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SIMULATION OF THE INFLUENCE OF THE ABSORPTION WINDOW FOR STACKED AND MONOLITHIC ORGANIC TANDEM SOLAR CELLS

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ABSTRACT

In order for organic bulk heterojunction solar cells to compete with the traditional inorganic cells, higher power conversion efficiencies are desirable. A way to improve the efficiency is to use a tandem configuration. In this article, we study the theoretical efficiency potential of organic bulk heterojunction tandem solar cells. We study the influence of the energy levels of donor and acceptor, as well as different absorption windows of the subcells for both a stacked and a monolithic configuration. Ideal material characteristics are obtained from these calculations, giving an idea of how the ideal organic tandem cell should look like. An interesting result shows that it would not pay off to develop photovoltaic organic materials with an absorption window broader than 400 nm, because hardly any efficiency gain can be achieved by a broader absorption window. The optimal bandgaps with a sufficient absorption window of 400 nm are E_{g1} = 1.8 eV and E_{g2} = 1.1 eV for both configurations. Furthermore, for a stacked organic tandem cell, both subcells need not necessarily a large absorption window. This does not apply for the monolithic cell. As soon as one subcell has a low absorption window, the efficiency decreases rapidly.

Keywords: organic solar cells; modelling; tandem solar cells; power conversion efficiency; optimal energy levels; absorption window;

1. INTRODUCTION

Photovoltaic solar cells based on organic compounds are promising candidates for solar energy conversion. They have the potential for cost effectiveness, mechanical flexibility and easy processing. Nowadays, efficiencies up to 5 % are reached for single junction cells [1]. However, in order to compete with the traditional inorganic cells, power conversion efficiencies above 10 % are desirable.

A characteristic of organic solar cells is their narrow absorption window, compared to the absorption band of inorganic semiconductors. A possible way to capture a wider band of the solar spectrum - and thus increasing the power conversion efficiency - is using two solar cells

with different bandgaps in a row, referred to as a tandem solar cell. The absorber of the first single solar cell in such a tandem cell has a large bandgap E_{g1}. High-energy photons with an energy $h\nu > E_{g1}$ are absorbed by the first cell. The second cell, with a lower bandgap E_{g2}, absorbs the low-energy photons with an energy between E_{g1} and E_{g2} (Figure 1). In this configuration, the photon energy is used more efficiently: the voltage at which electrical charge is collected in each subcell is closer to the energy of the photons absorbed in that subcell. The row can be extended with more single cells, i.e. a multi-junction solar cell.

Experimental and commercial tandem solar cells are usually of the monolithic (integrated or 2-terminal) type (Figure 1b). This configuration will never reach an efficiency that is higher than that of a stacked (4-terminal) tandem cell, because both single cells cannot be operating at their optimal working point at the same time (unless they have an equal maximum-power current).

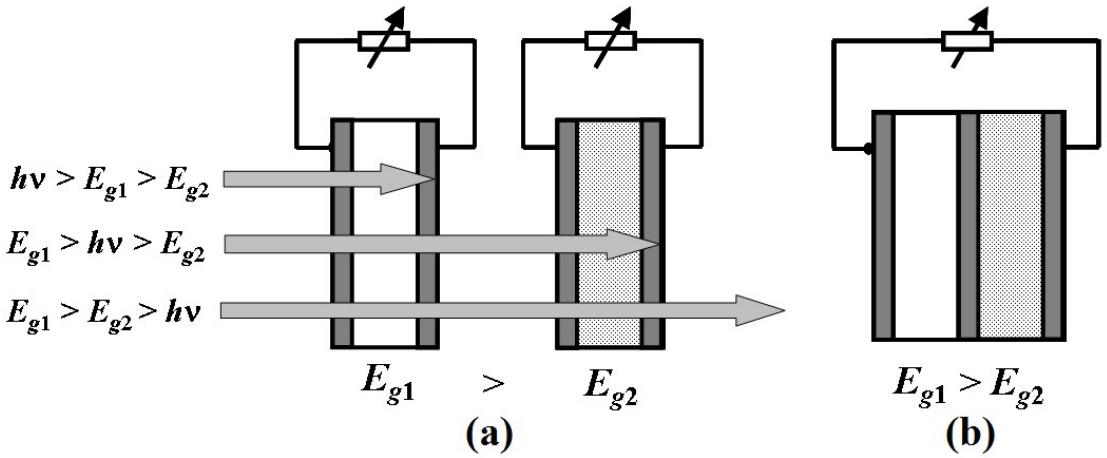

Figure 1: (a) A stacked or 4-terminal tandem solar cell: the first single cell absorbs photons with an energy higher than E_{g1}. The second cell absorbs photons with an energy between E_{g1} and E_{g2}. Photons with an energy below E_{g2} are not absorbed. The two subcells are electrically separated. (b) A monolithic or 2-terminal tandem solar cell: the single cells are electrically connected in series.

Organic tandem solar cells, where both single cells are of the organic solar cell type, have already been fabricated by several research institutes [2, 3, 4, 5], as well as fully organic multi-junction cells [6]. The efficiency of these cells hardly achieves the maximal efficiency of a single organic cell. Nowadays, efficiencies of more than 6 % are reached for organic tandem cells [7].

In this article, we calculate the theoretical upper-limit for the efficiency of organic tandem cells. Although this maximum efficiency itself is only interesting from a theoretical point of view, the ideal material characteristics obtained from these calculations can give us an idea of how the ideal organic tandem solar cell should look like. Previous work on predicting the efficiency of organic (tandem) solar cells has been done by multiple authors, e.g. [8, 9, 10, 11, 12, 13]. In this work, we include the influence of among others different absorption windows for each subcell. Moreover, our calculations are not only presented for a stacked organic tandem cell, but also for a monolithic organic tandem cell. The results presented in this paper are meant to increase the fundamental understanding of the relation between on the one hand the energy levels of donor and acceptor and the absorption window of the subcells, and on the other hand the light harvesting potential of the configurations.

2. ASSUMPTIONS

The active material in a single organic bulk heterojunction solar cell consists of an interpenetrating network of an *n*-type (electron acceptor, e.g. fullerene derivatives) and a *p*-type (semi)conductor (electron donor, e.g. conjugated polymer), sandwiched between two electrodes with different work functions. The optical bandgap E_g is defined as the difference between the lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) of the absorber material.

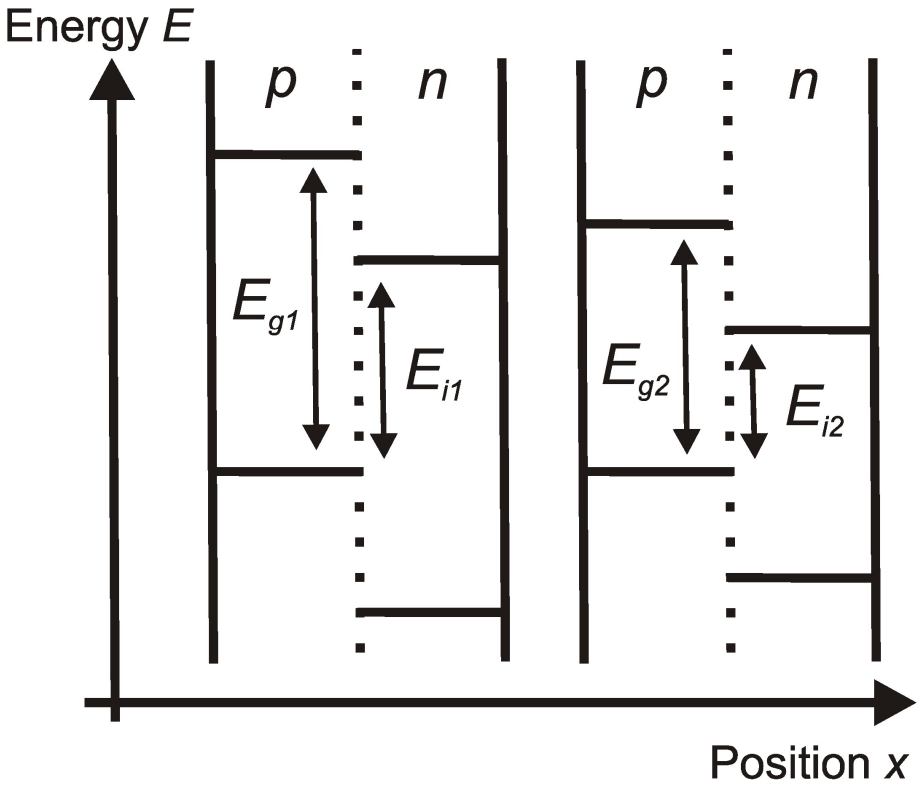

Figure 2: The schematic energy band diagram of a stacked organic tandem solar cell. Only the p-material is the absorber. The mutual position of the single cells does not matter, because the cells are only optically and not electrically connected in series. The absorber bandgap E_g and the interface bandgap E_i of each subcell are indicated.

We consider a 4-terminal tandem solar cell, consisting of two single organic photovoltaic cells (see Figure 2 for the schematic energy band diagram). We assume that in each single cell, only one material absorbs light. Usually, most of the light is absorbed by the *p*-type component; this is the case we will consider here onwards. In the other case, when the *n*-type material absorbs all light, the results remain the same by permutation of *n* and *p* [8]. Because we assume full absorption in each subcell, we neglect interference and optical coupling of the subcells, thus overestimating the efficiency potential. The organic cell with the widest absorber bandgap is at top (at the side of the sun), thus $E_{g1} > E_{g2}$. The distance between the HOMO of the *p*-type (donor) and the LUMO of the *n*-type (acceptor) is considered as the thermodynamic limitation of the useful energy [14]. We call this value the interface bandgap E_i. For an organic solar cell with ohmic contacts, the open circuit voltage V_{oc} is linearly dependent on the interface bandgap E_i. This linear relationship was proven for the variation of

the HOMO level of the donor [9, 15, 16] and of the LUMO-level of the acceptor [17, 18, 19]. For a cell with non-ohmic contacts, the V_{oc} is dependent on the work function difference of the electrodes [20]. In these calculations, we assume a cell with ohmic contacts.

For our simulation, the following fundamental assumptions are made about the stacked tandem cell (Figure 1a): (i) every photon with an energy $h\nu$ higher than the bandgap E_{g1} is absorbed by the first cell and leads to a useful energy E_{i1}. This assumption implies that each absorbed photon eventually leads to a free electron and a free hole, with an energy difference of E_{i1} between them. (ii) every photon with an energy $h\nu$ between E_{g1} and E_{g2} is absorbed by the second cell and leads to a useful energy E_{i2}. (iii) photons with an energy $h\nu$ lower than E_{g2} are fully transmitted. The maximum efficiency η_{max} is therefore given by:

$$\eta_{\max} = \frac{E_{i1}\int_{E_{g1}}^{\infty} N(E)\,dE + E_{i2}\int_{E_{g2}}^{E_{g1}} N(E)\,dE}{\int_{0}^{\infty} E\,N(E)\,dE}, \quad with \quad E_{g1} > E_{g2} \tag{1}$$

with $N(E)$ the incident photon flux. For all our simulations, we use the AM 1.5 experimentally measured solar spectrum [21]. Note that the denominator is the total incident photon power density of the solar spectrum and does not depend on any bandgap. In this ideal scenario, the open circuit voltage V_{oc} of the first and second subcell will be given by E_{i1}/q and E_{i2}/q respectively (with q the electric charge). The fill factor FF of both subcells is assumed to equal unity, as well as the external quantum efficiency EQE of the first cell for wavelengths below the cut-off wavelength λ_{g1} (corresponding with E_{g1}, see Figure 3). The EQE of the second cell equals unity for wavelengths between cut-off wavelength λ_{g1} and λ_{g2} (corresponding with E_{g2}). In real materials, however, the optical absorption and hence the EQE are confined to a more or less narrow wavelength range, usually about 200 to 300 nm wide. We idealize this behaviour by introducing the concept of absorption windows [8], which are defined in Figure 3, and will be treated further as a parameter.

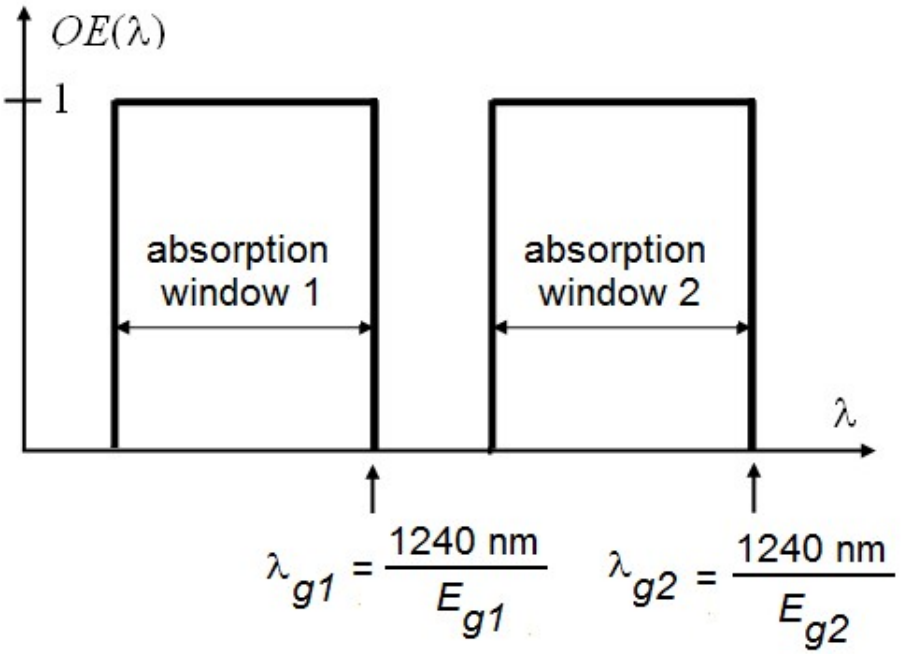

Figure 3: External quantum efficiency (EQE) as a function of the wavelength λ. Definition of the absorption window and the cut-off wavelengths λ_g. The absorption window of the second subcell ("absorption window 2") maximally extends to the cut-off wavelength λ_{g1} of the first subcell. Notice that the order of the first and second subcell can be changed if there is no overlap between both absorption windows.

In a monolithic or integrated tandem solar cell (Figure 1b), the individual cells are electrically connected in series. This means that the total voltage over the cell is the sum of the voltages over each individual cell, and thus equals the sum of the interface bandgaps of both single cells. Furthermore, the same current flows through both single cells. Hence, the maximum efficiency η_{max} for a monolithic organic tandem cell is given by

$$\eta_{\max} = \frac{(E_{i1} + E_{i2}) \cdot \min\left(\int_{E_{g1}}^{\infty} N(E)\,dE, \int_{E_{g2}}^{E_{g1}} N(E)\,dE\right)}{\int_{0}^{\infty} E\,N(E)\,dE}, \quad with \quad E_{g1} > E_{g2} \tag{2}$$

with min(*x*,*y*) the minimum of *x* and *y*. The open circuit voltage V_{oc} of the whole monolithic tandem cell will be given by $(E_{i1}+E_{i2})/q$, the fill factor *FF* equals unity, as does the external quantum efficiency *EQE* for wavelengths below than the cut-off wavelength λ_{g2}.

In organic bulk heterojunction solar cells, light absorption does not immediately lead to free charge carriers. Instead, an exciton is created. In an ideal scenario, the highest efficiency is reached when the LUMO of the *p*-material is as close as possible to the LUMO of the *n*-material [8]. However, a necessary condition for efficient dissociation of the created excitons is that the difference between the LUMOs of donor and acceptor (ΔLUMO) is higher than the exciton binding energy [22]. Thus, without a sufficient energy difference between the LUMOs of both materials, the solar cell cannot work. The value of the exciton binding energy (and the minimal ΔLUMO) in different materials is a subject of discussion, and values in a large range from 0.1 eV to 2 eV have been published [17, 19, 23, 24]. The excess of this

necessary minimum of the LUMO-difference corresponds with an energy loss. Figure 4 shows how the optimum efficiency decreases for increasing ΔLUMO for different absorption windows. With a full absorption window, each additional difference of 0.1 eV between the LUMOs results in approximately an additional 10 % relative efficiency loss in the maximum attainable efficiency. In the following calculations, we assume a difference of 0.2 eV between the LUMOs of our organic solar cell. This value was put forward as an empirical threshold necessary for exciton dissociation [25]. Just because of this necessary energy difference between the LUMOs, the attainable efficiency for the organic bulk heterojunction tandem solar cell drops by 16-17 % in comparison with their inorganic counterpart, purely because of the difficulties in exciton dissociation. Choosing another value for ΔLUMO would lead to qualitative similar results.

In the next section, first we discuss the results for the case where both subcells of the tandem cell have a maximum absorption window. Second, we consider the case where both subcells have the same, narrow absorption window. Finally, an organic tandem cell with different absorption windows for both subcells is discussed.

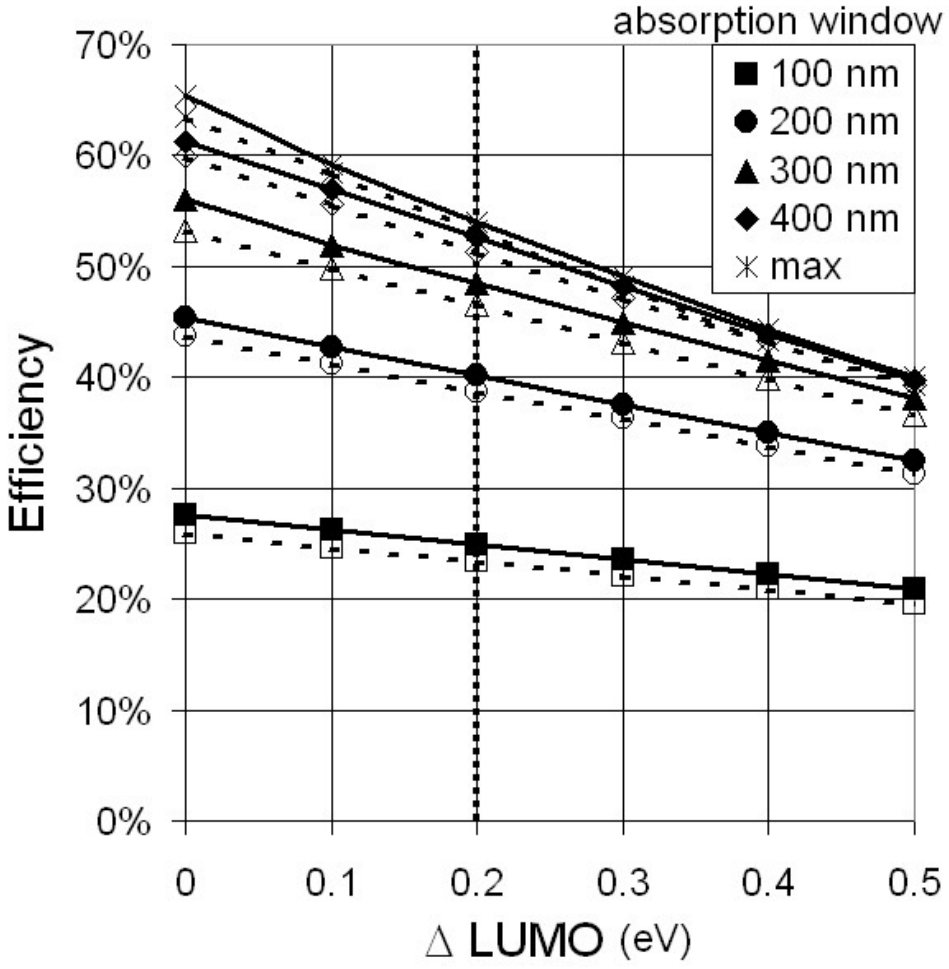

Figure 4: The maximum efficiency η_{max} for different absorption windows of a tandem cell in an ideal scenario as a function of the difference between the LUMOs of the p- and n-material, necessary for exciton dissociation. The solid line indicates the stacked configuration, the dashed line the monolithic configuration. The vertical dotted line indicates the empirical threshold of 0.2 eV.

3. RESULTS

3.1. Subcells with a full absorption window.

Figure 5 shows the maximum efficiency in the ideal scenario for a stacked and monolithic organic tandem cell with bandgaps E_{g1} and E_{g2}, a full absorption window for the subcells and a LUMO difference of 0.2 eV between *n*- and *p*-type. A maximum efficiency of 54.0 % and 53.3 % is reached for a stacked and monolithic tandem cell respectively. As mentioned already, the efficiency of a monolithic configuration will never be higher than that of a stacked configuration. In comparison with a single junction organic cell [8] with an optimal bandgap of 1.1 eV, adding a second subcell results in a relative gain of about 1/3rd in power conversion efficiency. For higher bandgaps, less photons are being absorbed from the solar spectrum, but the useful output energy of each absorbed photon is higher. Hence, there is an optimum for each bandgap. This maximum occurs for the stacked and monolithic tandem cell at a configuration (E_{g1}, E_{g2}) of (1.7 eV, 0.9 eV) and (1.6 eV, 0.9 eV) respectively.

The requirements for a close to optimal configuration of the stacked tandem cell are quite broad, permitting that the values of the bandgaps for optimal cells are not that strict. This is not the case for the monolithic configuration; especially the value of the bandgap E_{g1} of the first subcell is more critical than for a stacked cell.

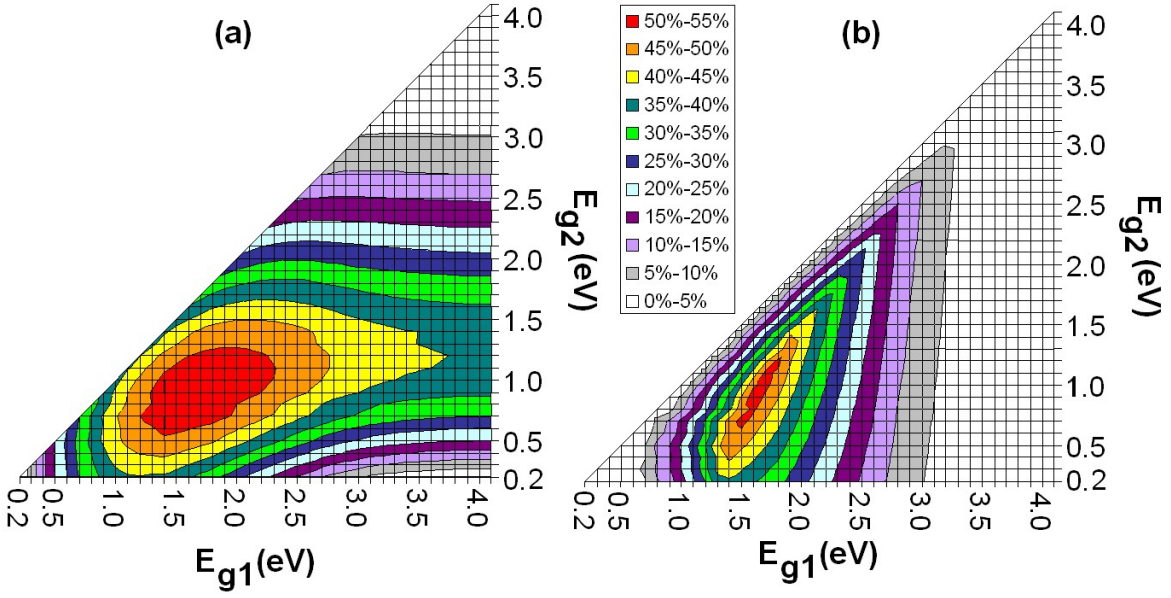

Figure 5: The maximum efficiency η_{max} in the ideal scenario for a stacked (a) and monolithic (b) organic tandem cell with bandgaps E_{g1} and E_{g2}, a full absorption window and a LUMO difference of 0.2 eV between n- and p-type.

Most organic absorbers have a wide bandgap and the production of suitable organic absorbers for photovoltaic applications with a narrow bandgap is problematic [26]. If we consider an organic cell with bandgaps E_{g1} = 2.5 eV and E_{g2} = 1.5 eV, the stacked cell still has a maximum efficiency of 43.6 %, whereas the monolithic cell only reaches 22.4 %. We may conclude that a monolithic tandem cell is much less efficient than a stacked cell in a non-optimal bandgap configuration. For an optimal bandgap configuration, however, the difference is negligible. This means that for the production of tandem cells, the choice of good

bandgap combinations (and thus material combinations) is much more limiting for a monolithic configuration than it is for a stacked configuration.

The current extracted from the monolithic tandem configuration is almost equal to the photocurrent of the subcell that generates the lowest current. If one subcell generates much more current than the other subcell, the excess of charge carriers cannot recombine at the intermediate contact between the subcells. This will cause a charging at the intermediate contact and will partially compensate the built-in voltage across the other cell until the current of both subcells matches. This will lower the power conversion efficiency and explains the inferior performance of monolithic cells for non-optimal bandgap configurations. Current matching is therefore necessary in a monolithic configuration. We want to stress that this effect is not implemented in the model presented in this paper.

3.2. Subcells with the same absorption window.

We now take into account the narrow absorption window which is characteristic for organic materials. For ease of presentation, we assume -for now- that both subcells of the tandem structure have the same absorption window in nm.

Figure 6 shows the maximum efficiency η_{max} and the optimum bandgaps E_{g1} and E_{g2} in the ideal scenario for a stacked and monolithic configuration as a function of the absorption window width. The broader the absorption window, the higher the efficiency. Only at an absorption window of 700 nm, the efficiency decreases because - by imposing the absorption window - we also impose limits on the allowed bandgaps. Notice that there is only a negligible difference between the stacked and the monolithic configuration (except at an absorption window of 100 nm). As explained above, the efficiency for non-optimal bandgap configurations of the monolithic tandem cell will be much lower than for the stacked cell. For example, if we look at an organic cell with bandgaps E_{g1} = 2.5 eV and E_{g2} = 1.5 eV and an absorption window of 100, 200 and 300 nm, the stacked cell still has a maximum efficiency of 20.0, 32.0 and 41.0 %, respectively, whereas the monolithic cell reaches only 17.9 % for a 100 nm broad absorption window and 22.4 % for an absorption window of 200 nm or more wide. Thus in the case of non-optimal bandgaps, we can conclude that for increasingly smaller absorption windows, the advantage of the stacked solar cell over the monolithic cell decreases.

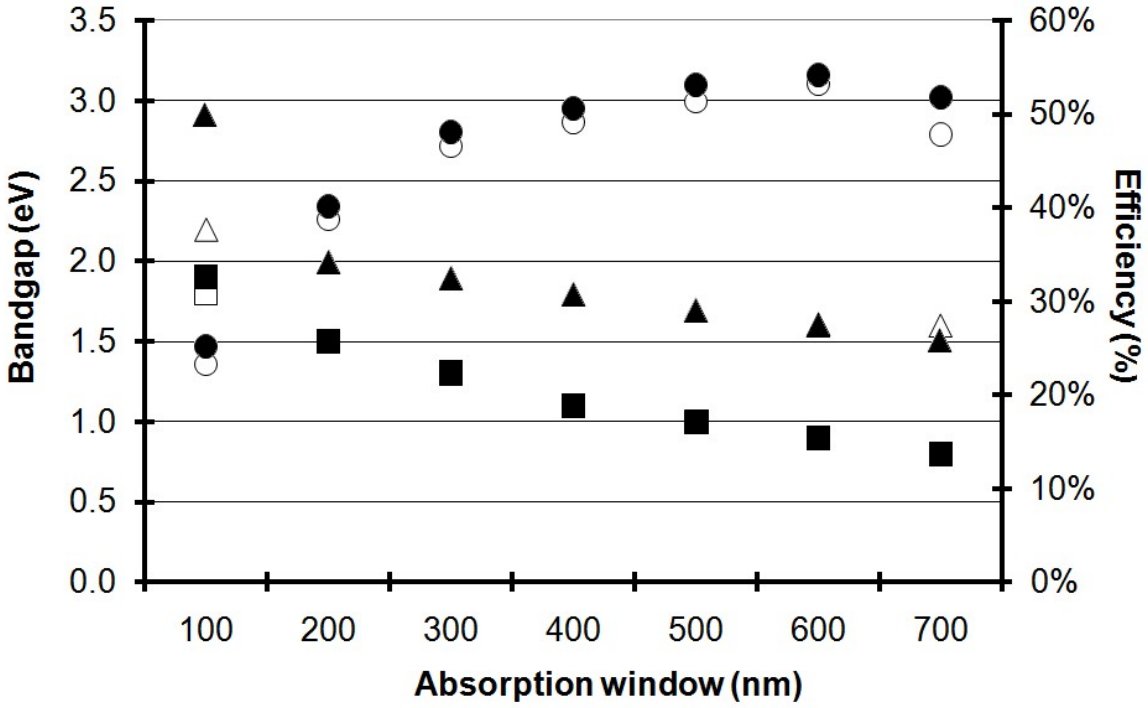

Figure 6: The maximum efficiency η_{max} is plotted in the ideal scenario (with ΔLUMO = 0.2 eV) for an organic stacked (●) and monolithic (○) tandem cell as a function of the absorption window. Also the optimum bandgaps E_{g1} (▲) and E_{g2} (■) for a stacked cell are plotted as a function of the absorption window. The optimum bandgaps E_{g1} (△) and E_{g2} (□) for the monolithic cell are plotted if they differ from the stacked cell.

Figure 6 shows that the optimum bandgap of the cells shifts towards higher energies for lower absorption windows. For example, the optimum bandgap shifts from E_{g1} = 1.6 eV and E_{g2} = 0.9 eV for a full absorption band monolithic cell to E_{g1} = 2.0 eV and E_{g2} = 1.5 eV for a cell with an absorption window of only 200 nm. This is a satisfying result, because, as we already mentioned, the production of suitable low bandgap organic materials is difficult. For an absorption window of 400 and 500 nm respectively, already 96 % of the maximum attainable efficiency (for a full absorption band) is reached for the monolithic configuration and even more for the stacked configuration. Hence, it would not pay off to try to develop organic materials with an absorption window broader than 400 nm, because hardly any efficiency gain can be achieved by widening the absorption window further. The optimum bandgaps with a sufficient absorption window of 400 nm are E_{g1} = 1.8 eV and E_{g2} = 1.1 eV for both configurations.

3.3. Subcells with an unequal absorption window.

We now look at the situation where both subcells of the tandem structure have a different absorption window. In Figure 7 we plot the maximum efficiency for different absorption windows of the subcells. Each data point was calculated using the optimal bandgap configuration for this particular absorption window combination.

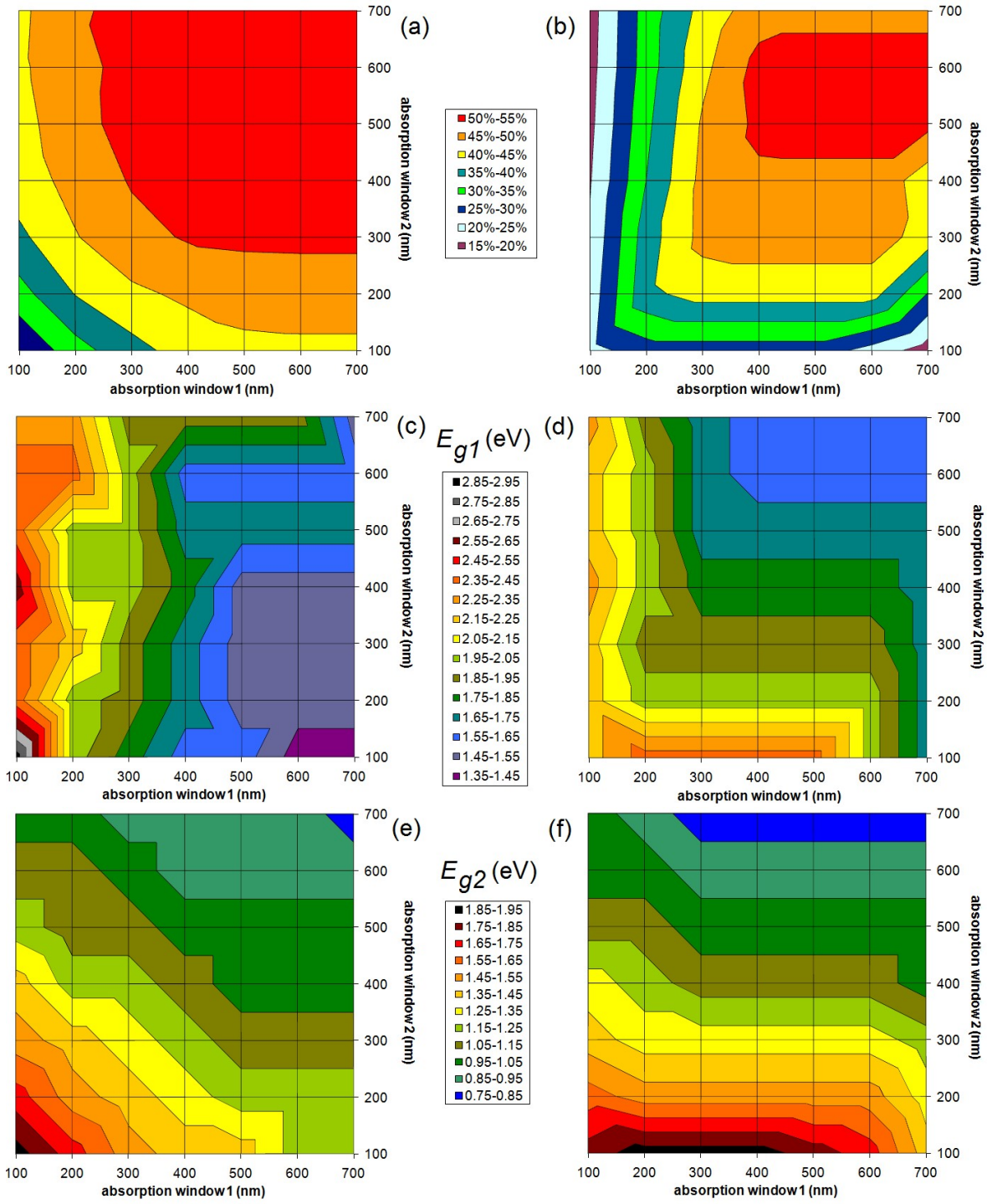

Figure 7: The maximum efficiency η_{max} is plotted in the ideal scenario for an organic stacked (a) and monolithic (b) tandem cell as a function of the absorption windows of the subcells. "Absorption window 1" refers to the first subcell with the highest absorber bandgap, i.e. the top cell directed at the sun. The optimal bandgap E_{g1} (c, d) and E_{g2} (e, f) in eV of the first subcell of a stacked (c, e) and a monolithic (d, f) organic solar cell as a function of the absorption windows of the subcells.

One notices that a small absorption window for just one of the subcells does not lead to a significant decrease in efficiency of the stacked cell, as long as the absorption window of the other subcell is wide enough (Figure 7a). If one subcell has an absorption window of only 100 nm or 200 nm, the maximum efficiency is still about 80 % or 90 % respectively of the absolute maximum obtained in the case of full absorption windows, as long as the other subcell has an absorption window of at least 400 nm.

This does not apply for the monolithic cell (Figure 7b). As soon as one subcell has a low absorption window, the efficiency decreases rapidly. A monolithic cell with an absorption window of 100 nm or 200 nm for the first subcell, and 400 nm for the second subcell, only has a maximum efficiency of less than half and three quarters, respectively, of the absolute maximum for full absorption windows. Again, the plots show for both configurations that absorption windows of more than 400 nm are not necessary for achieving good power conversion efficiency. Figures 7a and b are not symmetrical: for example, a monolithic tandem cell where the first subcell has an absorption window of 100 nm has a maximum attainable efficiency of 23.3 %, whereas if it is the second subcell that has an absorption window of 100 nm, the maximum attainable efficiency is 27.6 %. The asymmetry is only minor, but is more present for a monolithic cell.

Figures 7c to 7f show the optimal bandgap configurations for a stacked and monolithic organic tandem cell with unequal absorption windows. In general the optimum bandgap of the cells shifts towards higher energies for lower absorption windows. In an organic stacked tandem cell, the optimal bandgap E_{g1} of the first subcell only reaches values higher than 2.0 eV when the absorption window of the first subcell is 200 nm wide or less (Figure 7c). Narrowing the absorption window of the first cell increases the optimal bandgap E_{g1}. The value of the bandgap E_{g1} depends mainly on this first absorption window. It is not very dependent on the absorption window of the second subcell. In contrast with the stacked cell, the optimal bandgap E_{g1} of the first subcell for a monolithic configuration depends on both absorption windows (Figure 7d). The lower the absorption windows of the subcells, the higher the optimal bandgap E_{g1}.

The optimum of the bandgap E_{g2} of the second subcell is always such that the absorption window of the second subcell borders (or almost borders) the cut-off wavelength of the first subcell (not visible on the figures). This is the case for the stacked as well as the monolithic configuration. Only when the absorption window width of the second subcell is 100 nm or less, there is some significant space between both absorption windows, although never more than 50 nm. Hence, in all optimal bandgap configurations, (nearly) the entire solar spectrum between the outside borders of the absorption windows is absorbed.

The optimal bandgap E_{g2} of the stacked cell (Figure 7e) shifts (quite symmetrically) towards higher energies for lower absorption windows of both the first and the second subcell. The explanation is that the optimal bandgap E_{g2} is located in such a way that it (almost) borders the absorption window of the first subcell, as mentioned above. In the monolithic configuration, the bandgap E_{g2} is more or less independent of the absorption window of the first subcell. For narrow absorption windows of the second subcell, the monolithic configuration requires higher optimal bandgaps than the stacked configuration. This is a satisfying result, taking into account the characteristic narrow absorption window of organics and the difficulty of producing suitable low bandgap organic materials. Unfortunately, this has a negative influence on the efficiency.

3.4. The maximum obtainable efficiency in a more realistical situation.

To illustrate the influence of a non-ideal *EQE*, we consider a stacked solar cell with absorption windows of 400 nm wide for both subcells. Figure 8 plots the optimal bandgaps E_{g1} and E_{g2} as a function of different *EQE* for the subcells. One notices that the optimal bandgap increases for lower *EQE*s. The bandgap E_{g1} of the top subcell is influenced more than the bandgap E_{g2} of the bottom subcell. Qualitatively similar results are obtained for a monolithic configuration and for different absorption windows.

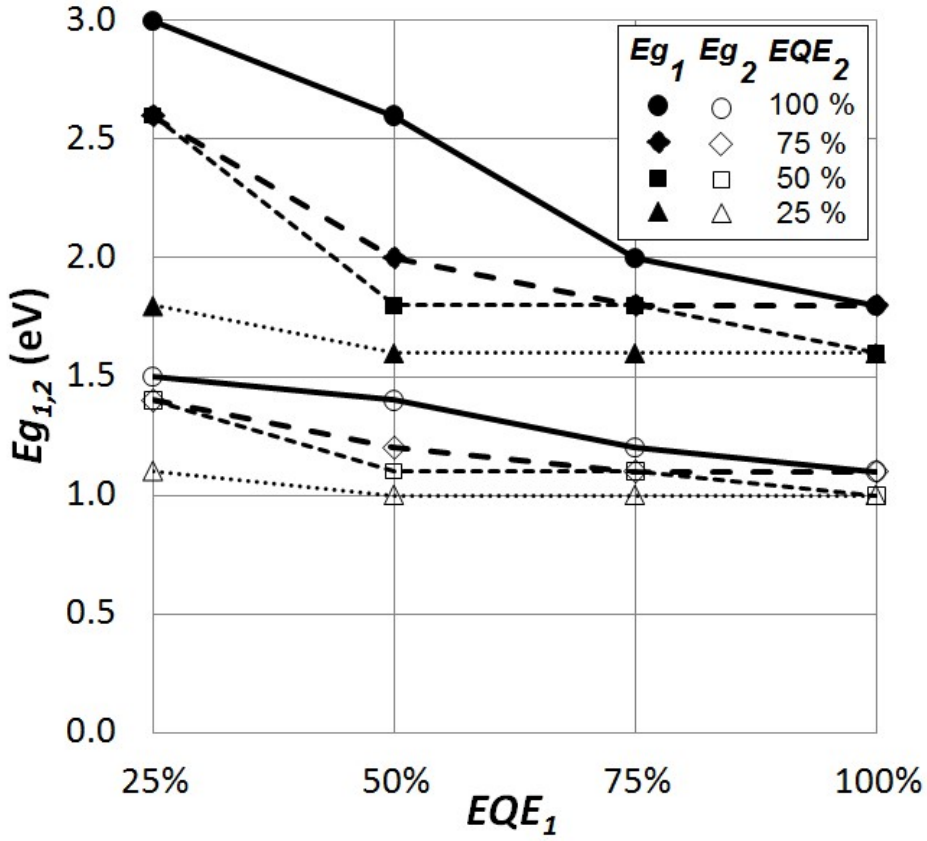

Figure 8: The optimal bandgaps E_{g1} and E_{g2} as a function of different EQEs for the subcells for a stacked solar cell with absorption window of 400 nm wide for both subcells.

To estimate the maximum obtainable efficiency in a less ideal situation, we assume a more realistic scenario. We consider an absorption window of 400 nm wide for both subcells, an *EQE* of 65 %, a fill factor *FF* of 60 %, and a voltage factor *f* of 60 %, with *f* defined by:

$$f = \frac{q \cdot V_{oc}}{E_{g,i}} \tag{3}$$

We consider that both single cells of the tandem structure have the same *EQE*, *FF* and *f*. This results in a maximum attainable efficiency of 11.9 % and 11.5 %, respectively, for stacked and monolithic organic solar cells, both at an optimal configuration (E_{g1}, E_{g2}) of (1.8 eV, 1.1 eV).

4. CONCLUSIONS

A maximum attainable efficiency of 54.0 % and 53.3 %, respectively, is reached in our ideal model for a stacked and monolithic tandem cell at a bandgap configuration of (1.7 eV, 0.9 eV) and (1.6 eV, 0.9 eV). This is about a third more when compared with a single junction organic cell. The requirements for a close to optimal configuration of the stacked tandem cell are quite broad. This is not the case for the monolithic configuration; the value of the bandgap E_{g1} of the first subcell especially is more critical than for a stacked cell. We found that a monolithic tandem cell is much less efficient than a stacked cell for a non-optimal bandgap configuration. We also studied the influence of the narrow absorption window on the efficiency. The optimum bandgap of the cells shifts towards higher energies for lower absorption windows. An interesting result is that it would not pay off to try to develop organic materials with an absorption window broader than 400 nm, because hardly any efficiency gains can be achieved by further broadening the absorption window. The optimal bandgaps with a sufficient absorption window of 400 nm are E_{g1} = 1.8 eV and E_{g2} = 1.1 eV for both configurations. Furthermore, for a stacked organic tandem cell, it is not necessary that both subcells have a large absorption window. This does not apply for the monolithic cell. As soon as one subcell has a low absorption window, the efficiency decreases rapidly.

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