Solar Energy Materials & Solar Cells 111 (2013) 66–73

Contents lists available at SciVerse ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Sensitivity analysis of current generation in organic solar cells—comparing bilayer, sawtooth, and bulk heterojunction morphologies

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ARTICLE INFO

Article history:
Received 28 March 2012
Received in revised form
14 June 2012
Accepted 3 December 2012
Available online 20 January 2013

Keywords:
Sensitivity analysis
Organic solar cells
Heterogeneous microstructure
Drift-diffusion

ABSTRACT

Organic solar cells (OSC) show great potential as a low-cost energy source. In addition, their mechanical flexibility allows the added advantage of use on a wide variety of surfaces. In recent years, progress in experimental strategies and modeling approaches have enabled enhancing the power conversion efficiencies of OSC. In particular, simulation based analysis has played a significant role in improving our understanding of the charge transport phenomena in the active layer of these devices. The excitonic drift-diffusion (EDD) model has been used widely to simulate the generation and transport properties of bulk heterojunction (BHJ) solar cells. The EDD model – which is derived from the Boltzmann transport is dependent on a number of input parameters such as (1) material properties (mobility and permittivity), (2) operating conditions (illumination and device thickness), and (3) active layer morphology.

A comprehensive sensitivity analysis of the short-circuit current, J_{sc}, to the input parameters is performed. This helps in rank ordering the input parameters and operating conditions – by strength and relevance – on their impact on J_{sc}. We particularly focus our investigations on understanding how the active layer morphology affects the sensitivity of J_{sc}. To accomplish this we analyze three classes of morphologies: bilayer, BHJ, and sawtooth. The results show significant differences in sensitivities between BHJ, sawtooth, and bilayer morphologies. Short-circuit current in BHJ structure shows higher sensitivity to material properties than either sawtooth and bilayer structure, suggesting that the necessity for finer control of material properties to counteract the increased disorder in the active layer morphology. The electrode current is found to be most sensitive to illumination intensity for all three morphologies. We report some interesting trends that may help choose the most sensitive parameters to vary for designing OSC's with better performance.

1. Introduction

Organic solar cells allow the use of low-temperature roll-to-roll, and hence low-cost manufacturing [1]. Mechanical flexibility is the other major advantage of these devices which makes them so attractive. Huge progress has been made in the last few years towards improving the problem of low efficiencies associated with organic solar cells. This advancement (highest reported value under laboratory conditions is 9% [2]) is made possible by experimental and numerical investigations of the operating mechanisms and device structure optimization. Numerical simulations not only help to enhance performance through device optimization [3] but also help to improve the fundamental understanding of the underlying charge transfer processes in the active layer which is not easily accessible to experimental tools [4–8].

Monte-Carlo method based microscopic models [9–11] have been utilized to probe the effect of morphology on the generation, mobility and recombination of charge carriers. But the high computational cost associated with microscopic simulations hampers their use for large scale simulations or in high-throughput analysis of OSC. A computationally efficient alternative in the form of excitonic drift-diffusion model [5,12–17] has been used successfully for simulating the charge transport properties of organic photovoltaics. With wide use of EDD equations it has become increasingly imperative to do a detailed investigation to understand how the material properties and operating conditions (inputs to EDD model) effect the device performance (output from EDD model). When input variables are not known or can be changed, sensitivity analysis informs us about the effect of a small change in input parameters on the output from EDD model. Sensitivity

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http://dx.doi.org/10.1016/j.solmat.2012.12.004

analysis can also help in ordering the inputs by their effect on performance of organic solar cells. This facilitates decision making process when one is presented with the option of modifying one among a number of competing input parameters, thus providing guidance on process optimization. Also, it would contribute towards increasing the understanding and credibility of the EDD model and making the results more persuasive. In recent years, such sensitivity analysis has been used extensively for a variety of systems like ecological models [18], chemical models [19], structural systems [20], pharmacokinetic models [21], and fluid flows [22].

For OSC devices, sensitivity analysis of 1D homogeneous model was undertaken by Häusermann et al. [23], wherein the sensitivity to input parameters for a range of device thicknesses, input voltages and time during turn-on was studied. The input parameters selected were electron and hole mobilities μ_n, μ_p, recombination efficiency r_{eff}, exciton pair separation a, and decay rate k_f. The pair separation a was found to have the highest influence on the short-circuit current.

However, experimental evidence suggests that the active layer morphology is an important factor determining the performance of organic solar cells [24–26], which indicates significant effect of morphology on the sensitivity of J_{sc}. This study aims to do an in-depth sensitivity analysis of a morphology dependent EDD model for organic solar cells. We analyze the sensitivity of the short-circuit current to the (a) material properties (electron mobility μ_n, hole mobility μ_p, acceptor dielectric constant ϵ_A, and donor dielectric constant ϵ_D), (b) device thickness (L), and (c) operating condition (illumination intensity Γ) under three different morphologies. To investigate the effect of the transition from ordered to disordered morphologies on sensitivity, we study bilayer, sawtooth, and BHJ based devices. As the sensitivity to a specific input parameter may be dependent on the values of other input parameters, we compute the sensitivity to the inputs for a range of values of all input parameters.

2. Method

2.1. Model

In order to determine the short-circuit current density output from the active layer of a BHJ device, we use the EDD model [5] as described below

$$\nabla \cdot (\epsilon \nabla \phi) = q(n-p) \tag{1}$$

$$\nabla \cdot \mathbf{J_n} = -qff' D_{[\nabla\phi,X]} + qff' R_{[n,p]} \tag{2}$$

$$\nabla \cdot \mathbf{J_p} = +qff' D_{[\nabla\phi,X]} - qff' R_{[n,p]} \tag{3}$$

$$\mathbf{J_n} = -qn\mu_n \nabla\phi + qV_t\mu_n \nabla n \tag{4}$$

$$\mathbf{J_p} = -qp\mu_p \nabla\phi - qV_t\mu_p \nabla p \tag{5}$$

$$\nabla \cdot (V_t \mu_X \nabla X) - ff' D_{[\nabla\phi,X]} - R_{[X]} = -G - ff' R_{[n,p]} \tag{6}$$

Eqs. (1)–(6) are solved to obtain the electron (n) and hole (p) densities, and electrostatic potential ϕ. The solution thus obtained is used to determine the electron and hole current densities, $\mathbf{J_n}$ and $\mathbf{J_p}$ respectively. Here, q is the elementary charge.

Morphology dependent parameters: The values of dielectric constant ϵ, electron mobility μ_n, and hole mobility μ_p are dependent on morphology, i.e. ϵ, μ_n, and μ_p at any location in the active layer depends on the type of material at that location. f and f' denote the parameters which are used to track the donor–acceptor (DA) interfaces. f is 1 at interface, 0 elsewhere. To identify the interfaces with pathways to electrodes – for both electrons and holes – we use parameter f'. f' is 0 at interfaces without direct connection to either cathode (for electrons) or anode (for holes). These interfaces are associated with either of the donor and acceptor regions forming a cul-de-sac. f' is 1 elsewhere.

The exciton generation, diffusion, and dissociation are represented by Eq. (6). G and $D_{[\nabla\phi,X]}$ denote rate of exciton generation and dissociation respectively. Exciton relaxation rate is denoted by $R_{[X]}$. The recombination of charges is given by $R_{[n,p]}$. The expressions for $D_{[\nabla\phi,X]}$, $R_{[n,p]}$, G, $R_{[X]}$ are

$$D_{[\nabla\phi,X]} = k_d X \tag{7}$$

$$R_{[n,p]} = \gamma_r np \tag{8}$$

$$G = \alpha\Gamma \exp(-\alpha x) \tag{9}$$

$$R_{[X]} = X/\tau_X \tag{10}$$

where Γ denotes illumination intensity and x is the distance from the illumination site. Here the exciton generation rate G is approximated using Beer–Lambert Law. The expression for

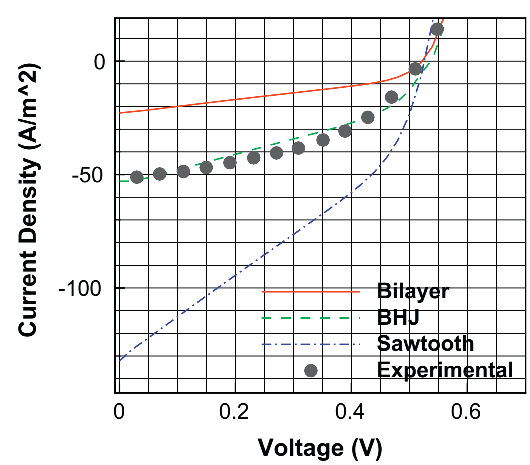

Fig. 2. Current–voltage characteristic plots for the bilayer, sawtooth, and BHJ morphologies compared with result from experiment [27].

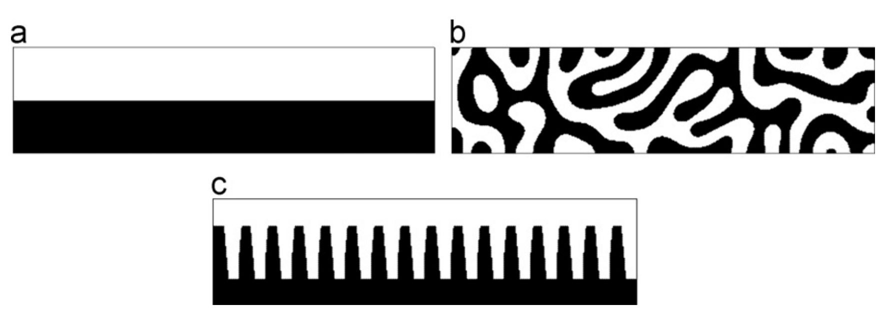

Fig. 1. Morphologies used for the sensitivity analysis. Dark region represents acceptor and lighter region is donor: (a) bilayer, (b) BHJ, and (c) sawtooth.