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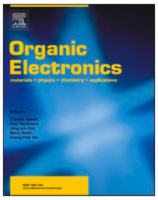

Efficiency limit of excitonic photovoltaic cells under phosphor-based white LED illumination

Grażyna Jarosz [*], Malgorzata Franz, Rafal Marczyński, Ryszard Signerski

Institute of Physics and Applied Computer Science, Faculty of Applied Physics and Mathematics, Gdansk University of Technology, Gdansk, Poland

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ABSTRACT

The limit of energy conversion of excitonic photovoltaic cells working under white light illumination generated by phosphor-based LED is analysed using the modified Giebink approach. Particularly, the impact of the optical energy gap and energy loss associated with the excitons dissociation at the heterojunction interface on power conversion efficiency of the device are discussed. From the results of our study it follows that the optimal optical energy gap value of organic materials equals 1.87–1.91 eV for the cool light and 1.80–1.82 eV for the warm light. The value of maximum power efficiency reaches 50%, if the energy loss related to excitons dissociation at the interface ED/EA is smaller than 0.3 eV and it decreases up to 40%, if the energy loss reaches the value of 0.5 eV. The obtained results reveal the direction of further improvement of efficiency of organic photovoltaic solar cells for indoor applications.

1. Introduction

Films of organic materials are usually characterized by a high value of linear absorption coefficient, exceeding $1{*}10^5$ cm^{-1} in the visible light spectrum, which is related generally to the exciton generation process [1,2]. Taking into account 10^5 cm^{-1} as the linear absorption coefficient, a film of a thickness 230 nm absorbs 90% of the incident radiation. From such results it follows that organic materials could be attractive as an active layer in thin film photovoltaic cells. However, the exciton absorption bands of organic materials cover much less the spectrum of solar radiation in comparison to inorganic semiconductors (e.g. Ref. [3,4]). In Fig. 1 the normalized absorption spectra for typical films of small-molecule organic materials commonly used in photovoltaics are shown (upper part of the figure) together with the spectrum of photon flux for the solar radiation AM 1.5G (bottom part of the figure). It is worth noticing the limited width of the exciton absorption bands which is much smaller than the width of the solar radiation spectrum. On the other hand, as it could be seen from Fig. 1, the absorption bands of organic materials could easily cover the solar radiation spectra in the visible range (see shaded area in Fig. 1), which is emphasized in many publications (e.g. Refs. [3–13]). Hence we can suspect that organic materials, from which thin, light and flexible layers could be produced in low temperature manufacture process, can successfully compete with inorganic semiconductors in the range of thin film photovoltaic devices devoted especially for indoor applications [3,5,11,12,14].

Efficient organic photovoltaic cells require the introduction of the electron donor-electron acceptor (ED-EA) heterojunction. This requirement follows from the fact that excitons generated in organic materials (Frenkel-Davydov excitons) are tightly bound and do not spontaneously dissociate into electron-hole pairs. Excitons due to their diffusive motion could reach the interface of the ED/EA heterojunction, where their dissociation can occur and the charge-transfer (CT) complexes are formed with the electron located on the EA molecule and hole on the ED molecule [1,2,17–23]. With this process additional energy loss is connected, which occurs in organic photovoltaic cells and is not observed for classical inorganic photovoltaic cells. Therefore it is commonly accepted that the maximum value of the power conversion efficiency of single junction photovoltaic cell must be lower than predicted by the Shockley-Queisser (SQ) limit, which was derived for inorganic photovoltaic cells using the principle of detailed balance [24]. In 2011, Giebink et al. have proposed a new approach for evaluating the efficiency limit of excitonic solar cells based on the Second Law of Thermodynamics [17]. According to Giebink, the efficiency limit of organic solar cells lies in the range from 22% to 27%, by taking into account the intrinsic energy losses to dissociate excitons at the interface ED/EA in the range 0.3–0.5 eV. From papers published in recent years it follows that the power conversion efficiency in organic solar cells has rapidly improved, exceeding the crucial efficiency of 15% (e.g. Ref. [25]). Such

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[*] Corresponding author.
E-mail address: grajaros@pg.edu.pl (G. Jarosz).

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result of the power conversion efficiency indicates significant progress, which has been done in the field of organic solar cells, as this is already exceeding a half of the upper limit of energy conversion expected for excitonic solar cells.

From the fact that absorption spectra of organic materials could overlap much better with the visible light spectrum than they cover the solar radiation spectrum AM 1.5G, we predict that organic photovoltaic cells could work under illumination produced by artificial light sources and serve for indoor application (e.g. as power supply in wireless sensors, mobile devices or circuits integrated with living organisms [5,12,14]). Furthermore, we suspect that indoor organic cells could approach faster their theoretical limit of energy conversion than solar cells. However, to the best of our knowledge, the limit of the efficiency of organic photovoltaic cells for indoor applications has not been estimated yet. The values of efficiency of organic photovoltaic cells, determined under solar radiation as well as artificial radiation, were commonly analysed accordingly to the theory of Shockley-Queisser, which was derived for inorganic photovoltaic cells (e.g. Ref. [3,14,26–28]). Therefore it is worth finding the limit of the efficiency of excitonic photovoltaic cells for indoor applications working under artificial light sources.

In this paper, we investigate the limit of energy conversion for excitonic photovoltaic cell working under white light illumination generated by phosphor-based LED using the modified approach of Giebink et al. The impact of the optical energy gap and energy losses associated with the dissociation of excitons at the interface between ED and EA are discussed. The obtained results reveal the direction of further improvement of efficiency of organic solar cells operating under indoors conditions.

2. Theoretical model

Unlike in most inorganic semiconductors, excitons generated in organic materials usually do not dissociate spontaneously into an electron and hole pair due to their high binding energy up to even 1.0 eV. Therefore a common strategy is the right choice of organic materials that form the ED/EA junction in order to receive convenient energy structure, allowing dissociation of excitons that reach the interface and creating CT complexes with an electron placed on EA and a hole on ED materials, respectively. With the formation of CT complexes, which are precursor states to free charge carriers, energy dissipation and additional energy loss processes are connected, which limits the energy conversion efficiency of excitonic photovoltaic cells.

It can be assumed that the additional energy loss is equal to the difference between the optical energy gap and the energy difference between the hole transporting band in ED and the electron transporting band in EA. The optical energy gap in organic materials is the edge of exciton absorption band (E_x). The difference between energy of the hole transporting band in ED and the electron transporting band in EA could be connected with the interfacial CT state energy, which is called by Giebink as the energy of a bound polaron pair (BP), and denoted by E_{BP}. The binding energy of BP is significantly smaller than that of the bulk excitons in organic materials. Hence the energy loss related to BP state dissociation could be neglected [17]. Thus the additional energy loss in excitonic photovoltaic cell ($-\Delta G_{CT}$) can be expressed as: $-\Delta G_{CT} = E_x - E_{BP}$. Here it is worth pointing out that a necessary condition which needs to be fulfilled to observe efficient exciton dissociation at the interface ED/EA is a proper energy gaps between energy levels of the LUMO and HOMO, both in ED and in EA, which need to be bigger than E_{BP}.

Following Giebink et al. we assume that for an ideal organic photovoltaic cell the bulk absorptivity $\alpha(E)$ is unity above the exciton energy gap (E_x) and significantly lower in the region between E_{BP} up to E_x. The bulk absorptivity of the cell equals unity as the exciton generation occurs in the bulk of organic materials. Since the BP is an interface state, its absorptivity is lower than that of the bulk exciton transitions. According to Giebink et al. [17] and Gruber et al. [23] the value of $\alpha_0 = 10^{-3}$ as typical of the range between BP and bulk exciton absorption is mentioned. However, it has to be pointed out that it is rather practical assumption since the absorptivity of bound pairs remains negligible in a realistic cell in which the bulk absorptivity is large [17]. Using this approach, the absorptivity of the ideal photovoltaic cell could be represented as follows:

$$\alpha(\hbar\omega) = \begin{cases} 0 & \text{for } \hbar\omega < E_{BP} \\ \alpha_o & \text{for } E_{BP} \le \hbar\omega < E_x \\ 1 & \text{For } \quad E_x \le \hbar\omega \end{cases} \tag{1}$$

where ħω is the photon energy and $\alpha_o = 10^{-3}$ following Giebink.

For an ideal single-junction photovoltaic cell, it is assumed that only radiative recombination process takes place in the device. Thus the

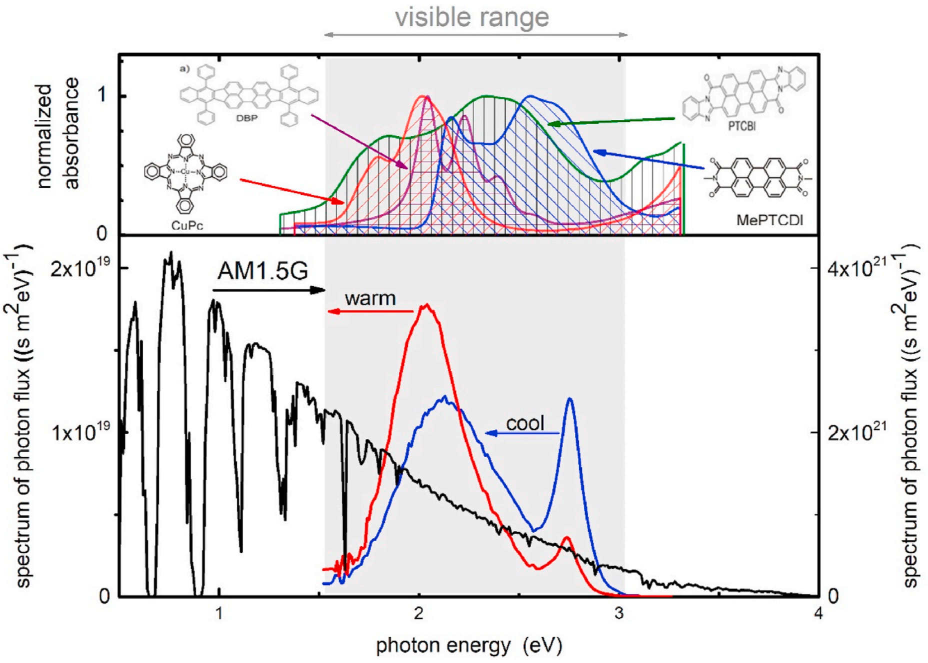

Fig. 1. Normalized absorption spectra of thin films of organic materials (at the top): on the left side examples of electron donor materials and on the right representatives of electron acceptor materials are placed respectively. Below the spectra of photon flux for the solar radiation AM 1.5G (black line) [15] and two phosphor-based LED bulbs emitting cool (blue line) and warm (red line) white light with a correlated colour temperature (CCT) of 6500 K and 3000 K respectively and luminous efficacy of 103 lm/W and 113 lm/W, respectively. The presented spectra of photon flux for the phosphor-based LED bulbs were taken from Ref. [16] and transformed to illuminance of 1000 lx according to the procedure described in details in Ref. [29]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

current-voltage relationship could be represented by a formula:

$$J = e\, I_{R0}\left[\exp\left(\frac{eV}{kT_C}\right) - 1\right] - e\, I_G \tag{2}$$

where J is the current density per unit area of the cell, e is elementary charge, k is Boltzmann constant, T_c is the cell's temperature and V is the potential difference between the cell's terminals. The current generated by sample irradiation (eI_G) results from the electron-hole pair generation, which is directly determined by the sample absorptivity:

$$I_G = \int_0^{+\infty} \alpha(\hbar\omega)^* J_{ph,\hbar\omega}\, d(\hbar\omega)\ , \tag{3}$$

where $J_{ph,\hbar\omega}$ is the energy spectrum of photon flux falling on the unit area of the cell. The value of the current eI_{R0} results from the thermal equilibrium condition between the cell and the environment. According to the detailed balance principle and under the assumption that the rear surface of the cell is isolated from the environment, the rate of radiative recombination assigned to a unit area of the cell at the temperature T_c, takes the form:

$$I_{R0} = \frac{\pi}{4\,\pi^3\,\hbar^3 c^2}\int_0^{+\infty} \alpha(\hbar\omega)^* \frac{(\hbar\omega)^2}{exp\left(\frac{\hbar\omega}{kT_C}\right) - 1}\, d(\hbar\omega) \tag{4}$$

In contrast to Shockley and Queisser model [24], in which the cell exchanges thermal radiation by top and rear surfaces, Eq. (4) is valid when the rear surface of the cell is thermally isolated and it does not participate in the exchange of thermal radiation with the environment. This condition can be easily met experimentally, hence it is often used in calculations of the efficiency of photovoltaic cells, e.g. Henry [30], Rühle [31], Würfel [32], Seki [18], Giebink [17] and Gruber [23].

According to the convention previously introduced (see Eq. (2)) the values of the photovoltaic current are negative, hence the maximum energy conversion efficiency must be determined by the formula:

$$\eta = \frac{MAX(-J\ \times V)}{P_{in}} \tag{5}$$

$$P_{\ in} = \int_0^{+\infty} \hbar\omega^* J_{ph,\hbar\omega}\, d(\hbar\omega)\ , \tag{6}$$

where the function *MAX(argument)* returns the maximum value of the argument and P_{in} is the power of the incident radiation per unit cell's surface.

3. Results and discussion

Nowadays, the artificial light sources market is dominated by phosphor-based LEDs. That follows from their exceptionally high luminous efficacy, reaching over 100 lumen/W. The spectra of phosphor-based LEDs have been used in our simulations and they are represented in the lower part of Fig. 1. Particularly those are spectra of two lamps, namely the first one with luminous efficacy of 113 lm/W and CCT of 3000 K (warm white light) and the second one with luminous efficacy of 103 lm/W and CCT of 6500 K (cool white light). The spectra of those lamps were adopted from Burattini et al. [16] and we transform them for our simulations so that they correspond to illuminance typical for indoor conditions which is from 100 lx (e.g. corridor) up to 1000 lx (e.g. the illumination of the desk). The details of the procedure used to change radiometric quantities to photometric quantities have been decribed in our previous work [29].

In Fig. 2 the power conversion efficiency of excitonic photovoltaic cells under black-body radiation that imitates solar radiation and under phosphor-based LED lamps have been represented. Please notice that values of parameters, of black body and of excitonic cells, used in the calculations are the same as those published in Ref. [17]. Grey arrows show the direction of the change in the device's performance resulting from the increase of -ΔG_{CT} from 0.1 eV, through 0.3 eV up to 0.5 eV. It can be clearly seen that under artificial light illumination, the excitonic photovoltaic cells can reach higher values of energy conversion efficiency than in the case of illumination by solar radiation. The value of maximum efficiency under small energy loss due to exciton dissociation (~0.1 eV) exceeds even 50% both for the warm light as well as for the cool light (see Table 1). The optimal value of the optical energy gap of organic materials should be higher than for solar radiation and equal 1.80 eV for the warm light and 1.87 eV for the cool light. Such absorption edges refer to visible light spectrum and hence seem to be much more easily to meet in organic materials than the absorption edges within near IR light. Furthermore it is worth noticing that even when the energy loss (related to exciton dissociation) is higher (e.g. -$\Delta G_{CT} = 0.5$ eV) the maximum energy conversion still stays high ~40%. However, as it follows from Fig. 2, to apply it in real device, the value of the optical energy gap of organic materials should be slightly higher than 1.80 eV for the warm light and than 1.87 eV for the cool light. Thus, it can be concluded that materials whose absorption edge falls on near IR light are not suitable for application in photovoltaic cells working under artificial light illumination.

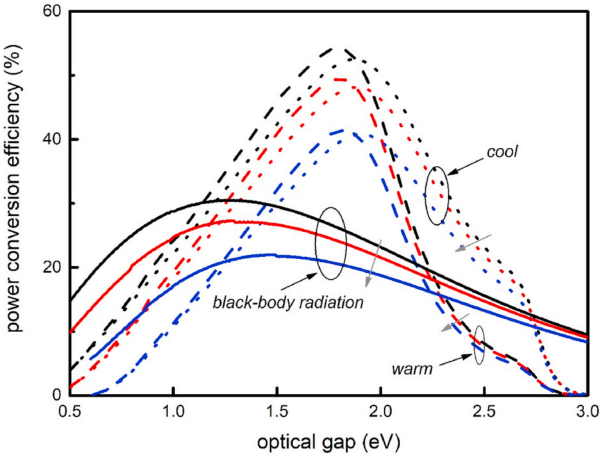

Fig. 2. The power conversion efficiency of excitonic photovoltaic cell as a function of exciton optical energy gap under black-body radiation for a temperature of 5778 K and for a solid angle subtended by the body of 6,822*10^{-5} sr (solid lines) and white light illumination produced by phosphor-based LED lamps with the illumination of 1000 lx (dashed lines refer to warm light and dotted lines to cool light, respectively). Grey arrows show the direction of the change in performance resulting from the increase of -ΔG_{CT} from 0.1 eV (black curves), through 0.3 eV (red curves) up to 0.5 eV (blue curves). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

In Fig. 3, the influence of the illuminance on the power efficiency is shown. It can be seen that reducing light illumination causes only a small (2–4%) decrease of the efficiency, however the value of an optimal optical energy gap does not change. The energy gap should be slightly bigger for photovoltaic cells for which the energy loss at the interface due to excitons dissociation is bigger. Namely, for the warm light an optimal value of the optical energy gap is 1.80 eV at -$\Delta G_{CT} = 0.1$ eV and 1.82 eV at -$\Delta G_{CT} = 0.5$ eV and for the cool light is 1.87 eV at -$\Delta G_{CT} = 0.1$ eV and 1.91 eV at -$\Delta G_{CT} = 0.5$ eV, respectively (see Table 1).

Currently the most efficient excitonic photovoltaic cells working under artificial light produced by fluorescence bulbs reach the values of efficiency up to 13.4% using PV2001:PCMB [33], 16.5% using PCDTBT: PC$_{71}$BM interface [4], 26.2% using TPD-3F:IT-4F [34] and using CD1: PBN-10 [35], 28% using BTR:PC71BM [11], and photovoltaic devices working under LED illumination reveals efficiency up to 9.5% using

Table 1

Maximum power conversion efficiency (η_{max}) and optimal optical energy gap ($E_{optimal}$) predicted for an ideal excitonic photovoltaic cell for different values of the energy loss at EA/ED interface for illuminance of 1000 lx.

$-\Delta G_{CT}$ (eV)		0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
warm	η_{max} (%)	54.2	53.1	49.4	45.4	41.4	37.4	33.5	29.7	26.0
	$E_{optimal}$ (eV)	1.80	1.79	1.81	1.81	1.82	1.84	1.85	1.85	1.86
cool	η_{max} (%)	52.5	51.5	48.1	44.4	40.8	37.2	33.6	30.2	26.7
	$E_{optimal}$ (eV)	1.87	1.87	1.88	1.89	1.91	1.92	1.94	1.95	1.97

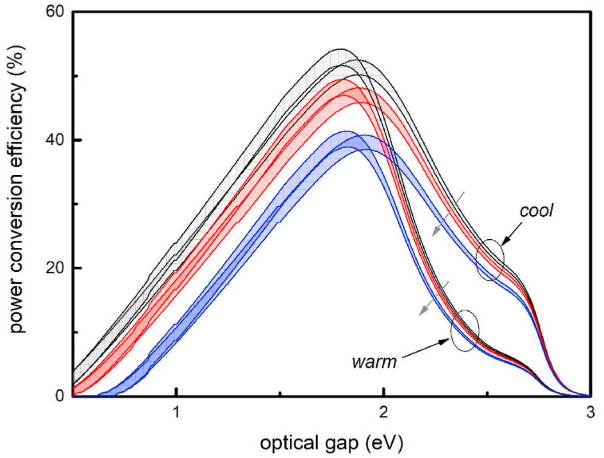

Fig. 3. The power conversion efficiency as a function of exciton optical energy gap under white light from phosphor-based LED lamp for different values of illumination from the range of 100 lx up to 1000 lx (shaded area). Grey arrows show the direction of the change in efficiency resulting from the increase of $-\Delta G_{CT}$ from 0.1 eV (dark region), through 0.3 eV (red region) up to 0.5 eV (blue region). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

P3HT:ICBA [36], 12.8% using P3HT:PC60BM [3], 13.0% using PV2001:PCMB [33], 16% using PPDT2FBT:PC70BM [37] and PCDTBT:PC71BM [4], 19.3% using 1DTP-ID:PNP [38] and 21.0% using PCE10:PC70BM [3], 22.8% using PBDB-TF:IT-M [39], 26.1% using PBDB-TF:IO-4Cl [40] and 26.4% using PBDB-T:ITIC-Th:PC71BM [41]. Many parameters have an influence on the performance of real excitonic photovoltaic cells. Apart of the absorption coefficient for example the layers morphology, charge carriers transport or the electrodes can play a role. To get more details about the records of real devices it is worth reading the last reviews such as [42–44]. The discussion about parasitic components can be also found in these works.

4. Conclusions

Based on Giebink et al. and taking into account the principle of detailed balance, the efficiency limit has been determined for excitonic organic photovoltaic cells working under illumination of white light from phosphor-based LED lamps with luminous efficacy 100 lm/W. From the results of our study it follows that the optimal optical energy gap value for organic materials equals 1.87 eV for the cool light and 1.80 eV for the warm light. The value of maximum power efficiency reaches 50%, if the energy loss related to excitons dissociation at the interface ED/EA is smaller than 0.3 eV and it decreases up to 40%, if the energy loss reaches the value of 0.5 eV. Such high values of the energy conversion efficiency refer to the excitonic devices with single ED/EA junction under illumination of artificial light produced by phosphor-based LED. So we can say it is possible to fully utilize the basic advantages of organic photovoltaic cells such as lightness, flexibility, high linear absorption coefficient in the visible light spectra and low production costs in indoor applications.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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