planar-mixed molecular heterojunctions further increases the efficiency as high as 5.5% [52].

In these directions, subphthalocyanines (SubPc, Fig. 5) which are the lowest homologues of phthalocyanine have attracted a great deal of interest due to their unique chemistry and photophysical properties [53,54]. SubPc based organic photovoltaic devices are found to be superior than other phthalocyanines based ones due to better energy level alignment and large open circuit voltage (V_{OC}). Kristin et al. [55] have reported that photovoltaic (SubPc/C₆₀) cell show increase in V_{OC} as compared (CuPc/C₆₀) which further increases the efficiency of the cell. The other oligomer organic materials which are successfully served as an electron-donor are the porphyrines and their derivatives [56–58]. The problem with the oligomer organic materials is that they are not very much compatible with the flexible electronics. Most of these are not solution processable and are only thermally evaporated to be used as an active layer in thin film form. Also the requirement of thermal evaporation techniques makes the devices more costly than the solution processable materials based devices.

On the other hand, the polymers based OPV cell are mostly solution processable which make them compatible with the flexible substrate and cost effective. There are many polymer electron-donor materials that are successfully incorporated as an active layer. Among the polymer based electron donor materials, Poly(3-alkylthiophene)s (P3AT) have been used extensively as active material for photovoltaic cells. P3HT (Fig. 5) is the best one in P3ATs as photovoltaic material. P3HT:PCBM ([6,6]-phenyl C61-butyric acid Methylester is abbreviated as PCBM) based solar

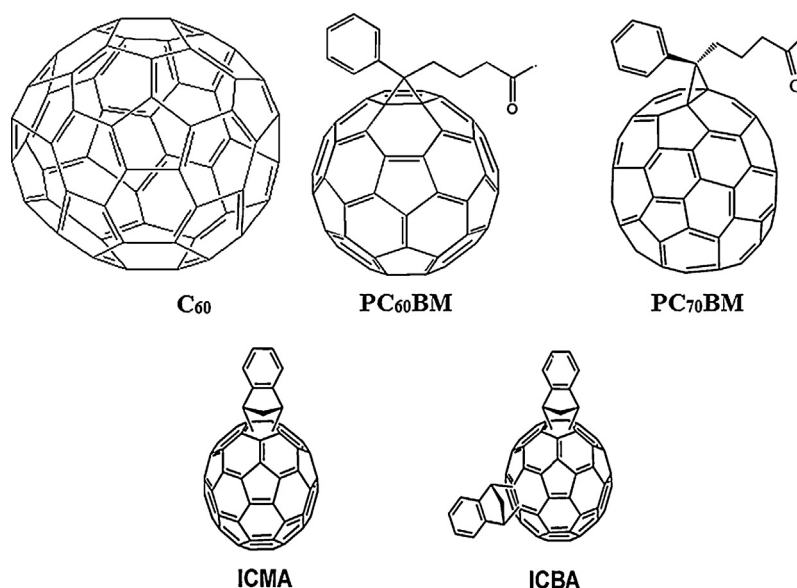


Fig. 6. Structure of fullerene and their derivatives.

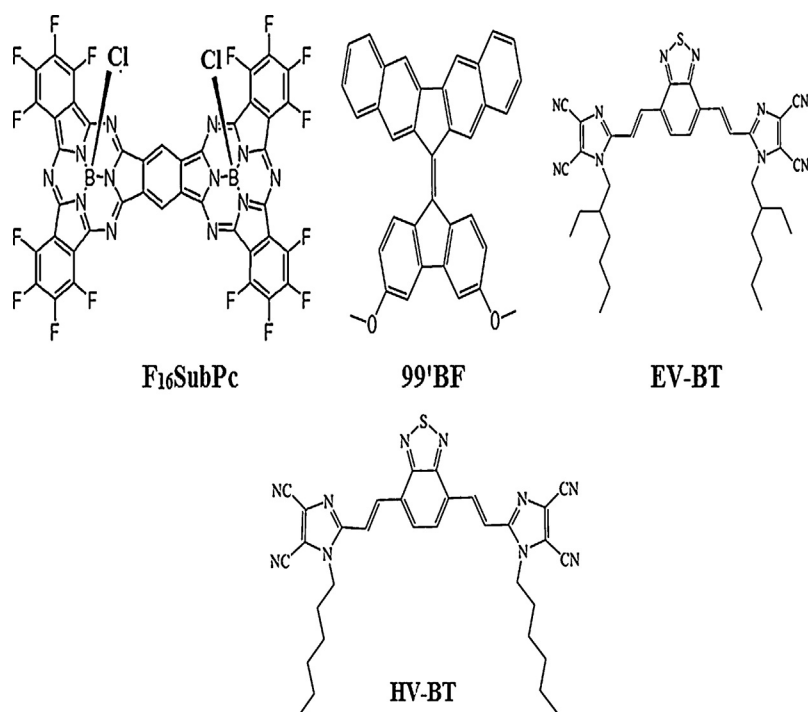


Fig. 7. Structure of some new electron-acceptor materials.

cell has reported an efficiency as high as 5% [59,60]. P3HT is a kind of conducting polymer and it is the excitation of the π -orbital electron in P3HT that gives the photovoltaic effect in the blend [61]. Few of the other efficient polymer based electron donor materials are polymers based on 2,1,3-benzothiadiazole (BT) [62,63], pyrrolo[3,4-c]pyrrole-1,4-dione (DPP) derivatives [64,65] and benzo[1,2-b;4,5-b']dithiophene-based polymers (BDT) (Fig. 5) [66,67]. Table 1 shows the oligomer and polymer based OPV cell and their reported efficiency.

5.2. Electron-acceptor materials

Over the year, many electron-acceptors organic materials including conjugated polymers and small molecular compounds has been used in OPV. Out of these, a very few electron acceptor materials can be used in highly efficient OPV devices. Fullerenes and their derivatives (Fig. 6) are well known and most successful electron-acceptor materials. Fullerene (C_{60}) exhibit a high degree of symmetry and the arrangement of its molecular orbitals provide the foundation for a number of interesting chemical and physical properties [68]. The ability of C_{60} to be reduced reversibly with up to six electrons has led to the synthesis of a large number of donor-acceptor systems in which it acts as an electron acceptor. The C_{60} exhibits very limited solubility in most of the commonly used organic solvents. In order to improve its solubility and also to avoid severe phase separation of D/A blend, [6,6]-phenyl- C_{61} -butyric acid methyl ester ($PC_{60}BM$) was applied in OPVs [69]. In the past few years, due to the solution processable advantage of $PC_{60}BM$ and its corresponding C_{70} derivative ($PC_{70}BM$), they have been extensively used as acceptors in OPVs. Although in comparison with $PC_{60}BM$, $PC_{70}BM$ possesses stronger absorption in the visible range, the C_{70} is much expensive than that of C_{60} due to its tedious purification process, which limits its application [70]. In addition to these fullerenes, there is another fullerenes derivative known as Indene-fullerene which was also used as electron acceptor materials in OPVs [71–73]. These are indene-fullerene monoadduct (ICMA) and indene-fullerene bisadduct (ICBA) (Fig. 6) and can be synthesized

easily by a one-pot reaction. ICMA/P3HT- and ICBA/P3HT-based devices shown higher V_{oc} than that of P3HT/ $PC_{60}BM$ -based devices. The use fullerene and its derivatives as an electron-acceptor in OPV and their related efficiencies are shown in the Table 1. So far fullerene and its derivatives are the dominate electron-acceptors materials for OPV and are also most efficient one. Recently many other electron-acceptor materials are reported as an alternative to the fullerene but yet they didn't approached the efficiency of the fullerene based devices.

Conventionally SubPc has been used as electron-donor material in OPV due to its better energy-level alignment. But recently Beaumont et al. used tetracene (TC) as a electron-donor and SubPc as a electron-acceptor in Tc:SubPc OPV devices and found the efficiency as high as 2.9% [74]. This work found to be very good in the direction to find the alternative of fullerene. Another SubPc derivative, fluorinated subphthalocyanine ($F_{16}SubPc$) found to be a good candidate for OPV as electron-donor [75]. The other electron-acceptor materials include 9,9'-bifluorenylidene[18] (99'BF), vinazene derivatives and 4,7-bis(2-(1-(2-ethylhexyl)-4,5-dicyano-imidazol-2-yl)vinyl)benzo[c][1,2,5]thiadiazole (EV-BT) (Fig. 7) [76,77].

6. Conclusion

A better understanding of the fundamental mechanism of organic photovoltaic process, introduction of new efficient electron donor/acceptor materials and new device architectures leads to the great progress in OPVs in the last few years. For the commercialization of OPV cell having performance comparable to the inorganic counterparts requires an effort from the industry and research institute based organizations. For instant, scientists work on the synthesis of alternate of fullerene and other new organic molecules with multifunctional properties, study of charge transport properties of the organic materials and good understanding of organic molecule structures is appreciable. This rapid progress in the field suggests that the commercialization of OPVs will be realized soon.

Acknowledgement

The Ministry of Education, Youth and Sports of the Czech Republic, Project CZ.1.07/2.3.00/30.0021 “Strengthening of Research and Development Teams at the University of Pardubice”, financially supported this work. This article was written with support of Technology Agency of the Czech Republic, project TE 1010022, FLEXPRINT. One of the author (MS) wish to thank Prof. Luisa Torsi, Department of Chemistry, University of Bari “Aldo Moro”, Italy for continous support and motivation forthe work.

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