



Review

Solution-processable polymeric solar cells: A review on materials, strategies and cell architectures to overcome 10%



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ABSTRACT

Organic photovoltaics will become 30 years old relatively soon. In spite of the impressive development achieved throughout these years, especially in terms of reported power conversion efficiencies, there are still important technological and fundamental obstacles to circumvent before they can be implemented into reliable and long-lasting applications. Regarding device processing, the synthesis of highly soluble polymeric semiconductors first, and fullerene derivatives then, was initially considered as an important breakthrough that would definitely change the fabrication of photovoltaics once for all. Nowadays, the promise of printing solar cells by low-cost and high throughput mass production techniques still stands. However, the potential and the expectation raised by this technology is such that it is considerably difficult to keep track of the most significant progresses being now published in different and even monographic journals. There is therefore the need to compile the most remarkable advances in well-documented reviews than can be used as a reference for future ideas and works. In this letter, we review the development of polymeric solar cells from its origin to the most efficient devices published to date. After analyzing their fundamental limits, we separate these achievements into three different categories traditionally followed by the scientific community to push devices over 10%

Abbreviations: BCP, bathocuproine; BHJ, bulk heterojunction; $C_2C_6GeIDT-BT$, germanium heteroatom substituted indacenodithiophene-benzothiadiazole; Ca, calcium; C-PCBSD, cross-linked [6,6]phenyl- C_{61} -butyric styryl dendron ester; Cs_2CO_3 , cesium carbonate; CsF, cesium fluoride; DIO, 1,8-diiodooctane; DTS(FBTTh₂)₂, 7,7'-[4,4-bis(2-ethylhexyl)-4H-silolo[3,2-b:4,5-b']dithiophene-2,6-diyl]bis[6-fluoro-4-(5'-hexyl-[2,2'-bithiophen]-5-yl)benzo[c][1,2,5]thiadiazole]; F-DTS, p-DTS(FBTTh₂)₂; E_g , energy bandgap; FF, fill factor; HOMO, highest occupied molecular orbital; ICBA, 1',1'',4',4''-tetrahydro di[1,4]methanonaphthaleno[1,2:2',3',5,6:2'',3''] [5,6] fullerene- C_{60} ; I_{sc} , short circuit current; ITO, indium tin oxide; J_{sc} , short circuit current density; LiAc, lithium acetate; LiF, lithium fluoride; LUMO, lower unoccupied molecular orbital; MDMO-PPV, poly[2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylenevinylene]; MEH-PPV, poly[2-methoxy-5-(2'-ethyl-hexyloxy)-p-phenylenevinylene]; MgF, magnesium fluoride; MoO₃, molybdenum oxide; NiO, nickel oxide; NREL, national renewable energy laboratory; ODT, 1,8-octanedithiol; OLED, organic light emitting diode; OPV, organic photovoltaics; P3HT, poly(3-hexylthiophene); P3HTTz, poly(3-hexyl-2,5-bithienyl); pBBTDPP2, poly[3,6-bis-(4'-dodecyl-[2,2']bithiophenyl-5-yl)-2,5-bis-(2-ethyl-hexyl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione]; PBDTTPD, poly(benzo[1,2-b:4,5-b']dithiophene-thieno[3,4-c]pyrrole-4,6-dione); PBDTTT-C-T, poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-b:4,5-b']dithiophene-2,6-diyl-alt-(4-(2-ethylhexanoyl)-thieno[3,4-b]thiophene-)-2,6-diyl]; PC₆₀BM, [6,6]-phenyl-C₆₁-butyric acid methyl ester; PC₇₀BM, [6,6]-phenyl C₇₁-butyric acid methyl ester; PCDTBT, poly N-9-hepta-decanyl-2,7-carbazole-alt-5,5,4,7-di-2-thienyl-2,1,3-benzothiadiazole; PCE, Power conversion efficiency; PCPDTBT, poly[2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta [2,1-b:3,4-b']dithiophene)-alt-4,7(2,1,3-benzothiadiazole)]; PDPP3T, poly[2,5-bis(2-hexyldecyl)-2,3,5,6-tetrahydro-3,6-dioxopyrrolo[3,4-c]pyrrole-1,4-diyl]-alt-2,2':5',2''-terthiophene]-5,5''-diyl]; PDPP-TPT, poly-2,5-bis(2-hexyldecyl)-2,3,5,6-tetrahydro-3,6-dioxopyrrolo[3,4-c]pyrrole-1,4-diyl]-alt-(2,2'-(1,4-phenylene)bisthiophene)-5,5'-diyl]; PDPP-TT-T, thieno[3,2-b]thiophene-diketopyrrolopyrrole; PDTP-DFBT, poly[2,7-(5,5-bis-(dimethyloctyl)-5H-dithieno[3,2-b:2',3'-d]pyran)-alt-4,7-(5,6-difluoro-2,1,3-benzothiadiazole)]; PEDOT:PSS, poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate); PEIE, polyethylenimine, 80% ethoxylated; PFN, poly[(9,9-bis(3'-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)]; PIDTDTQx, indacenodithiophene-quinoxaline; P_{in} , incident light intensity; PMDPP3T, poly[[2,5-bis(hexyldecyl)-2,3,5,6-tetrahydro-3,6-dioxopyrrolo[3,4-c]pyrrole-1,4-diyl]-alt-[3',3''-dimethyl-2,2':5',2''-terthiophene]-5,5''-diyl]; PPV, poly(p-phenylenevinylene); PTB7, poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]]; R2R, roll-to-roll; TiO_x, titanium oxide; V₂O₅, vanadium oxide; V_{oc}, open circuit voltage; WO₃, tungsten trioxide; ZnO, zinc oxide; ZnS, zinc sulfide.

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power conversion efficiency: Active materials, strategies -fabrication/processing procedures- that can mainly modify the active film morphology and result in improved efficiencies for the same starting materials, and all the different cell layout/architectures that have been used in order to extract as high photocurrent as possible from the Sun. The synthesis of new donors and acceptors, the use of additives and post-processing techniques, buffer interlayers, inverted and tandem designs are some of the most important aspects that are in detailed reviewed in this letter. All have equally contributed to develop this technology and leave it at doors of commercialization.

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0. Broader context

Organic photovoltaics (OPV) was born as a new possibility to decrease the fabrication cost of solar devices and be validated as one of the most promising renewable energy sources. Thirty years later they have still however not fulfilled this expectative. The non-stop development of existing technologies makes it still difficult for OPV to compete with traditional systems such as silicon solar cells in terms of efficiency and reliability. However, the unique selling properties of this technology extend the potential of OPV from particular niche markets to generic energy production. These are mainly based on mechanical flexibility, transparency and processing of arbitrary shape devices. In this way, OPV is still a real and promising alternative for certain applications for which added functionality to already existing elements is the key selling point. Portable/flexible/wearable electronics and building integrated photovoltaics are some of the most significant examples toward where this technology is being addressed. Moreover, the possibility of printing modules at production speeds up to several meters per second with low-cost and high throughput techniques such as inkjet, slot die, screen or gravure printing allows the coating of the same photoactive area in a single day as that of a traditional silicon foundry in a year. Thus, OPV is also a technology subject to further study for on and off-grid applications for which energy production is still the leitmotif.

1. Introduction

Abundance of raw material, simplicity in device fabrication and easy integration into different applications thanks to their lightweight, semitransparency, flexibility and color tunability, have become organic photovoltaics (OPV) into an attractive source of green energy. Nowadays research on this technology is in one way focused on understanding the physics behind and in the other way on achieving as high efficiency as possible.

Nelson calculated the limiting efficiency for an ideal single solar cell as a function of the semiconductor bandgap taking into account the incident and extracted power from the photon fluxes. She considered a two band system for which the ground state -lower band- is initially full and the excited state -upper band- is empty. The bands are separated by a bandgap, E_g , and electrons in each band are supposed to be in a quasi thermal equilibrium at the

ambient temperature. In the case that no potential is lost through resistance anywhere in the circuit, and being radiative recombination of electrons with energy larger than the bandgap the only unavoidable loss, a limiting efficiency of about 33% at a bandgap of 1.4 eV (885 nm) [1] was calculated providing all incident light with energy larger than the bandgap is absorbed. Each absorbed photon generates exactly one electron-hole pair and excited charges are completely separated. In principle, all the assumptions made for this model are perfectly valid also for organic semiconductors. However, more accurate models explicitly developed for polymer:fullerene bulk heterojunction solar cells and that hence take into account their detailed work mechanisms predict maximum efficiencies of around 15% [2] and 21% [3] for single and tandem cells respectively. The main reasons why ideal performances are not achieved are incomplete absorption of the incident light due to either reflection and/or non perfectly opaque contacts, non-radiative recombination of photogenerated carriers, i.e. excited charges that are trapped at defect sites and recombine before being collected leading to transport losses, and voltage drop due to non-ideal series and/or parallel resistance within the bulk and between the active film and the external circuit. All these aspects need to be tackled in order to overcome the current reported record efficiencies and take them closer to the theoretical limits. As we will show, this can be done by actuating on active materials, fabrication/processing procedures (strategies) and device layout/architectures.

Let us first consider the key performance characteristics of an organic solar cell and the influence of the later issues on their performance. The efficiency is defined as the ratio between the voltage at open circuit conditions (V_{oc}), the output current at short circuit conditions (I_{sc}), the fill factor (FF) of the device and the incident light intensity (P_{in}) (see Eq. (1)).

$$\eta = \frac{V_{oc} \cdot I_{sc} \cdot FF}{P_{in}} \quad (1)$$

It is clear that maximizing the efficiency is thus a matter of increasing V_{oc} , I_{sc} and FF as much as possible.

V_{oc} is ideally limited by the energy difference between the LUMO level of the acceptor and the HOMO level of the donor, therefore, it can be theoretically adjusted up to a certain extent by modifying the energy levels of the materials [4–7]. In consequence, the election of the donor and acceptor material will define the upper limit. However,

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