ORGANIC SOLAR CELL STRATEGY AND DEVELOPMENTS IN FLANDERS

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ABSTRACT: IMEC started up its activity on organic solar cells in 1998 and since 2005 this topic became one of the key technologies of the Flemish Innovation Policy in the field of Photovoltaics. The aim of the paper is to present the overall Flemish approach and strategy to this field as well as to highlight the recent achievements of the Organic Photovoltaics Technology Program. The strategy of this program is built around three focal points: enhancement of the conversion efficiency, improvement of the cell stability and development of a printing technology to realize monolithic modules on a flexible foil. This strategy is being executed mainly within IMEC with its large expertise in the field of solar cell technology development and its associated lab, IMOMEC, and the University of Hasselt which has a strong background in synthesis and characterization of conjugated polymers. In addition, there is considerable analytical and modeling support by the University of Antwerp and the University Ghent.

Keywords: polymer film – organic solar cell – tandem

1 STRATEGIC BACKGROUND

Solar cells, using organic materials in the active part of the device, have been the object of research since many years in view of the potential cost reduction achievable with these devices relative to solar cells based on inorganic semiconductors (Si, CdTe, $CuInSe_2$ and related compounds). This low cost potential is often ascribed to the minute amount of the required organic compounds for the thin films and the low cost of the substrates on which the devices are built. The large cost reduction potential is actually rather linked to the quantum leap achievable in terms of manufacturing throughput by the prospect of a reel-to-reel production technology with such materials and substrates. In principal, all these factors could lead to a direct solar module cost of 0.5 Euro/W_p or below. Despite this promise of lower costs, progress was hindered by the low conversion efficiencies reported until the beginning of the nineties [1].

However, the interest in the domain of organic solar cells has been growing fast since the discovery of the Graetzel cell [2] in the beginning of the nineties and the appearance of polymer-based bulk donor-acceptor heterojunction solar cells half of the nineties[3, 4]. Both types of cells deviate considerably from classical solar cell device structures in that they both depend strongly on the presence of nanoscale phases and volume-distributed carrier-collecting junctions. Both features are crucial to obtain efficient exciton dissociation and charge carrier generation. This opened the door for impressive progress on the level of efficiencies since then with efficiencies of 11% for the Graetzel cell and above 5% for the bulk donor-acceptor approach but a number of important challenges remained.

In order to become an economically viable photovoltaic technology the conversion efficiency is to be improved further for the solid-state version of the Graetzel cell and polymer solar cells, stability has to be shown and improved and, last but not least, a manufacturing technology is to be developed which realizes the potential of very high manufacturing throughputs.

IMEC has a long tradition in the field of crystalline Si solar cells, in which it definitely took advantage of the large semiconductor expertise available within the institute. A similar synergy between solar cells and electronics was perceived in the second half of the nineties when IMEC decided to start activities in the field of polymer and molecular electronics. Moreover, the presence of a polymer synthesis and analysis laboratory at the University of Hasselt with a strong background in conjugated polymers was an invaluable asset in combination with the device processing and device physics background of IMEC.

Besides the context of future large-scale energy supply, organic solar cells can also play a role as an enabling technology within the so-called “ambient intelligence” vision. The term ‘ambient intelligence” refers to a vision on future electronic systems in which ubiquitous computing power will be distributed over our near environment. Many of these electronic systems will contain a sensing part associated with data processing capability as well as RF-features for data communication. Within this vision, ensuring the energy autonomy of freestanding and (or) portable circuits, is a crucial task. It turns out that, even at low illumination levels of typically 1% of standard sunlight or below, photovoltaic cells are the most obvious means to ensure this required energy autonomy with lowest area or volume requirements, and organic (flexible) solar cells are an obvious enabling technology to do so.

The potentially low cost of the technology, the synergy with activities on polymer electronics and the complementarity of several Flemish players resulted in the start of an activity on organic solar cells in 1998 with a focus on polymer-based solar cells, addressing the challenges described in the previous paragraph. The Organic Photovoltaics Technology Program is since 2002 an integral part of the overall Strategic Photovoltaic Program $SOLAR^+$ at IMEC and since 2005 it became one of the key technologies of the Flemish Innovation Policy in the field of Photovoltaics. It is supported by a Strategic Basic Research Project with the acronym NANOSOLAR.

2 DESCRIPTION OF FLEMISH PARTNERSHIP

2.1 IMEC/IMOMEC/University Hasselt (UH)

The improvement of solar cell conversion efficiency and the development of the final cell and module manufacturing technology is mainly done at IMEC. Within IMEC there is a large equipment base to perform these tasks. It comprises all necessary tools for the growth (by means of vacuum evaporation or vapor phase deposition) or casting (spinning or screenprinting) of the active organic layers as well as the necessary equipment to process and analyze the final devices in a N_2 glove box environment.

The research group "Organic material Synthesis" at IMEC (located at the premises of the University of Hasselt and part of the IMOMEC-division) performs research in the field of the synthesis and characterization of functional polymers with new electrical conductive and/or special optical properties. Besides the study of novel synthetic methods for special monomers, new polymerization reactions, mechanisms of polymerization reactions, also modifications of existing polymerization pathways are studied. In this area, attention is also paid to the theoretical basis related to these material systems, in order to get an insight into the underlying structure-property relations with rational material design as a final objective.

2.2 University Ghent (UG)

The University Ghent has a strong background in the field of electrical modeling of polycrystalline compound solar cells. The modeling software package SCARF [5] is heavily used by many groups active in the field. Their recent research activity is in the field of solar cells with volume-distributed junctions like the eta-cell [6] and the bulk donor-acceptor heterojunction cell concept [7].

2.3 University Antwerp (UA)

The University Antwerp has a strong background in advanced analysis of organic materials [8,9] with special emphasis on the measurement and analysis of spectroscopic properties. Besides the basic optical absorption and fluorescence spectroscopy, the Antwerp partner disposes of picosecond time-resolved laser spectroscopy and high-sensitivity resonant Raman scattering. In addition, electron paramagnetic (or spin) resonance techniques (EPR or ESR) are available ranging from conventional X-band (9.4 GHz) to high-frequency W-band (94 GHz) and pulsed EPR. This also includes combinations of magnetic resonance and optical techniques, i.e., light-induced (LI) EPR, optically detected magnetic resonance (ODMR) and transient EPR after laser pulse excitation.

3 SYNTHESIS OF NOVEL ACTIVE MATERIALS

Firstly, there is the development of reliable synthetic methods for organic low-bandgap semiconductors to improve the matching of the absorption spectrum of the material system with the emission spectrum of the sun. Two approaches are being followed: derivatives of poly-thiophene (PT) or derivatives of poly-thienylene-vinylene (PTV), results of which will be shown in paragraph 3. In recent years we have put major efforts to develop a reliable synthetic route toward PTV's with the development of the "dithiocarbamate precursor route".

Secondly, efforts are oriented towards material systems that can be processed from more environmentally friendly solvents. This implies that one should develop electron donor and acceptor materials which are more polar. One beneficial side effect could be that because of the more polar environment exciton dissociation may go faster. The on-going work focuses on PPV derivatives with polar side chains soluble in various solvents – from apolar to polar and to water. The results for MDMO-PPV and the three polar PPV derivatives are depicted in Table 1. The reference material MDMO-PPV (OC_1C_{10}-PPV) gives a relative permittivity of ~3. Replacing the $OC_{10}H_{23}$ side chain (OC_1C_{10}-PPV) by a $(CH_2CH_2O)_3CH_3$ side chain (PEO-PPV) increases the permittivity to roughly 4. Substituting the OCH_3-side chain by an OC_9H_{19} side chain ((PEO-OC_9-PPV) further increases the permittivity slightly from 4.0 to 4.1. Finally, introducing a second $(CH_2CH_2O)_3CH_3$ side chain (diPEO-PPV), results in a permittivity as high as 5.5.

Table 1: Summary of the electrical properties of the newly synthesized polymers with increased polarity

Polymer	ε_r	μ [10^{-4} cm^2/Vs]
OC_1C_{10}-PPV	*3.0*	*2 - 3*
PEO-PPV	*4.0*	*1 - 5*
diPEO-PPV	*5.5*	*1 - 4*
(PEO-OC_9)-PPV	*4.1*	*3 - 6*

At this moment the morphology consists of a frozen structure resulting from demixing of PCBM in a polymeric matrix. This morphology will evolve in time to stronger demixing depending on the T_g of the matrix which means that the morphology is hard to stabilize. The positive effect of materials with higher glass transition temperatures (T_g) on the morphology stability has been recently demonstrated [10]. It is very important to develop methods which can stabilize the once optimal morphology in a definitive way. A successful solution has been demonstrated using a so-called high T_g-PPV, the structure of which is shown in Figure 1.

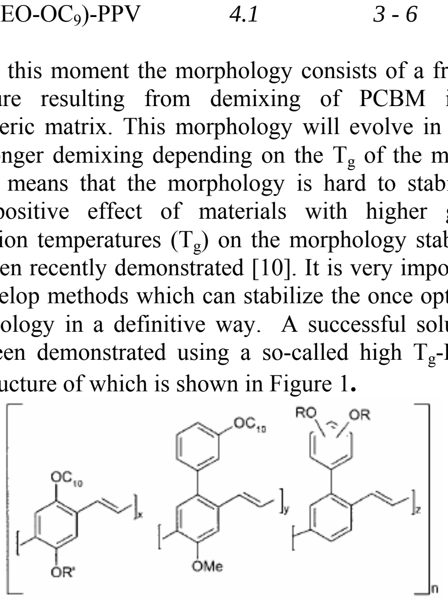


Figure 1: Structure of high-T_g PPV, synthesized at IMEC/IMOMEC

4 CELLS AND MODULES

4.1 Improving the efficiency of basic bulk donor-acceptor heterojunction devices

Initially, a lot of effort has gone into the improvement and understanding of bulk donor-acceptor heterojunction cells based on blends of PPV (poly-paraphenylene vinylene) with PCBM. This resulted in efficiencies near 3.5%[11]. Gradually, the focus has moved to P3HT (poly-3-hexylthiophene) as donor material because it has a higher absorption coefficient close to the maximum photon flux in the solar spectrum. P3HT is also known in the field of organic electronics as a high hole mobility material. Solar cells based on the P3HT:PCBM system normally require a short thermal anneal to increase their performance [12]. A thorough analysis has revealed that addition of the second compound, PCBM (a bucky ball derivative) to the active layer, serving as acceptor and transporter of excited electrons, disturbs the natural tendency of P3HT to form a crystalline structure. During this annealing step the P3HT is able to recrystallize [13], which was proven by several techniques like XRD, TEM and SAED (Selected Area Electron Diffraction), as shown in Figure 2 . This has a positive influence on the solar cell's absorbance and the mobility of the charges inside the P3HT, as deduced from transistor and space charge limited current measurements. Spectroscopic data (see Sec. 5.2) have on the contrary demonstrated that exciton dissociation becomes even slightly less effective.

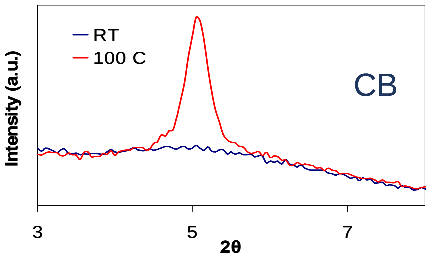


Figure 2: XRD-spectrum before and after anneal of a P3HT:PCBM-film (the film was casted from chlorobenzene)

The crucial parameter for the thermal anneal turned out to be the evaporation speed of the solvent. In more slowly evaporating solventsthe P3HT polymer segments remain mobile for a longer time during the film formation, allowing for the creation of crystalline P3HT-network. This was experimentally evidenced by testing solvents with a higher boiling temperature. Figure 3 shows the XRD-spectrum of a film casted from tetraline.

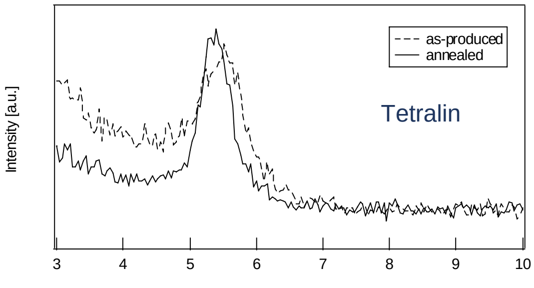


Figure 3: XRD-profiles of P3HT:PCBM films when casted from tetraline (annealed vs. non-annealed)

The XRD-spectrum clearly indicates that the P3HT donor material is already present in its crystalline form in the film without annealing . Careful optimization of the solvent and the evaporation conditions of the solvent allowed to realize P3HT:PCBM solar cells with efficiencies of 4.5%., which is comparable to the best conversion efficiencies reported for polymer-based solar cells [14].

4.2 Improving the efficiency: organic multijunction devices

One drawback of organic photoactive materials is their narrow absorption window compared to solar cells based on inorganic semiconductors. A possible way to extend the spectral sensitivity over a broader wavelength region is stacking different solar cells on top of each other.

In practice, the stacking can be done by placing the two cells in parallel or in series. Several groups throughout the world are working towards tandem organic cells [15,16,17]. The challenge of making tandem cells is in finding an efficient recombination center in between the two subcells to ensure their efficient series connection. Different metallic nanomaterials were studied as recombination centers to interconnect subcells based on pentacene and fullerene (C_{60}). Pentacene combined with C_{60} as active layer for organic solar cells has recently gained interest [18,19]. Both materials show a high transistor mobility [20] and pentacene has a peak absorption around 670 nm which is close to the maximum of the AM1.5 solar spectrum. We tested vacuum deposited metal layers as recombination centers with pentacene and buckminsterfullerene (C_{60}) as donor and acceptor respectively. As shown in Figure 4, S-shaped curves were visible in the I-V characteristics when using thin layers of aluminum, indicating a barrier for extraction inside the device.

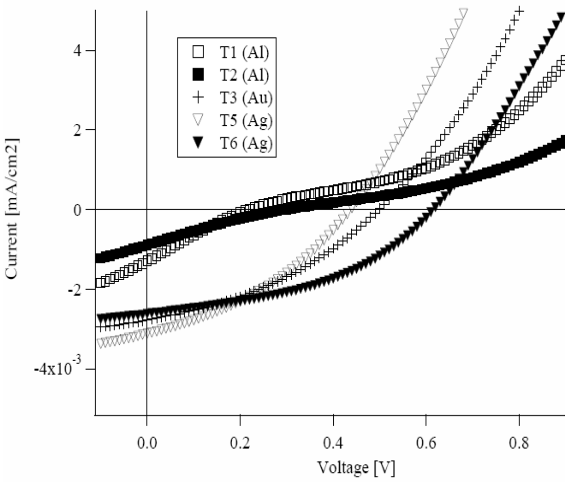


Figure 4: IV curves of tandem cells. The structures are tandem planar heterojunction cells with (T1, T3, T5) and without (T2, T6) PEDOT:PSS layer on top of the ITO. Further layer structure: 40nm pentacene/ 30nm C_{60}/ 0.5nm metal layer/ 40nm pentacene/ 30nm C_{60}/ 10nm BCP/ 70nm Al. The metal layer is aluminum (T1 and T2), gold (T3 and T4) or silver (T5 and T6)

Thin metal layers of gold or silver result in a doubling of the open-circuit voltage, as shown in Table 2, without the appearance of these S-shaped features [21].

Table 2: Overview of solar cell performance parameters for single and tandem cells. S1 is a single planar heterojunction cell whereas for the other the nano-metal interconnection is indicated.

Cell	Jsc [mA.cm-^{2}]	Voc [mV]	FF [%]	η [%]
S1	6.7	0.29	51	0.55
Al	1.29	0.22	23	0.03
Ag	2.34	0.52	37	0.3
Au	2.6	0.62	46	0.47

4.3 Towards a high-throughput manufacturing technology: printed active layers

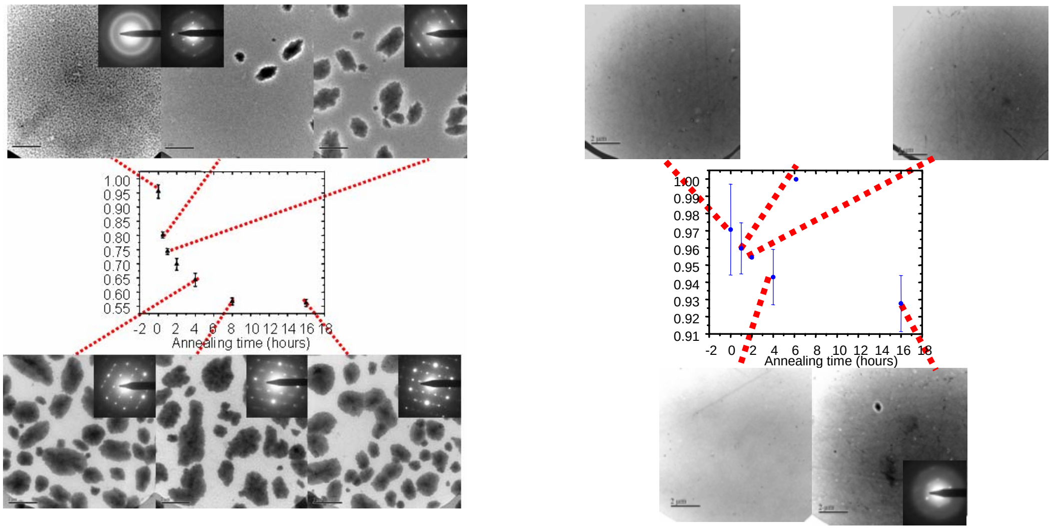
Besides single cell optimization and efficiency improvement, the development of a monolithic organic solar cell module is also under investigation. Screen printing was investigated for the deposition of the active layer of a bulk donor-acceptor heterojunction solar cell using MEH-PPV as donor and PCBM as acceptor material. This technique has the advantage that the active layer can be patterned and that material losses during deposition are lower as compared to spin coating. Solar cells and modules were realized with energy conversion efficiencies above 1% and fully operational modules were applied as driver for a pocket calculator as shown in Figure 5. Monolithic modules have been obtained with active areas of approx. 10cm^2 with patterned frontside contacts and a printed organic active layer. The versatility of this production process offers the possibility to use glass as well as flexible foils for the substrate [22].

Figure 5: Organic solar cell module with printed active layers driving a small pocket calculator

5 ADVANCED CHARACTERIZATION

5.1 Nanomorphological changes and stability

A study of the thermal stability of the morphology has been performed for various material combinations including MDMO-PPV:PCBM, P3HT:PCBM and high T_g-PPV:PCBM. The combined study on the thermal stability of the morphology of MDMO-PPV:PCBM and P3HT:PCBM results in the important conclusion that the morphology of the investigated blended material systems are not thermally stable, but their morphology changes continuously as a function of thermal annealing. The blended material systems are not in thermodynamic equilibrium but are subject to temperature driven effects such as diffusion of PCBM yielding continuous growth of PCBM-crystals [23].



a) b)

Figure 6 a) TEM-images of bulk morphology and corresponding degradation curve of the short-circuit current for MDMO-PPV:PCBM blends annealed at 110°C; b) images of bulk morphology and corresponding degradation curve of the short-circuit current for high Tg-PPV:PCBM blends annealed at 110°C.

For the newly developed high-T_g material (see paragraph 2 of this paper) the glass transition temperature T_g is in the range of 140°C-150°C, while for MDMO-PPV the T_g is only around 45°C. Since the glass transition temperature T_g of the new material is higher than the typical operation and annealing conditions (room temperature to 125°C) the material structure remains quite rigid at these temperatures and therefore the thermal stability of the morphology is drastically increased as illustrated in Figure 6.

5.2 Exciton dissociation and polaron properties

In the photovoltaic cell, photocreated excitons are dissociating at the polymer/fullerene interface and the electrons are transferred from the donor to the acceptor component of the composites. The evaluation of this very important step is routinely performed using several techniques. The quenching of the polymer fluorescence in the blends is determined from steady state measurements and the corresponding decrease in lifetime is verified by picosecond streak-camera experiments. In parallel, the concentrations of the radicals created under illumination (i.e.: the positive polaron of the polymer and the fullerene anion) can be quantitatively determined by light-induced EPR measurements, with sufficient resolution in W-band to resolve both species. This is applied both as a test of the charge transfer process of blends with new polymers [24], and in comparative studies of the charge transfer efficiencies in different polymer/PCBM blends. Such experiments have also demonstrated [25] that in morphology-optimized P3HT blends the charge separation efficiency is lowered while higher solar cell performance is obtained. The latter is ascribed to higher polymer chain ordering leading to an increased hole mobility. A similar approach is followed for the evaluation of novel n-type polymers which can be applied as donors in polymer/polymer blends.

6 EFFICIENCY POTENTIAL

Given the progress seen in the last decade, the question about the limits for the achievable efficiencies arises. In order to shed more light on this, a calculation was made taking into account a number of assumptions. Firstly, we consider a solar cell with one active material with a bandgap E_g. We make the following fundamental assumptions: (i) every photon with an energy $h\nu$ higher than the bandgap E_g is absorbed. (ii) photons with an energy $h\nu$ lower than the bandgap E_g are fully transmitted. (iii) every absorbed photon leads to a useful energy E_g. In this case, the maximum efficiency η_{max} is given by:

$$\eta_{\max} = \frac{E_g \int_{E_g}^{\infty} N(E)\ dE}{\int_0^{\infty} E\ \ N(E)\ dE}$$

with $N(E)$ the photon flux. For all our simulations, we use the AM 1.5 experimentally measured solar spectrum. A maximum efficiency of 48.7 % is reached when E_g=1.14 eV, which is a well-known result. However, producing suitable organic absorbers with such a low bandgap is a problem. When we take also into account a narrow absorption window, characteristic for organic materials, the optimal bandgap shifts towards higher energies. For an absorption window of e.g. 300 nm, the optimal bandgap is 1.7 – 1.8 eV, which is in line with current organic absorbers. To estimate the maximum obtainable efficiency with the current state of technology, we assume the following realistic values, which are nowadays reached in organic photovoltaics. We assume an absorption window of 400 nm, a quantum efficiency QE of 70%, a fill factor FF of 65%, and a voltage factor f:

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

of 60%, with V_{oc} the open circuit voltage. This results in a maximum attainable efficiency of 11 %. If we consider optimistic values, which however are credible to be achieved in the future, an efficiency of 22 % becomes possible (i.e. an absorption window of 500nm, QE=90%, FF=80%, f=70%).

The active material in a bulk heterojunction solar cell consists not of one material, but 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. For our simulation, the following assumptions are made (Figure 3 for definition of terms): (i) only one material absorbs light (usually, this is the p-type component) (ii) every photon with an energy $h\nu$ higher than the bandgap $E_{g,absorber}$ is absorbed, with the bandgap defined as the difference between the LUMO and the HOMO of the absorber. (iii) not any photon with an energy $h\nu$ lower than the bandgap $E_{g,\ absorber}$ is absorbed. The distance between the HOMO of the donor, and the LUMO of the acceptor is considered as the thermodynamic limitation for the useful energy. This value is often called the interface bandgap $E_{g,i}$. We assume that (iv) every absorbed photon leads to a useful energy $E_{g,i}$. In this case, the maximum efficiency η_{max} is given by:

$$\eta_{\max} = \frac{E_{g,i} \int_{E_g}^{\infty} N(E)\ \ dE}{\int_0^{\infty} E\ \ N(E)\ \ dE}$$

The HOMO of the n-type component is taken as a reference ($HOMO_n$ = 0 eV), and because of energetic reasons, $HOMO_n < HOMO_p < LUMO_n < LUMO_p$ (see Figure 7).

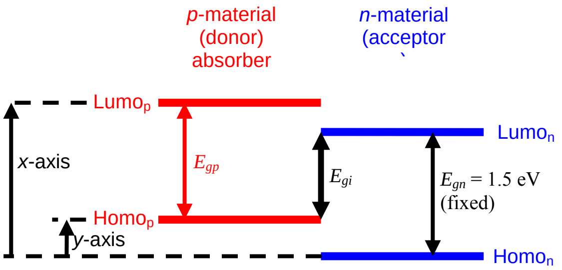


Figure 7: Definition of terms: donor and acceptor material; absorber; HOMO and LUMO levels; parameters varied and fixed in Figure 9.

One notices from Figure 8 that, with a full absorption window, the optimal bandgap of the absorber is again 1.1 eV. The highest efficiency is reached when the LUMO of the p-material is as close as possible to the LUMO of the n-material. This was expected, because the difference between the LUMO's corresponds with an energy loss of the absorbed photon. However, a necessary condition for efficient dissociation of the created excitons is that the difference between the LUMO's of the donor and acceptor is higher than the exciton binding energy. The value of the exciton binding energy in different materials is a subject of discussion, values between 0.1 eV and 2 eV are published [26]. For the simulations, we assume a difference of 0.2 eV between the LUMO's. This value was put forward as an empirical threshold necessary for exciton dissociation [27]. If we also take into account the realistic present and future values of the absorption window, *QE*, *FF* and *f* mentioned above, the maximum obtainable efficiency of organic bulk heterojunction solar cells is respectively 9.4 % and 19 %.

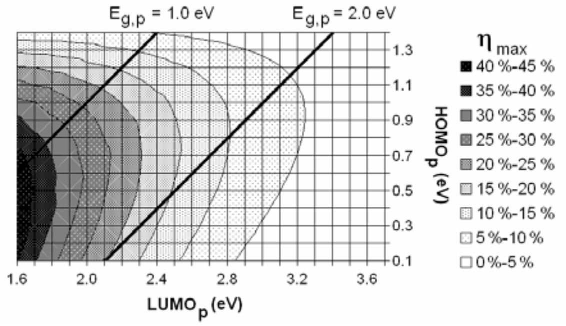


Figure 8; The maximum efficiency plotted as a function of the LUMO and the HOMO of the *p*- material for the case where the p-type material absorbs the light, and the n-type has a bandgap of 1.5 eV (e.g. PCBM). The HOMO of the *n*-material is taken as a reference ($HOMO_n$ = 0 eV). Two straight lines indicate where the bandgap of the p-material is 1.0 eV and 2.0 eV.

7 CONCLUSIONS

Within IMEC and a number of Flemish universities organic solar cell research has been significantly gaining momentum. A close partnership was created involving several Flemish R&D-institutes and universities. This resulted in impressive progress on the level of device performance, improvement of stability as well as in the fundamental understanding of the device performance.

8. ACKNOWLEDGEMENTS

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