



Measurement of contact surface photo-voltage from forward bias C-V characteristics of P3HT:PCBM based BHJ solar cells



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ABSTRACT

The surface photovoltage (SPV) and carrier accumulation in open circuit conditions are sensitive indicators of the contact quality and band bending. However, these are not accessible directly in experiments. We analyze the capacitance-voltage (C-V) characteristics in P3HT:PCBM based bulk heterojunction to investigate SPV for both conventional (transparent anode) and inverted (transparent cathode) architectures. The C-V characteristics under dark conditions are analyzed using drift-diffusion model to extract built-in voltage, while under white light illumination it yields SPV. The SPV and the corresponding carrier density at the anode are quantified as a function of light intensity for conventional structure and compared with those of inverted structure to distinguish between the mechanisms in the two cases. The inverted structure with higher open circuit voltage is marked with less SPV compared to the conventional one. The technique of obtaining SPV as described here and the underlying mechanism can be used to suggest modifications in device architecture.

1. Introduction

Organic solar cells (OSCs) have become an intensive area of research with advantages of being flexible, light-weight, inexpensive, environmental friendly and biodegradable [1,2]. Amongst several available material combinations, blend of poly(3-hexylthiophene-2,5-diy) (P3HT) and [6,6]-Phenyl C₆₁ butyric acid methyl ester (PCBM) has become the workhorse of OSC research [3–5] and serves as a model system for characterization techniques. Despite being intensively studied, OSCs suffer from low efficiency and short life time of the devices as compared to their inorganic counterparts. In the recent years, efforts have been made to enhance the device efficiency as well as the life time through the understanding of the governing physical mechanisms [4,6–12]. However, the performance of OSCs still needs to be maximized in order to make them feasible for practical applications. This can be accomplished by exploring the underlying physical phenomena influencing the device performance. Since the efficiency of OSCs is simultaneously controlled by the open circuit voltage (V_{OC}), short circuit current (J_{SC}) and the fill factor (FF), a detailed understanding of each of these parameters becomes crucial. Therefore, in OSCs, these three parameters are intensively researched to understand their dependence on material properties and the device architecture [6,13–18]. Though all three are important and interrelated, the study of V_{OC} is of special importance from the point of view of physics of these devices for a

variety of reasons. The principal motive among them is that it allows monitoring of *loss mechanisms*, especially in the absence of transport of carriers.

The open circuit voltage (V_{OC}) in a typical solar cell is the voltage developed across the device at which the photocurrent is equal to the dark current and the net current flow is zero [19]. In terms of the parameters of *ideal* J-V characteristics of a solar cell, V_{OC} can be expressed as [20]:

$$eV_{OC} = nk_B T \ln \left(\frac{J_{SC}}{J_0} + 1 \right) \quad (1)$$

here n is the ideality factor, J_{SC} is the short circuit current and J_0 is the reverse saturation current under dark condition.

From the band structure point of view, in an ideal organic bulk heterojunction solar cell, V_{OC} is the separation between the respective quasi Fermi levels of electrons and holes [21,22]. Determining the upper limit of V_{OC} for any technology is important since one can estimate and monitor the loss mechanisms of V_{OC} with that limit as the point of reference. Open circuit voltage also depends on the device architecture, specifically on the quality of contact. In fact the above expression in Eq. (1) often assumes that the contacts are perfectly ohmic so that the built-in potential of the diode does not play a role and the contacts are perfectly aligned with the respective quasi-Fermi levels in the illuminated condition. However, in practical devices the metals

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used as anode and cathode may have a barrier at the interface, or may have other interactions with the material.

In addition to V_{OC} , a closely related phenomenon is the accumulation of photogenerated carriers at the interface. Such charge accumulation specifically at the interfaces can generate photovoltage which would contribute to the loss of overall photovoltaic effect [23,24]. Therefore, in addition to intrinsic properties of the material, device parameters such as contact related phenomena can have significant effect on built-in and open circuit voltage, even though contact resistance itself has no role to play in absence of a current. A close look at the role of contacts from the point of view of open circuit voltage is therefore attracting recent attention from many research groups [25–28]. The band bending at the contacts can be influenced by *pinning* of the Fermi level, or accumulation leading to space charge, or even formation of dipolar layer at the metal/polymer interfaces [29,30].

The quality of the interface is closely related to the fabrication processes, especially because phase distribution and morphologies of the blend are crucial determinants. The loss in photovoltage due to such device related issues, and their effect on overall performance of the solar cell has been mostly studied so far by simulation of solar cell characteristics under various assumptions. The loss of open circuit voltage has been attributed to the occurrence of contact limited regime of injection [31]. Solak et al. have recently proposed that non-ohmic barrier at one of the contacts leads to its decreased sensitivity at high intensity [15]. Tessler recently suggested through simulations that if one can avoid accumulation of carriers at the contact, it is possible to increase open circuit voltage by 0.2 V and hence increase in efficiency by almost 40% in bulk heterojunction OPV cells is possible [32]. Most of the ideas relating to electrical characterization of contact related phenomena in organic solar cells are difficult to isolate experimentally, though there have been some studies involving Kelvin probe [33–35] and photoelectron spectroscopy [36,37]. Lee et al. have significantly shown that surface photovoltage as measured by Kelvin probe follows open circuit voltage [24]. Often the models and ideas are tested by fitting $J-V$ characteristics. There have been efforts to obtain qualitative insights from $C-V$ characteristics as well [23,29,30].

Our specific aim in this work is to be able to measure the built-in potential using $C-V$ characteristics under illumination and operating conditions. In the literature, often $C-V$ is used under Mott-Schottky assumptions, which is not valid for undoped organic semiconductors [38]. We specifically use capacitance analysis under drift-diffusion conditions to obtain the surface potential. This is equivalent to measurement of the voltage across the device at which injection of carriers commence. This measurement is significant to the study of electrical models of solar cells and monitoring their performance. Since the *quantitative C-V analysis* allows us to determine the threshold for injection processes, it is possible

- (i) to determine effective built-in potential using capacitance measurements in the forward bias,
- (ii) to monitor surface photovoltage at the contacts under illumination and hence determine the voltage loss due to accumulation, and
- (iii) to compare these results for a conventional BHJ OPV with one having inverted architecture.

We demonstrate the differences between conventional architecture and inverted cell which may be of help in understanding the origin of beneficial factors in inverted cells, which is not very well understood as yet.

2. Experimental details

2.1. Device structure

The structures used in this study are conventional and inverted bulk heterojunction solar cells containing blend of P3HT and PCBM as the

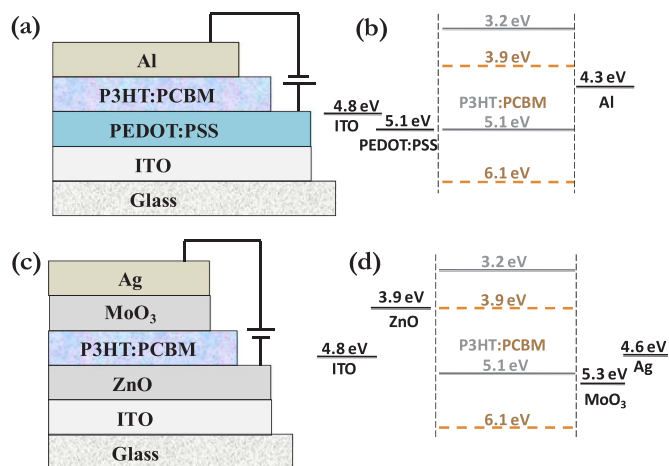


Fig. 1. Device schematic of (a) conventional structure (b) corresponding energy band diagram (c) inverted structure and (d) corresponding energy band diagram.

active layer. Poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) and Molybdenum trioxide (MoO_3) are used as hole transport layers (HTL) in conventional and inverted structures respectively whereas zinc oxide (ZnO) served as electron transport layer (ETL) in inverted structure. The device structure for conventional organic solar cell is ITO|PEDOT:PSS|P3HT:PCBM|Al and for inverted organic solar cell is ITO|ZnO|P3HT:PCBM| MoO_3 |Ag. Fig. 1 shows the device schematic for both the structures along with their corresponding energy band diagram. The light is incident from the anode side in conventional structure while in the case of inverted structure it is from the cathode side.

2.2. Fabrication and characterization

The materials P3HT and PCBM were commercially obtained from Sigma Aldrich. PEDOT:PSS was purchased from Clevis. For the fabrication of conventional organic bulk heterojunction solar cell, the PEDOT:PSS layer is formed on ITO patterned glass substrates by spin coating followed by annealing at 120 °C for 20 min on a hot plate in ambient. PEDOT:PSS solution was filtered using 0.45 μm PVDF filter. It was diluted by mixing deionized water in a ratio of 2:1 to obtain optimized thickness and surface smoothness through spin casting. The thickness of PEDOT:PSS thin film was 35–40 nm approximately. P3HT:PCBM layer is spin coated onto PEDOT:PSS layer and annealed at 140 °C for 20 min on a hot plate in nitrogen (N_2) glove box. Finally, 100 nm thick aluminum (Al) is deposited via thermal evaporation in high vacuum. The blend solution of P3HT:PCBM is prepared by dissolving 20 mg mixture of P3HT:PCBM in 1 ml of Chlorobenzene in a weight ratio of 1:1. The thickness measurement of the solution processed layers is carried out using the stylus based profilometer from Dektak.

In case of inverted structure, zinc oxide (ZnO) layer is formed on ITO patterned glass substrates using solution process and annealed at 250 °C for 10 min on hot plate in ambient, resulting in a thickness of 30 nm. ZnO solution is prepared using sol-gel method [39]. The ingredients for the preparation of ZnO solution were Zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$) as precursor and monoethanolamine ($\text{H}_2\text{NCH}_2\text{CH}_2\text{OH}$) as stabiliser, dissolved in 2-methoxy ethanol ($\text{CH}_3\text{OCH}_2\text{CH}_2\text{OH}$). At first, 0.384 g zinc acetate dihydrate is dissolved in 4.895 ml 2-methoxy ethanol and subsequently 0.105 ml of monoethanolamine is added to the solution. Finally, the solution is stirred overnight at room temperature and filtered using 0.45 μm PVDF filter before being processed. P3HT:PCBM layer is spin cast on top of it and annealed at 140 °C for 20 min on hot plate in N_2 glove box. Subsequently, 5 nm thick Molybdenum trioxide (MoO_3) and 100 nm thick silver (Ag) were deposited using sequential thermal evaporation in high

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