

A Review of the Development of Organic Solar Cells Efficiency

Jawad Shoqeir^{1,†}, Samah Al-Sourkhi^{2,†}, Husain Alsamamra^{3,*}

¹Department of Earth and Environmental Sciences, Al-Quds University, Jerusalem, Palestine

²Renewable Energy and Sustainability Master Program, Al-Quds University, Jerusalem, Palestine

³Physics Department, Al-Quds University, Jerusalem, Palestine

Email address:

halsamamra@staff.alquds.edu (Husain Alsamamra)

*Corresponding author

† Jawad Shoqeir, Samah Al-Sourkhi and Husain Alsamamra are co-first authors.

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Abstract: In recent years organic solar cells have been recognized to have tremendous potential as alternatives to their inorganic counterparts, because they have many distinctive features such as they are flexible, colorful, have a lightweight and they are environmentally friendly. They have the ability to produce the cheapest electricity due to their low costs and be competitive with the silicon solar cells. Furthermore, the number of benefits of organic solar cells is expected to increase greatly as the technology is further developed. However, they have some obstacles and challenges to be utilized commercially on a large scale have been highlighted by their relatively low power conversion efficiencies and the relatively short device lifetime. Despite these challenges, the tunability and versatility of organic materials offer promise for future success. The evolution of the organic solar cells' performance over time has been addressed in this work, the present study provide the differences between organic and silicon solar cells which reveal the challenges that affect the development of organic solar cells efficiencies. In addition, it shows the historical development of efficiency of the organic solar cells in each generation: (i) single layer organic solar cells, (ii) donor-acceptor bilayer heterojunction organic solar cells and (iii) bulk heterojunction organic solar cells. Finally, the paper concludes by suggesting that future research should focus on addressing the identified challenges and developing new materials and technologies that can further improve the performance and efficiency of organic solar cells.

Keywords: Organic Solar Cells, Donor, Fullerene, Acceptor, Bulk Heterojunction

1. Introduction

Energy plays an important role in our life especially the electric energy [1]. The fast growth of population around the world every year besides the industrial and technical developments means that there is fast increase of energy demand [2]. In 2019, the global energy supply was 606.6 EJ and most of this demand is met by fossil fuels as presented in figure 1 [3]. However, the depletion of these sources make their prices rise every day which cause the rise in the costs of the daily services.

In addition, carbon (CO₂, CO) and greenhouse gases (SO_x, NO_x,...) emissions from using fossil fuels is causing many environmental problems such as the rise of global

temperatures, acid rain, ozone layer depletion and climate change [4]. Figure 2 summarizes the source of CO₂ emissions in 2019 which reached around 33600 Mt CO₂ and the share of fuel consumption in these emissions was about 15400 Mt CO₂ [5]. This needs a shifting towards sustainable and renewable energy sources, specially wind and solar, to replace the fossil fuels [6-8].

According to Brabec and his co-workers, 2011, solar energy alone has the potential to meet the global demand of energy and the highest energy demand for a whole year can be satisfied by only one hour of sunshine [9]. The renewable energy shares in energy production are increasing every year. In 2019, the solar energy used to generate 2.6% of the global electricity [10].

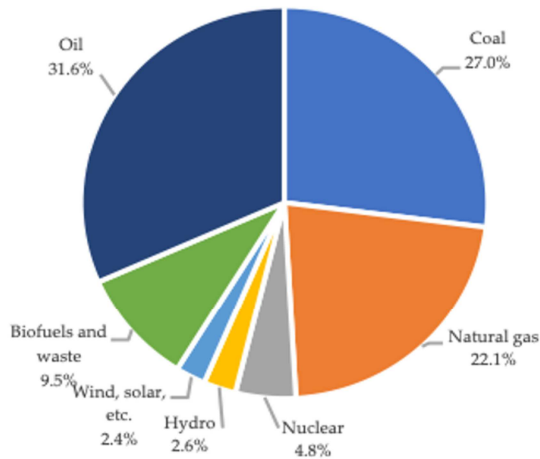


Figure 1. The worldwide energy supply, 2019 [3].

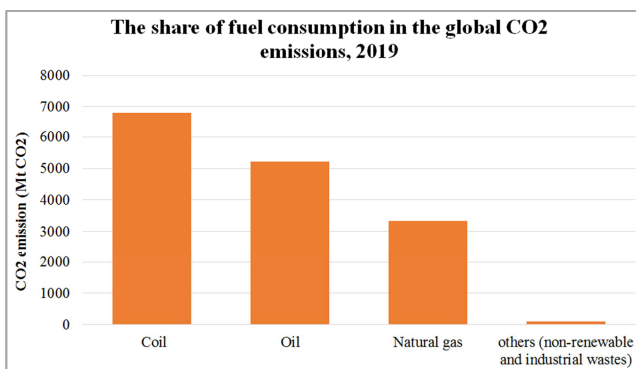


Figure 2. The global CO₂ emissions by fuel consumption, 2019 [5].

Solar cells, also called the photovoltaic cells, are devices used to produce electricity by converting the sunlight based on a chemical and physical phenomenon known as the photovoltaic effect [11]. Currently, the crystalline silicon photovoltaics are dominating the solar market [6]. However, there are many under development technologies have potential to deliver electricity at lower cost [12]. One of the most progressive one is organic solar cells (OSCs) [13].

Organic solar cell is a form of plastic solar cells (polymer solar cells) that use the sunlight to produce electric current by depending on a carbon-based photovoltaic active layer (organic semiconductors). They consist of two organic layers or a homogeneous blend of two organic materials. One of these materials is p-type organic semiconductor which donates electrons (donor organic dye or donor semiconducting polymer) and the other accept them (acceptor, n- type organic semiconductor) [14, 15].

There are many advantages for OSCs that attracted attention in the last two decades, some of them are: (1) organic semiconductors (OSs) can be produced for a specific purpose and via various synthetic pathways, (2) the raw material used are non-toxic and abundant [16], (3) primary vacuum coating, solution processing and can processed by low-cost manufacturing technologies, like inkjet printing [17, 18], (4) able to coat large areas fast and cheap, (5) need nearly one gram of organic semiconductor per m², (6) produce at low temperature, (7) they are light weight, flexible,

can be colorful, fashionably designed and attracted to buildings and houses [6, 19], and (8) has the potential to generate the cheapest electricity [6, 20].

This paper aims to highlight the most important historical development in OSC field with focus on the efficiency. In addition, it aims to list some recent research on OSC efficiency to increase it and surpass one of the main obstacles in front of OSC to be competitive with silicon solar cells in the market.

2. Comparison Between OSC and the Silicon Solar Cells

Even though the organic photovoltaics (OPVs) has many characteristics that made it competitive with inorganic photovoltaics (IPV), it's not used widely like silicon solar cells. Therefore, there are many difference between these solar cells and some of these differences make the silicon semiconductors better than OSs to be used.

The most important difference in operation between OSCs and silicon solar cells is that the absorption of light in OS thin film doesn't lead to efficient free charge carrier's generation (electron-hole pairs) with weak binding between them but to excitons' generation having limited diffusion length and relatively strong binding between electrons and holes [21]. One of the major parameters to determine the performance of PV cells is the life time. The life time of silicon solar cells are long comparative with the life time of OSC [22].

Moreover, one of the most important differences between them that made silicon favorable is that the power convergence efficiencies of silicon solar cells are higher than the efficiencies of OSCs because band gaps in OSCs is higher than IPV's band gaps [23]. The Silicon PVs are stable, strong and have high carrier mobility because they have 3D rigid lattice which provide this mobility whilst OPVs are weaker, less stable than them and have low mobility. The low mobility leads to the limitation in light absorption efficient and the thickness of the device active layer [24].

3. Historical Development of OSC Efficiency

In 1839, the photovoltaic effect was discovered by Becquerel when he observed photocurrent while AgCl electrode were illuminated in an electrolyte solution (photoelectrochemical process) [25]. In 1873, Smith reported the selenium photoconductivity and Adams reported the same in 1876 [26, 27]. Pochettino (1906) reported the photoconductivity in the first organic compound called Anthracene (Figure 3) [28]. The first solar cell was built by Fritts in 1883 by using junctions made of selenium coated with very thin layer of gold and the first OSC was built in 1958 by Calvin and Kearns and was made of magnesium phthalocyanines (MgPc) between two glass electrodes [29, 30].

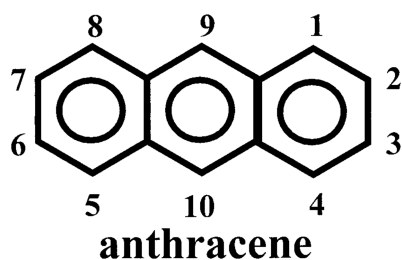


Figure 3. The structure of anthracene [31].

3.1. The First Generation of OSCs

In this generation, also known as the single layer OSCs, figure 4a, the charge separation occurred because of Schottky barrier (the different between the metal that has the lower

work function and the organic layer) and they had a poor power conversion efficiency nearly 0.7% because the potential bias wasn't sufficient to separate the electrons and holes efficiently [32, 33]. For many years after 1958, small molecule OSs single films were used, phthalocyanines and merocyanine SC were typically used. A little photocurrent, less than several microamperes, could be generated between organic film, which had the characteristics of p-type, and a low-work function metal like Al [34, 35]. In 1957, the photoconductivity in the poly polymers (N-vinyl-carbazole, PVK) was testified and it was discovered that many common dyes has the properties of semiconductors in the early 1960s [14, 36]. After many years of research, the results have shown that the simple organic single dye PVs efficiency will probably remain less than 1% [37].

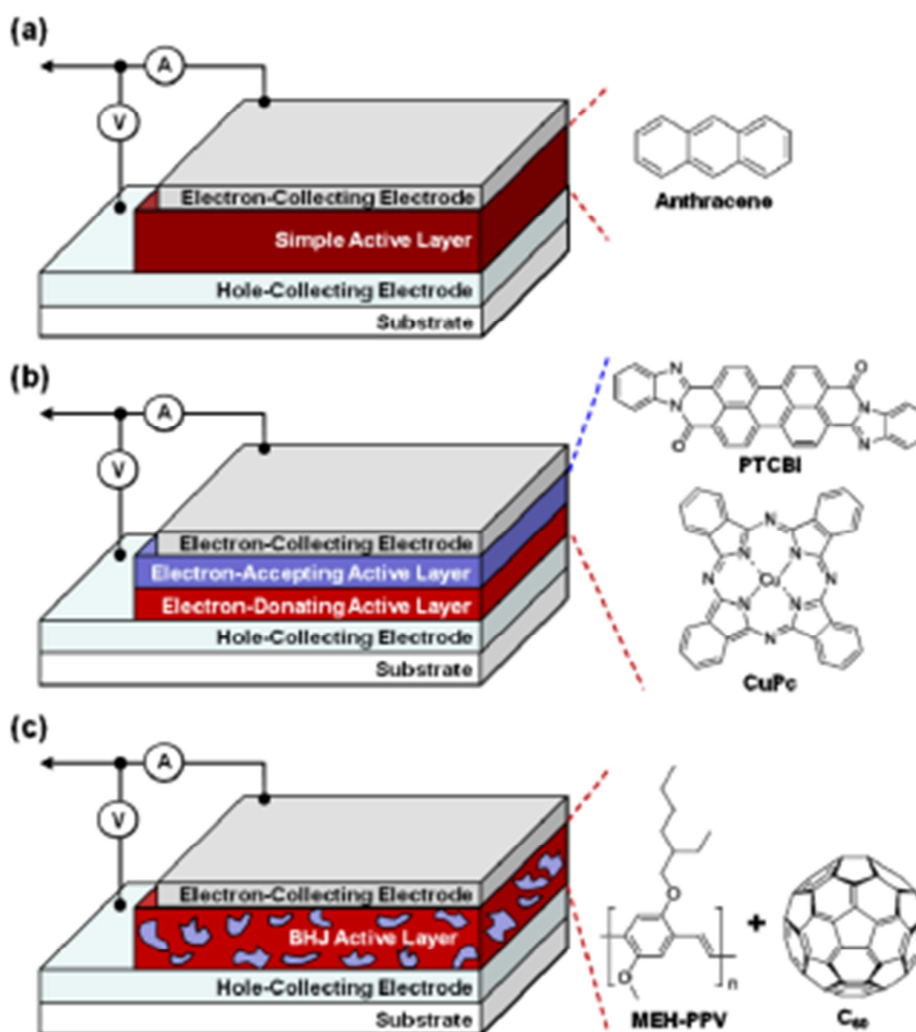


Figure 4. The structure of the three generations of OSC: (a) single layer OSC, (b) donor-acceptor bilayer planar hetero-junctions OSC, (c) bulk heterojunction OSC. The shown chemical materials were the first material used for the first device of each generation.

3.2. The Second Generation of OSCs

This generation is also called the donor-acceptor bilayer planar hetero-junctions (1986), figure 4b. This generation was impacted by one of the most important developments in

the OSCs field. This development was done by Tang in 1986 and was an efficient organic display based on organic light emitting devices (OLEDs) which shown the availability of organic electronic compounds. Before it, the highest efficiency of OSC was 1% [38]. These OSC also had certain limitations.

3.3. The Third Generation of OSCs

There is another important development in OSC field which leads to the third generation, the bulk heterojunction (BHJ) cells, figure (4c), where the active layer is consist of donor and acceptor materials mixed with each other in a form of mixture. This development was in 1991 where Hiramoto et al. co-sublimated donor and acceptor molecules and build the first blended junction (dye/dye hetrojunction) for small molecule cells. the bulk heterojunction is the key to overcome the limitation of exciton diffusion length [14, 39, 40].

In 1992, another development was done by Sariciftci when he reported the first polymer heterojunction device made of fullerene (C_{60}), shown in figure 5a, as an acceptor with poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV), figure 5b, as a donor. This led to discover that the transition of electron from the excited donor polymer to C_{60} happens very fast and the quantum efficiency of it is very high. This is promising for the separation of charge carriers in photovoltaic cells (PV cells) [14, 34, 41].

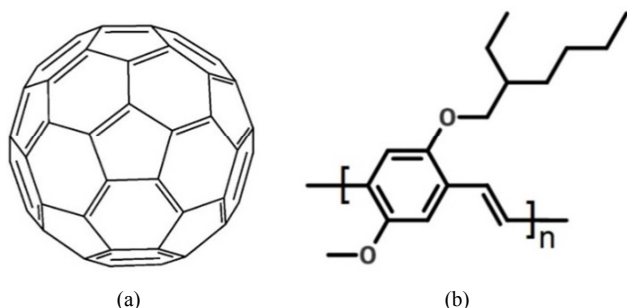


Figure 5. (a) The structure of fullerene [42] and (b) the structure of MEH-PPV [43].

The first bulk polymer/polymer heterojunction PV cell was made by Yu in 1995. He used the MEH-PPV film as donor in this PV cell and the used acceptor was C_{60} with an energy conversion efficiency of 2.9% [44]. In 1996, Halls and his co-workers studied a heterojunction made of Poly (p-phenylenevinylene) (PPV) and C_{60} . The PPV/ C_{60} heterojunction external monochromatic quantum efficiency reached 9% [45].

The efficiency of OSC increased rapidly from 1% in 2000 and reached 12% by 2013. The bulk-heterojunction (BHJ) OSCs had developed greatly in the last years because they have a light weight, able to be manufactured into semitransparent and flexible devices and their costs are low [20]. Nowadays, it has achieved high power conversion efficiencies that exceed 18% [46].

One of the most used acceptors is C_{60} because it is fairly transparent, has a fair electron conductance and high electron affinity. The OSC that uses fullerene as an acceptor has reached power conversion efficiency of 12% [34]. However, the fullerene acceptors have some shortcomings, such as the

limitation of the variability of energy levels and they have weak absorption of light in the region of visible spectra [47].

Recently, there are focus in the development of non-fullerene acceptors (NFAs) because they overcome the failings of fullerenes, have tunable energy levels and have a strong absorption in the region of near-infrared (NIR). With these characteristics, they have high potential to achieve high power conversion efficiencies and upgrade the performance of OPVs in efficient ways [48].

4. Work Mechanism of Bulk-Heterojunction Organic Solar Cells

In the BHJ organic solar cells (figure 6a), the exciton can be generated from donor and acceptor. The OSC absorbs only 30% of the sun light. The work mechanism is shown in figure 6b [15, 34, 48, 49]:

- 1) The photons absorption and the excitons creation: the incident photons absorbed by donor and acceptor strike the electrons. The excited electrons transfer from the highest occupied molecular orbital (HOMO), also called ionization potential (IP), to the lowest unoccupied molecular orbital (LUMO), which is known also as electron affinity (EA), in the donor which generates the excitons (electron-hole pairs that it is bounded together by the electrostatic) if they have energy exceeds the band gap. These pairs have a low life time and returning to the ground state fast (within few nanoseconds) that's because they are strongly bound thus, the dissociation must happen fast.
- 2) The diffusion of generated excitons to donor/acceptor interfaces.
- 3) The dissociation of excitons, which is the key factor to free and generate charge carriers (holes and electrons) on OSs at the interfaces of the donor and acceptor. The energy offset between the LUMO of the donor and the acceptor LUMO forms the overall driving force of electron transfer from the donor to the acceptor. The research defined that to affect the charge dissociation and the exciton splitting, 0.3 eV is the minimum required difference of energy. Moreover, if the LUMOs energy difference is larger than the minimum, it doesn't mean it's a good thing. It became a wasted energy [50, 51].
- 4) The charge carriers transport to the electrodes.
- 5) The extraction of the carriers of the charge at electrodes.
- 6) The organic solar cells performance is determined by three parameters: the fill factor (FF), the short circuit current (J_{sc}) and the the open circuit voltage (V_{oc}). Therefore, these parameters has been rising in the last years so the OPVs can meet the requirement of the market [52].

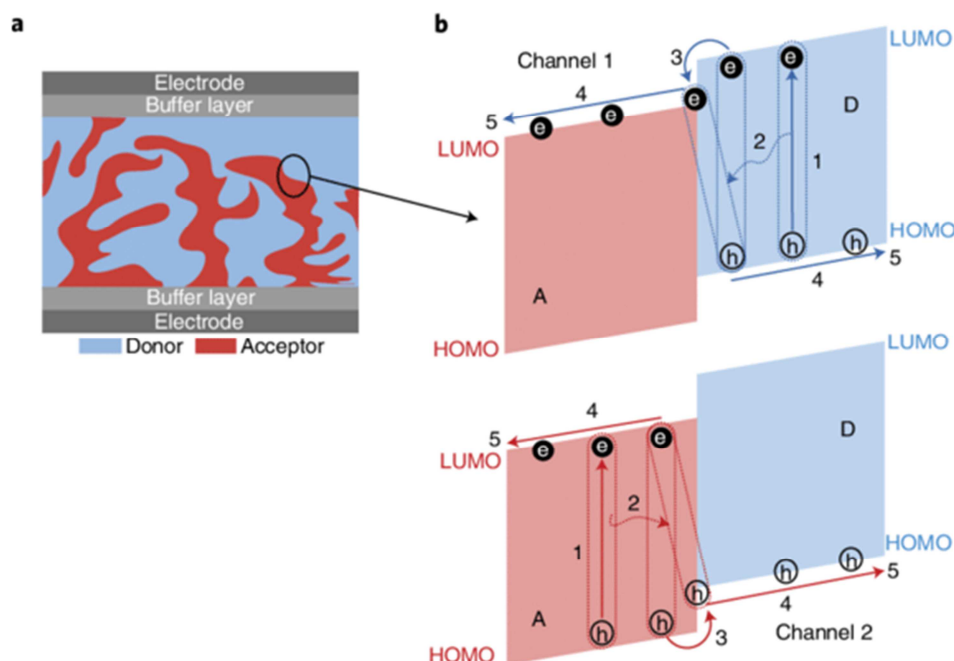


Figure 6. (a) The BHJ solar cell and (b) the work mechanism in it [48].

5. Challenges

There is fundamental challenge that needs to be bypass to achieve higher performance organic photovoltaics (OPVs). This challenge is the necessity of low voltage loss (materials with small energy offsets between their states) and efficient charge separation/transport (high charge-carrier mobility) at the same time [53, 54]. In addition, to maximize the charge generation and transport, it's important to optimize the blend composition and morphology and the energy levels of HOMO and LUMO in the NFA must be positioned with respect to the energy levels of the donor [53, 55].

Many research has been focus in the voltage losses, which can be divided into: (1) Energy transfer losses [56]. (2) Radiative recombination: its unavoidable but can be reduced by lowering the interfacial area of donor-acceptor [57]. (3) Non-radiative recombination, which happens without emitting a photon and occurs due to the coupling to intramolecular vibrations of organic materials themselves and decreasing this lose is the main key to increase the organic solar cells V_{oc} [6, 58].

6. Recent Experiments to Develop OSC Efficiency

There are many experiments in the field of OSC efficiency. This section of paper will discuss two experiments in this field in 2019 and 2020 and present the result efficiencies of their experiments.

In 2019, Yuan *et al.* had achieved a 14.7% efficiency organic cell by introduce a new NFA called Y6, figure 7 [53]. This acceptor uses ladder-type electron-deficient-core-based

central fused ring (dithienothiophen [3, 2-b]-pyrrolobenzothiadiazole) with a 2, 1, 3-benzothiadiazole (BT) core, TPBT. They designed the the NFA as mentioned to fine-tune its affinity, electron absorption and to match with the donor which was PBDB-T-2F polymer (PM6). This donor is is a commercially available polymer with 5.56 eV HOMO energy level and 3.50 eV LUMO energy level.

They were able to design a OPVs based on Y6 and displayed a high efficiency reached 15.7% in two architectures: conventional and inverted. Table 1 summarize the results and the performance of the photovoltaic of the OSC PM6: Y6 at different ratios under AM 1.5G illumination, 100 mW cm⁻². As shown in the table, the highest recorded efficiency was 15.72% when Y6 was blended with the donor PM6 with ratio 1:1.2 for the devices with the conventional architecture and inverted one. The V_{oc} was 0.83 V, J_{sc} was 25.32 mA cm⁻² and the FF of the device was 74.8%.

In conclusion, they designed a new NFA (Y6) which is a new class of NIR-absorption (931nm onset absorption). It's optical bandgap was 1.33 eV and the coefficient of absorption was 1.07×10^5 cm⁻¹.

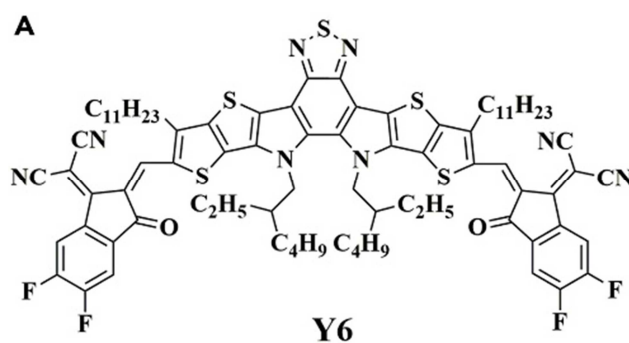


Figure 7. Y6 structure [53].

Table 1. Optimization of the conventional devices D-A ratio and annealing temperature for Y6-based blend films [53].

PM6: Y6 (w/w)	Additive ^a (%)	Annealing (°C)	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF (%)	PCE (%)
1:1.2	-	-	0.86	24.31	73.2	15.31
1:1.5	-	-	0.86	24.21	72.3	15.14
1:1	0.5	110	0.83	25.25	72.9	15.27
1:1.2	0.5	110	0.83	25.32	74.8	15.72
1:1.2	0.3	110	0.83	25.20	73.7	15.42
1:1.2	0.8	110	0.83	24.38	74.7	15.12
1:1.2	0.5	90	0.84	24.52	76.0	15.67
1:1.2	0.5	130	0.82	24.74	74.1	15.02
1:1.2	0.5	150	0.81	23.98	74.8	14.54
1:1.5	0.5	110	0.84	24.96	74.6	15.64
1:1.7	0.5	110	0.83	25.42	73.3	15.48

^a The additive is chloronaphthalene (CN)

In 2020, Luo and his co-workers increased the efficiency of organic solar cells by using the asymmetric end groups to increase the fine tuning of levels of energy [59]. They used PM6 as the donor and Y6 (BTP-4F) as the NFA in this experiment as the previous experiment.

The importance of fine-tune energy levels of both the acceptor and donor is the potential to reach the minimum energy offsets of the HOMO. They achieved this by modifying the end group of acceptor from IC-2F to CPTCN-Cl. However, the upshift of the energy level of HOMO was too much which caused a problem in the energy alignment between donor and acceptor. Thence,

guarantee sufficient separation of charge is important. To achieve that, they used the asymmetric end groups and achieved higher V_{oc} and PCE.

Table 2 presents the performance of the photovoltaic of the OSC based on PM6: acceptors under the same conditions of previous experiment. Its noticed that the efficiency of fine tune energy levels without asymmetric OSC has a lowest efficiency (14.49 %) while the highest efficiency of 17.06% is the result of the highest asymmetric OSC and they have higher V_{oc} compared with the main device which is PM6: BTP-4F.

Table 2. The Optimization of the photovoltaic performance of the OSC based on PM6 with different acceptors [59]. The data were obtained from 20 devices.

Device	J _{sc} (mA cm ⁻²)	V _{oc} (V)	FF	PCE _{avg} (%)
PM6: BTP-4F	25.19 (25.4 ± 0.15)	0.841 (0.837 ± 0.002)	0.773 (0.761 ± 0.007)	16.37 (16.18 ± 0.17)
PM6: BTP-2ThCl	23.46 (23.38 ± 0.35)	0.885 (0.884 ± 0.003)	0.698 (0.693 ± 0.006)	14.49 (14.33 ± 0.18)
PM6: BTP-2F-ThCl	25.38 (25.37 ± 0.28)	0.869 (0.869 ± 0.005)	0.774 (0.761 ± 0.008)	17.06 (16.77 ± 0.19)

7. Conclusion

There is a great attention to the field of organic solar cells. This increase in attention on these cells is due to the potential of these cells to be one of the most important sources of the cheapest electricity in the future. They also have many advantages such as the ability to produce them in different technologies and being cheap.

However, the main prevention of using them widely until now is the low efficiencies of them and improving the organic solar cells efficiencies is essential to enhance using them and be competitive with silicon solar cells. Every now and then, there is a new development that increase the efficiency. There is many ways to increase the efficiency like using the non-fullerene acceptors and the inverted structure of bulk heterojunction solar cells. Nowadays, the efficiencies of organic solar cells have reached over 23% in laboratory.

The organic bulk heterojunction solar cells and the non-fullerene acceptors are the topics that currently receive the largest attention in this field. They have a strong potential to increase the efficiency, absorption and the mobility of organic solar cells and overcome the challenges faces the use of them in commercial ways.

The solar cells in general are the most used devices to convert the solar energy to electricity and meet the global demand of energy by sustainable and friendly source. The ability to use this renewable energy in efficient and large scales is important for our future thus, it's important to concentrate on developing the organic solar cells.

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