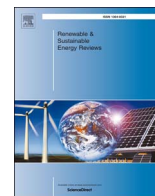




Contents lists available at ScienceDirect

# Renewable and Sustainable Energy Reviews

journal homepage: [www.elsevier.com/locate/rser](http://www.elsevier.com/locate/rser)

## Fundamentals of bulk heterojunction organic solar cells: An overview of stability/degradation issues and strategies for improvement



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### ARTICLE INFO

#### Keywords:

Bulk heterojunction organic solar cells (BHJ OSCs)  
Performance characteristics  
Working principle  
Stability  
Degradation  
Strategies to improve

### ABSTRACT

In the last few years, the performance of organic solar cells (OSCs) based on bulk heterojunction (BHJ) structure has remarkably improved. However, for a large scale roll to roll (R2R) manufacturing of this technology and precise device fabrication, further improvements are critical. This article highlights the fundamentals of a BHJ OSC, including its working principle and performance characteristics. The importance of stability for the device lifetime is underpinned and different degradation factors affecting the operational life of OSCs are discussed. Lastly, the strategies to improve the stability of OSCs, including the encapsulation of device, morphology control in BHJ layer, interfacial engineering in terms of buffered layers, use of inverted geometry and alternative electrode materials are highlighted. Moreover, a simple mathematical model of degradation trends in OSCs is proposed. This review provides a comprehensive insight into the current status of BHJ OSCs regarding stability/degradation and covers almost all critical aspects that are considered important to understand it.

### 1. Introduction

Solar energy is believed to have the highest potential among other alternative energy resources such as hydroelectric, biomass and wind energy. It is inexhaustible and environmental friendly, especially, organic solar cells (OSCs) have attracted immense attention as a possible alternative to their inorganic counterparts [1]. Potential for cost effective and fast roll to roll (R2R) production, as well as their light weight and fabrication on flexible substrates, provide them an edge over traditional inorganic solar cells [2–5]. Although power conversion efficiencies (PCEs) of more than 10% have been recently reported [6,7]. However, significant work is required to translate the progress made at lab-scale towards the large scale industrial production [8,9].

An ideal photovoltaic device should possess a consistent performance during its operational lifetime; however, organic semiconductors are often regarded as inherently unstable when they are subjected to the cyclic environmental changes [10]. Undoubtedly, significant degradation pathways have been posited to occur at virtually every layer and interface of OSC devices. Therefore, the stability of the OSCs is of paramount importance to identify and improve the degradation factors affecting the device performance. Until recent years, the search for

highly efficient OSC materials and device structures has been of tremendous interest in the OSC community, with device lifetime being largely ignored. Therefore, the scientific community is changing the focus to the stability constraints of the OSCs, largely driven by the recent critical needs to understand and optimize the stability and reliability of organic photovoltaic based products [11–13]. Several strategies have been adopted to improve the performance of OSCs such as synthesis of new donor and acceptor fragments of the photoactive blend [14–16], controlling the morphology of photoactive layer [17–19], employing interfacial layers [20–22], and designing and implementation of new device architectures [23–25]. As a result, despite the limited demonstration of actual large-scale installations of OSC modules, a steady increase in their efficiency and stability is frequently published [26].

This review article discusses fundamental concepts of BHJ OSCs, with a focus on factors limiting the device stability and strategies to improve the stability. Despite the notable progress in both the efficiency and stability, the future of OSCs is uncertain because of several stability challenges. Here we discuss and proposed solutions to these challenges. Firstly, we briefly described the degradation factors that limit the device lifetime, such as morphology of photoactive layer, diffusion of

**Abbreviations:** BHJ, Bulk heterojunction; FF, Fill factor; HOMO, Highest occupied molecular orbital; HTL, Hole transport layer;  $J_{sc}$ , Short circuit current density;  $L_D$ , Diffusion length; LUMO, Lowest unoccupied molecular orbital; OSCs, Organic solar cells; P3HT, Poly(3-hexylthiophene-2,5-diyl); PCBM, [6,6]-Phenyl-C61-butyric acid methyl ester; PCDTBT, Poly[N-9'-heptadecanyl-2,7-carbazole-*alt*-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)]; PCE, Power conversion efficiency; PEDOT:PSS, Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; PV, Photovoltaics; R2R, Roll to roll;  $V_{oc}$ , Open circuit voltage; WF, Work function

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<https://doi.org/10.1016/j.rser.2017.12.008>

Received 2 November 2017; Accepted 16 December 2017

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electrodes and interfacial layers, oxygen and water ingress, photo-degradation and mechanical degradation. Lastly, we surveyed and analyzed the strategies to increase the stability, such as encapsulation from outside atmosphere, interfacial engineering to enhance performance, morphology control and use of inverted device architecture.

## 2. Fundamentals of bulk heterojunction solar cells

A BHJ OSC essentially consists of a multilayer structure in which each layer in the device architecture could be deposited by an individual fabrication technique. The absorber layer is comprised of two constituents; a donor material is usually a conjugated polymer, conjugated pigments or oligomers, and for an acceptor material often fullerene derivatives are used. The photoactive layer is sandwiched between the anode and top low work function (WF) cathode. Often the interfacial layers namely hole transport layer (HTL) and an electron transport layer (ETL) are inserted between the anode-photoactive and cathode-photoactive interfaces, respectively to improve performance and stability of the BHJ OSCs [27]. Over the years, inverted device architecture has also been established for BHJ OSCs. In an inverted device, the bottom transparent electrode serves as the cathode while the top electrode is an anode. The performance of inverted devices is comparable with the normal architecture solar cells; in addition, they also exhibit relatively higher environmental stability. Typical device architecture for normal and inverted solar cells is illustrated in Fig. 1.

### 2.1. Operation principle

The simplified working principle of BHJ OSC device can be described in at-least four fundamental steps namely (i) photons absorption and exciton formation, (ii) exciton diffusion and splitting, (iii) charge transportation and, (iv) charge collection [27–29]. In a BHJ OSC device, light is usually absorbed by the donor material, e.g., a conjugated polymer. Upon absorption of photons, an electron is excited from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO). The offset between donor (LUMO) and acceptor (LUMO) must be in the range of 0.1–1.4 eV to generate the electron-hole pairs also known as the excitons. The excitons must diffuse to the donor-acceptor interface where there is sufficient potential energy drop to split these excitons into the free charge carriers, i.e., the electrons and the holes [30]. After splitting into free charge carriers, each carrier must be transported to the respective electrode through the bicontinuous interpenetrating pathway while avoiding recombination and trapping of charges. Some limitations and losses could occur during these steps such as absorption loss due to spectral mismatch, thermalization loss, the insufficient energy required for exciton splitting, and charge recombination, etc. [31]. A detailed description of each of the steps involved from light absorption to the charge carrier collection is presented below.

#### 2.1.1. Light absorption and exciton generation

As the first and essential requirement to achieve high efficiency, the photoactive layer must absorb the maximum of the incoming sunlight.

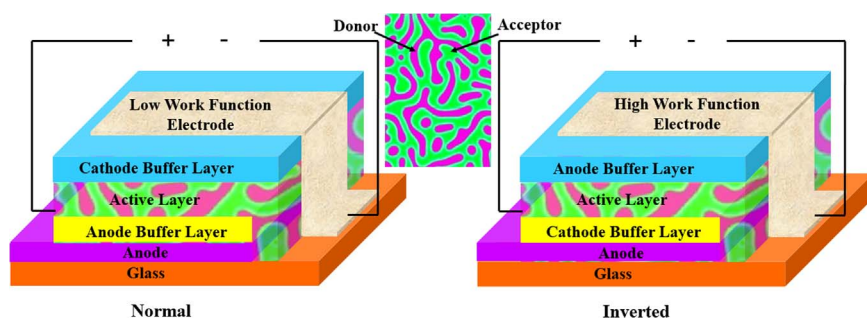


Fig. 1. Device architecture of the (a) normal and (b) inverted BHJ OSCs.

Light is usually absorbed by donor fraction of the BHJ photoactive layer. Due to high absorption coefficient of conjugated polymers ( $10^7 \text{ m}^{-1}$ ), they can effectively absorb light at maximum of their absorption spectrum [32] with very low thickness of the photoactive (usually up to 100 nm) layer as compared to their inorganic silicon (an indirect semiconductor) based counterparts where thicknesses of hundreds of micrometres are required. Moreover, the thickness of the polymer based photoactive layer is also limited to 100 nm due to low charge-carrier mobilities in most of the polymers that lead to an absorption of only 60% of the incident light at the absorption maximum (excluding back reflection of the electrode) [32]. In contrast, the inorganic semiconductors can effectively absorb the whole visible solar spectrum [28,32]. Thus, low absorption in conjugated polymers leads to low photocurrent generation. Interestingly, the absorption of light can be enhanced by lowering the band gap of donor polymers that results in absorption of a maximum number of photons that lead to higher PCEs [33,34]. Therefore, materials with lower band gap are necessary to optimize the photon harvesting. For instance, a material with band gap lower than 2 eV is considered as a low band gap material that leads to the possibilities of improving the efficiency of OSCs due to a better overlap with the solar spectrum. For example, a band gap of 1.1 eV can cover 77% of the AM 1.5 solar photon flux as compared to the band gap of 1.9 eV that can hardly cover 30% of the AM1.5 photon flux [34]. Thus, a low band gap material can significantly improve the photocurrent generation.

When the light is illuminated from the transparent electrode side, and upon absorption of the photon; an electron is excited from the HOMO to the LUMO. It is similar to inorganic semiconductors where the electron is excited from valence band to the conduction band. As a result, an electron-hole pair (exciton) with binding energy typically in the range of 0.1–1.4 eV is generated [30]. The excitons are then migrated to the donor-acceptor interface. A schematic representation of the steps in energy production and corresponding band diagram of an OSC is shown in Fig. 2.

#### 2.1.2. Exciton diffusion and charge dissociation

The energy offset in LUMO between donor and acceptor materials breaks the Coulomb attraction that ultimately causes the excitons to dissociate [35–37]. As most of the conjugated polymers exhibit a shorter lifetime of the excitons, the diffusion lengths are limited to a few nanometers (less than 20 nm), which is much shorter than the optical absorption pass length ( $\sim 100\text{--}200 \text{ nm}$ ). It is, therefore, a prerequisite that excitons must be generated within their diffusion length ( $L_D$ ) for efficient charge generation [2]. Exciton diffusion length is defined as the distance traveled by an exciton before recombination [38]. The reported excitons diffusion length for various conjugated polymers significantly varies from 5 to 20 nm [39–41]. Thus, the thickness of the photoactive layer is very much critical for an efficient charge generation.

In organic semiconductors, photogenerated holes and electrons at the donor/acceptor interface experience a strong Coulomb binding energy [42,43]. These Coulomb bound electron-hole pairs have to be dissociated to get free charge carriers. However, they either recombine

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