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Light dependent open-circuit voltage of organic bulk heterojunction solar cells in the presence of surface recombination

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ABSTRACT

An analytical model for the light intensity dependence of open circuit voltage V_{oc} in the presence of bimolecular, trap assisted and surface recombination mechanisms was proposed. The model quantitatively explains reported experimental deviations from the bimolecular and bimolecular/trap assisted recombination models.

V_{oc} was found to be the most sensitive photoelectric parameter to surface recombination. The relative effect of surface recombination on V_{oc} increases with the increase of trap density as well as V_{oc} becomes more sensitive to the presence of deep traps due to surface recombination. In the presence of surface recombination slope V_{oc} vs. light intensity *A* does not reach value of 2.0 kT/q even at very high density of traps.

Also a possible misinterpretation of the experimental V_{oc} vs. light intensity dependences in the presence of trap assisted and surface recombination was outlined.

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1. Introduction

Organic bulk heterojunction (BHJ) solar cells are very attractive from the both fundamental and practical points of view. There are many papers focused on the understanding of the main efficiency limiting factors in BHJ solar cells. The open circuit voltage V_{oc} of organic BHJ solar cells is one of the main photoelectrical parameter, which is still under intensive investigation [1–5].

The expression for the V_{oc} in the case of the pure bimolecular recombination, which explains the experimental light intensity dependence of V_{oc} for different BHJ solar cells was derived a decade ago [6]. Afterward, the effect of trap assisted recombination on V_{oc} was taken into account [7–9]. It was shown that the slope of V_{oc} vs. light intensity dependence $A = \Delta V_{oc}/\Delta \ln[Intensity]$ increases from 1.0 kT/q to 2.0 kT/q with the increase of trap density [7,9]. Therefore, the light intensity dependence of V_{oc} became a widely used simple experimental method for the identification of the dominating recombination processes in organic BHJ solar cells under working conditions.

However, there are a number of experimentally induced reasons for the further and deeper investigation of V_{oc} vs. light intensity dependence for organic BHJ solar cells. The values of *A* for the small molecule BHJ solar cells 3,6-bis(5-(benzofuran-2-yl)thiophen-2-yl)-2,5-bis(2-ethylhexyl)pyrrolo[3,4-c]pyrrole-1,4-dione (DPP(TBFu)$_2$):PC$_{71}$BM and 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) (*p*-DTS(FBTTh$_2$)$_2$):PC$_{71}$BM are smaller than 1.0 kT/q [10] that can not be explained in the scope of the bimolecular recombination model and moreover in the scope of the bimolecular/trap assisted recombination model. This fact is usually suggested to originate from the presence of surface recombination at a BHJ layer/contact interface [11–13], but there is no a quantitative model. At the same time the experimental investigation of V_{oc} vs. light intensity of organic BHJ solar cells, made of a blend of the copolymer poly[N-9″-hepta-decanyl-2,7-carbazole-alt-5,5-(4′,7’-di-2-thienyl-2′,1′,3′-benzothiadiazole) (PCDTBT) and the fullerene derivative PC$_{60}$BM, with the intentionally introduced high density of well-defined [6,6]-phenyl C 84 butyric acid methyl ester (PC$_{84}$BM) traps shows the values of *A* smaller than 2.0 kT/q (maximum slope 1.62 kT/q at the maximum introduced PC$_{84}$BM trap density $N_t \approx 10^{18}$ cm^{-3}) [9]. However, the slope of 2.0 kT/q is theoretically predicted by the bimolecular/trap assisted recombination model in the case of high trap density.

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These deviations from the bimolecular and bimolecular/trap assisted recombination models should be analyzed in details in order to understand other physical processes which may decrease V_{oc} of organic BHJ solar cells and affect on its light dependence.

In this contribution a simple analytical model for the light intensity dependence of V_{oc} in the presence of the bimolecular, trap assisted and surface recombination and shunt resistor is presented and modeled under different conditions.

2. Theoretical part

An organic bulk heterojunction solar cell will be considered in the presence of bimolecular recombination, trap assisted recombination, surface recombination and shunt resistor. The rate of the bimolecular recombination is given as [14,15].

$$R_{BMR} = \frac{q}{\varepsilon\varepsilon_0}\xi(\mu_n + \mu_p)\left(np - n_i^2\right), \quad (1)$$

where ε is the dielectric constant of the BHJ layer, $\varepsilon 0$ is the permittivity of free space, ξ is the Langevin prefactor, μn and μp are the electron and hole mobilities, n and p are the electron density in the conduction band and the hole density in the valence band, respectively, $n_i = np = N_c^2 \exp(-Eg/kT)$ is the intrinsic charge carrier concentration in the BHJ layer, where Nc is the effective density of states, Eg is the band gap of the BHJ layer. The rate of trap-assisted recombination is governed by the Shockley-Read-Hall (SRH) model:

$$R_{SRH} = \frac{C_n C_p N_t (np - n_i^2)}{[C_n(n + n_1) + C_p(p + p_1)]}, \quad (2)$$

where C_n and C_p are the capture coefficients for electrons and holes, respectively, N_t is the density of traps, the values of n_t and p_t are determined by the location of the trap level E_t: $n_t = N_c\exp(-(E_c-E_t)/kT)$ and $p_t = N_v\exp(-(E_t-E_v)/kT)$. The rate of trap assisted recombination is maximized when $E_t = E_i$ (E_i is the mid-gap energy), indicating that only deep energy levels, located near the mid-gap are effective recombination centers. Considering only these deep traps and taking into account that in organic BHJ solar cells under illumination typically $n \approx p$, $np = n^2 >> n_1 p_1 = n_i^2$, the Eqs. (1) and (2) may be reduced to Eqs (3) and (4), respectively [14–16].

$$R_{BMR} = \frac{q}{\varepsilon\varepsilon_0}\xi(\mu_n + \mu_p)n^2 = \gamma_{BMR} n^2, \quad (3)$$

$$R_{SRH} = \frac{C_n C_p N_t}{(C_p + C_n)} n = \frac{q}{\varepsilon\varepsilon_0}\mu_p N_t n, \quad (4)$$

The effect of surface recombination (at BHJ layer/contact interface) can be taken into account through the dimensionless field dependent coefficient $h(V)$ [17–21].

$$h(V) = \left(1 + \frac{S}{\mu_p E}\right)^{-1} = \left(1 + \frac{SL}{\mu_p(V_{bi} - V)}\right)^{-1}, \quad (5)$$

where S is the surface recombination velocity, L is the thickness of the BHJ layer. The current transport and recombination processes in organic BHJ solar cells are considered to be determined by the slowest charge carriers. Thus the hole mobility is put in Eq. (5).

Now, basing on the discontinuity equation for electrons under stationary open circuit condition [6], the following equation can be written in the presence of shunt resistance, bimolecular, trap assisted and surface recombination:

$$PG\left(1 + \frac{SL}{\mu_p(V_{bi} - V_{oc})}\right)^{-1} = (1 - P)\gamma_{BMR} n_{oc}^2 + \frac{q}{\varepsilon\varepsilon_0}\mu_p N_t n_{oc} + \frac{V_{oc}}{qLR_{sh}}, \quad (6)$$

where n_{oc} is the free charge carrier concentration in the BHJ layer under open circuit conditions. At open circuit the drift and diffusion current (photocurrent and injected current) compensate each other. Thus the quasi-Fermi levels for electrons and holes are constant and the energy difference between them is equal to qV_{oc} [6].

$$n_{oc} p_{oc} = n_i^2 \exp\left[\frac{qV_{oc}}{kT}\right], \quad (7)$$

Since the densities of electrons and holes are considered to be equal in organic BHJ solar cells we get the final expression for n_{oc}:

$$n_{oc} = n_i \exp\left[\frac{qV_{oc}}{2kT}\right] = N_c \exp\left[-\frac{E_g}{2kT}\right]\exp\left[\frac{qV_{oc}}{2kT}\right], \quad (8)$$

Eq. (6) may be rewritten in the form of a quadratic equation regarding n_{oc}:

$$a n_{oc}^2 + b n_{oc} + c = 0, \quad (9)$$

where

$$\begin{aligned} a &= (1 - P)\gamma_{BMR} \\ b &= \frac{q}{\varepsilon\varepsilon_0}\mu_p N_t \\ c &= -\left[PG\left(1 - \frac{SL}{\mu_p(V_{bi} - V_{oc})}\right)^{-1} - \frac{V_{oc}}{qLR_{sh}}\right] \end{aligned}, \quad (10)$$

The physically based solution of Eq. (9):

$$n_{oc} = \frac{-b + \sqrt{b^2 - 4ac}}{2a} = \frac{\sqrt{\left(\frac{q}{\varepsilon\varepsilon_0}\mu_n N_t\right)^2 + 4(1 - P)\gamma_{BMR}\left[PG\left(1 + \frac{SL}{\mu_p(V_{bi} - V_{oc})}\right)^{-1} - \frac{V_{oc}}{qLR_{sh}}\right]} - \frac{q}{\varepsilon\varepsilon_0}\mu_n N_t}{2(1 - P)\gamma_{BMR}}, \quad (11)$$

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