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Three-Layered Photovoltaic Cell with an Enlarged Photoactive Region of Codeposited Dyes

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Abstract. — Three-layered organic solar cells with an interlayer of codeposited dyes of p-type Copper Phthalocyanine (CuPc) and n-type 5,10,15,20-Tetra (4-Pyridil)21H,23H-Porphine (TPyP) between the respective dye layers were prepared and characterised. The analysis of their dark current-voltage \( (I - U) \) characteristics at room temperature has been presented in order to elucidate the conduction mechanisms and to evaluate the cell parameters. The analysis of photovoltaic properties shows that the photocurrent of three-layered cells is about ten times larger than that of a double layered cell, due to the efficient carrier photogeneration in the codeposited layer. The best power conversion efficiency, of 0.35%, was obtained under illumination with monochromatic light of 30 \( \mu \)W cm\(^{-2}\), at 520 nm.

Résumé. — Nous avons élaboré des cellules photovoltaïques avec trois couches minces de colorants organiques entre deux électrodes : ITO et Al. Les cellules ont été faites en utilisant CuPc comme semi-conducteur de type-p et TPyP comme semi-conducteur de type-n. Entre les deux couches de CuPc et TPyP, une couche mince de CuPc+TPyP a été déposée par co-évaporation thermique des deux matériaux organiques. L’analyse des caractéristiques électriques \( (I - U) \) des cellules dans l’obscurité est présentée pour comprendre les mécanismes mis en jeu et pour évaluer leur paramètres électriques. L’analyse des propriétés photovoltaïques a montré que le photocourant des cellules avec trois couches minces est 10 fois plus grand que celui mesuré sur les cellules à deux couches minces, grâce à une création plus efficace des photoporteurs dans la couche mince co-évaporée. Le rendement de conversion énergétique est maximal à 0,35 % pour la lumière avec une longueur d’onde de 520 nm et une intensité lumineuse de 30 \( \mu \)W cm\(^{-2}\).

1. Introduction

There has been a great deal of interest in the photovoltaic properties of the organic semiconductors because of their potential use as inexpensive material for solar cells and because of their high optical absorption in visible region of the solar spectrum. Among the organic materials, which have been studied from this point of view, were the merocyanines [1,2], phthalocyanines [3–6] and porphyrins [7–9]. Most of the organic cells have been of Schottky type.

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in which an organic layer is sandwiched between two electrodes having suitable work function, one of them forming a rectifying contact and the other one forming an ohmic contact with the dye layer [1–3, 7–9]. There are some factors which limit the performances of an organic Schottky cell, generally having a low power conversion efficiency, smaller than 1%. A higher power conversion efficiency, of about 0.1–2%, has been achieved from the two-layer organic photovoltaic cells [2, 5, 6, 10, 11]. In these devices, the interface between the two organic materials, rather than electrode/organic contacts, is crucial in determining the photovoltaic response. The absorption characteristics of the two layers, if complementary, enhance substantially the utilisation of wavelengths of solar spectrum by the two-layer cell as compared with single-layer cell.

Recently, an internal power conversion efficiency of 0.7% was obtained [12], from a three-layered organic solar cell with photoactive interlayer of codeposited pigments, which has been found to act as an efficient carrier photogeneration layer. Afterwards, we have reported the strong "cosensitization" effect [5], presented by the two-layer organic photovoltaic cell fabricated from Copper Phthalocyanine (CuPc) as p-type organic semiconductor and 5,10,15,20-tetra-(4-pyridil)21H,23H-porphine (TPyP) as n-type organic semiconductor. Motivated by the encouraging results on heterojunction CuPc/TPyP devices and in an effort to obtain a thicker photoactive region at the interface between the two-organic layers, we have fabricated the three-layered organic cells ITO/CuPc/(CuPc+TPyP)/TPyP/Al with a mixed layer of CuPc and TPyP between them. In this paper we present the electrical and photovoltaic properties of ITO/CuPc/(CuPc+TPyP)/TPyP/Al devices as compared with two-layered ITO/CuPc/TPyP/Al cells. The performance of the three-layered cells has been improved over that of two-layered cells in terms of decreased internal resistances, decreased dark current and voltage, increased fill factor and increased power conversion efficiencies; however, the cell efficiencies are still too low for their commercial applications.

2. Sample Preparation and Experimental Procedures

The CuPc dye was supplied by Eastman Kodak Company and TPyP was supplied by Aldrich-Chemie Company; they were used without further purification. The chemical structures of dyes were illustrated in a previous paper [5]. The investigated cells were prepared by the following procedure. The Indium Tin Oxide (ITO) coated glass provided the transparent conducting substrate (with transparency of 60% for the average solar spectrum region), on which a layer of CuPc, about 80 nm thick, was deposited by conventional vacuum evaporation. The deposition of the mixed interlayer was carried out by the coevaporation of CuPc and TPyP from two separated sources. The evaporation rates were separately controlled. We tried to obtain the volume ratio 1:1 for the mixed layers CuPc/TPyP. The total thickness of coevaporated layers was about 80 nm. A third layer of TPyP having the same thickness, was deposited on top of the codeposited layer, also by vacuum evaporation. Finally, an Al layer was evaporated having an area of 0.25 cm² and defining the active area of the cell. The cell configuration is shown as an insert in Figure 1. Both CuPc and TPyP are thermally very stable, thus allowing their deposition by vacuum evaporation. During sublimation the source temperature was maintained at about 380 °C for CuPc and at 280 °C for TPyP. The ITO transparent electrode has been found to form an ohmic contact with p-type CuPc and the Al electrode a nearly ohmic contact with the n-type porphyrin [5, 10]. Two-layered photovoltaic cells ITO/CuPc/TPyP/Al were also prepared, to compare their performances with those of the three-layered cells. Films of common component in this cells, i.e. CuPc, TPyP or Al, were prepared at the same time in such a way that they had the same thickness and morphology. The current-voltage (I – U) characteristics in the dark were measured by biasing the devices with a voltage stabilised power
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source. The current and voltage were measured with a Philips microammeter and a Keithley 614 electrometer, respectively. The illumination of the cells was provided by a 650 W halogen lamp. A monochromator (spectral range of 300 to 800 nm) was used to obtain the spectral responses. The light power density at each wavelength has been measured with a powermeter to have the normalised spectra of photocurrent. The optical absorption spectra of the organic layers were obtained on a Perkin Elmer UV- VIS spectrophotometer (Lambda 2S). The power output of the cells was measured by introducing external resistance (10 kΩ – 2 TΩ) in the circuit.

3. Experimental Results and Discussion

3.1. Dark Current-Voltage Characteristics. — Figure 1 shows the dark current through a typical ITO/CuPc/(CuPc+TPyP)/TPyP/Al cell as a function of the applied bias, at room temperature. The forward bias corresponds to a positive voltage on ITO with respect to Al electrode. The cell exhibits a strong rectifying effect in the dark, characterised by the rectification ratio (forward bias current/reverse bias current) of nearly 500 at the voltage of 1 V. Since CuPc [13] and TPyP [9] make ohmic contacts with ITO and Al electrodes, respectively, we infer that the rectification shown in bipolar (I – U) characteristics in Figure 1 is due to the energy barrier at the junction between the CuPc and TPyP layers. The presence of an energy barrier at the interface CuPc/TPyP has been already mentioned by us in [5], but its width and height were probably smaller than those of the three-layered cells, which show higher asymmetry in (I – U) characteristics. This behaviour shows that the codeposited layer (CuPc+TPyP) gives rise to a thicker and higher energy barrier between CuPc and TPyP, than that existing at the CuPc/TPyP interface in the two-layered cell. Some structures fabricated with only one layer of codeposited dyes (CuPc + TPyP) showed a symmetrical bipolar (I – U)
characteristics and under illumination behave like a photoconductor (results to be published).

For a rigorous analysis of \((I - U)\) curves with respect to conduction mechanisms and cell parameters, firstly we found out the implication of Shockley mechanism by seeking out the linear region in the \(\ln(I) - U\) plot of the forward current-voltage characteristics of the cell. The semilogarithmic plot for the three-layered cell is shown in Figure 2. As can be seen from this figure, the plot consists of a linear region between 0.3-0.6 V and two curved regions at biases higher than 0.6 V and lower than 0.3 V. It is known that for a Shockley diode having high series resistance and small shunt resistance, the \(\ln(I) - U\) plot contains a linear region only in a small range of moderate voltage, while at higher voltages a downward curved region exist. That is why, we consider that the forward characteristic of the cell up to 1 V can be fitted well by the modified Shockley equation:

\[
I = I_0 \left\{ \exp \left[ \frac{q(U - IR_s)}{nkT} \right] - 1 \right\} + \frac{U - IR_s}{R_{sh}}
\]  

(1)

where: \(I_0\), \(n\), \(R_s\) and \(R_{sh}\) are the reverse saturation current, diode quality factor, series and shunt resistance of the cell, respectively and \(q\) is the electronic charge. The source of the series resistance, \(R_s\), is mostly the combined effect of bulk of the organic layers of the cell (having a large thickness \(\sim 0.4 \mu m\)) and the barrier; for an ideal barrier, \(R_{sh} \rightarrow \infty\).

Now, to improve the linearity of the \(\ln(I) - U\) plot, to find out the exact voltage range where the Shockley mechanism is valid, it is essential that the effect of \(R_s\) and \(R_{sh}\) to be removed. But to do so, one needs to know the values of \(R_s\) and \(R_{sh}\). Here, we give a method to determine these parameters \(R_s\) and \(R_{sh}\), but also \(n\) and \(I_0\), by fitting the experimental data with equation (1).
3.1.1. Evaluation of $R_s$ and $R_{sh}$. — From equation (1), the junction resistance, $R_0$, is:

$$R_0 = \frac{dU}{dI} = R_s + \frac{1}{\{\beta I_0 \exp[\beta(U - IR_s)] + 1/R_{sh}\}}$$  \hspace{1cm} (2)

where $\beta = q/nkT$.

For higher forward bias, where $R_s$ affects the curves, equation (1) can be approximated as $I = I_0 \exp[\beta(U - IR_s)]$, and since $1/R_{sh} < \beta I$, equation (2) can be written as follows:

$$R_0 \approx R_s + 1/\beta I$$  \hspace{1cm} (3)

Thus by plotting $R_0$ vs. $1/\beta I$ for high forward biases, one can obtain $R_s$ and the value of $n$ (although its accurate determination will be made from $\ln(I) - U$ plot).

For low voltages, where $R_{sh}$ acts, the approximation $\beta I_0 \exp[\beta(U - IR_s)] \ll 1/R_{sh}$ is valid, and equation (2) becomes:

$$R_0 \approx R_s + R_{sh}$$  \hspace{1cm} (4)

Usually $R_s \ll R_{sh}$, $R_0$ thus approaches $R_{sh}$ at low biases. The graph of $R_0$, the junction resistance, versus $1/I$ is shown in Figure 3, for one of our three-layered cell. One can divide the plot in two regions: A and B, for high and low forward currents respectively. For region [A] (expanded in the insert), one notices that $R_0$ as a function of $1/I$ shows linearity up to the applied voltage of ~0.7 V (the voltage from $J - U$ plot in Fig. 1, corresponding to the value 0.6 GΩ of $R_0$) and curves thereafter, for high forward biases. This fact may suggest that the modified Shockley equation is valid up to 0.7 V and there may be other conduction mechanism at play for higher values. From the extrapolated linear region A, the determined value of $R_s$ is 5.82 MΩ; one can also obtain the value of $n$, though approximately, from the slope, which is 4.9. The value of $R_{sh}$, obtained from region [B] is ~ 2 GΩ. These values of $R_s$, $n$ and $R_{sh}$, depending upon experimental conditions are comparable to those obtained by other authors for porphyrin [7] and phthalocyanine [14] cells.

3.1.2. Evaluation of $n$ and $I_0$. — To improve the linearity of $\ln(I) - U$ plot, for determination of $n$ and $I_0$, we first remove the effect of $R_s$. This is achieved by making the following change in variable:

$$Y = U - IR_s$$  \hspace{1cm} (5)

With this change, equation (1) becomes:

$$I = I_0[\exp(\beta Y) - 1] + Y/R_{sh}$$  \hspace{1cm} (6)

where $Y$ is the voltage only across the junction. For high forward biases, equation (6) can be written as:

$$I \approx I_0 \exp(\beta Y)$$  \hspace{1cm} (7)

Thus, from the plot of $\ln I$ vs. $Y$, $n$ and $I_0$ can be determined. Further, to increase the precision it is advisable to remove the effect of $R_{sh}$ from the $\ln(I) - U$ plot, as well. This is achieved by plotting $\ln(I - Y/R_{sh})$ vs. $Y$, because $(I - Y/R_{sh})$ can be considered to describe the current flowing in the diode junction only. Such a plot for our cell is shown in Figure 4 [A]. One can see that the removal of $R_{sh}$ and $R_s$ has lead to the increase in the linearity of the curve at lower biases from 0.3 V to 0.1 V and at higher biases from 0.7 to 0.8 V, respectively. Then the whole linear region of $\ln(I) - U$ plot is extended in the range 0.1 - 0.8 V. The values of $n$ and $I_0$ obtained from the slope and the intercept are 2.79 and $6.2 \times 10^{-13}$ A, respectively and are more reliable than those obtained after the removal of $R_s$ only.
For biases $> 0.8$ V, other conduction mechanism seems to be operative. Although we have only few experimental data in this range of voltages (we tried to prevent the breakdown of the cells), we observed that the current shows a power dependence of voltage, i.e. follows the relation $I \sim U^m$ where $m = 7$, as seen from $\log(I) - \log(U)$ plot in Figure 4 [B]. This superquadratic power dependence suggests that the dark current is a Space-Charge-Limited-Current (SCLC) in the presence of exponentially distributed traps. According to Mark and Helfrich [15], the SCLC for a solid with a trap distribution decreasing exponentially with the trap depth is given by:

$$J_{\text{SCLC}} = N_{\text{eff}} \mu q^{1-\gamma} \left[ \frac{\varepsilon \gamma}{N_t(\gamma + 1)} \right]^{\gamma} \left[ \frac{(2\gamma + 1)}{(\gamma + 1)} \right]^{\gamma+1} \left( \frac{U^{\gamma+1}}{d^{2\gamma+1}} \right)$$

(8)

where $N_{\text{eff}}$ is the effective density of states in conduction or valence band, $\mu$ is the mobility of majority carriers, $\varepsilon$ is the permittivity of material, $d$ is the thickness of the layer, $U$ is the applied voltage and $\gamma = T_c/T$, $T_c$ is a "characteristic temperature", that describes the trap distribution. Considering that the CuPc is mainly responsible for SCLC region, and taking $\gamma = m - 1 = 6$ we obtain $T_c \sim 1650$ K, value in good agreement with that obtained from our previous results [6,9,13].

Fig. 3. — The variation of junction resistance $R_o$ as a function of reciprocal forward current $1/I$. The insert zooms on region A of high currents.
3.2. THE PHOTOVOLTAIC PROPERTIES

3.2.1. The Action Spectra. — The photovoltaic action spectra can provide considerable information as regarding to how the charge carriers are generated. The action spectra of short-circuit photocurrents of ITO/CuPc/(CuPc+TPyP)/TPyP/Al [A] and ITO/CuPc/TPyP/Al [B] cells are shown in Figure 5, while the absorption spectra of the organic layers CuPc and TPyP are shown in Figure 6. The action spectra are measured at illumination through the ITO electrode. These are obtained by plotting the short-circuit photocurrent $J_{ph}$ divided by the number of incident photons on the ITO/CuPc interface and then normalised to unity. Although the absorption spectra of TPyP and CuPc layers, are nearly complementary (the main maxima of TPyP and CuPc layers are centered at 430 and 620 nm, respectively), both the TPyP and CuPc layers contribute to the photogeneration of carriers in the two cells. A marked feature to be noted in Figure 5 is that both curves show greater photocurrents in the range of the broad absorption band of CuPc film (at wavelength between 500 and 750 nm). In the Soret region of all action spectra presented in Figure 5, a distinct peak appears, as a result of the photons weakly absorbed in CuPc, but strongly absorbed in the Soret band of TPyP. It is
important to note that, although moderate absorption there is both in TPyP and CuPc layers at wavelength of 520 nm, a peak of photocurrent exists at this wavelength, which is greater than that from Soret band of TPyP. The filtering effect of CuPc layer has to be considered. This behaviour shows strongly that the photoactive region (i.e. in the internal field region) is at the CuPc/TPyP interface.

The photocurrent of the three-layered cell Figure 5 [A], was about ten times larger than that of two-layered cell Figure 5 [B] all over the spectral region. The thickness of the front CuPc layers is the same (80 nm) in both typed cells, i.e. the same masking effect due to CuPc layer. This result strongly suggests that the codeposited interlayer, which has a large amount
of direct molecular contacts between TPyP and CuPc molecules and consequently there is a thicker depletion region, may provide an efficient carrier photogeneration layer. It is known [2] that an efficient photogeneration mechanism is that of exciton dissociation on the dopant complex states present in the volume of an organic photoconductor. That is why, presently, we suspect that the charge photogeneration via exciplex of the two dyes, i.e. \( (\text{TPyP}^- \text{CuPc}^+) \), from codeposited layer is responsible for the efficient photogeneration of the charge carriers. The better agreement of the absorption spectrum of the codeposited layers and the action spectra of the cells illuminated through ITO electrode, suggests that the singlet excitons are the precursors of the charge carrier by the previous described process and that the photovoltaic region is located in the codeposited interlayer.

The dependence of the short-circuit photocurrent \( (J_{sc}) \) on the incident light intensity \( (I_{inc}) \), when illuminated through ITO, was examined too. At the wavelengths 520 and 590 nm, the dependence of \( (J_{sc}) \) vs. \( (I_{inc}) \) of the cell follows the relation \( J_{sc} \sim I_{inc}^m \), where the exponent \( m < 1 \). This is a common behaviour for the organic photovoltaic cells and could be explained by taking into account the model based on the combined effect of traps and recombination centers [3–5, 7].

3.2.2. The \( (J_{ph} - U) \) Characteristics. — Figures 7 [A], [B] show the \( (J_{ph} - U) \) characteristics in the fourth quadrant of \( (J - U) \) diagram of a typical ITO/CuPc/(CuPc+TPyP)/TPyP/Al cell, obtained at 520 and 590 nm, the wavelengths where a high sensitivity of the cell exists. These curves were obtained by varying the load resistance in the range 100 kΩ to 2 TΩ at constant input light intensities, which are given in the legend of each plot. Several parameters of interest such us open-circuit photovoltage \( (U_{oc}) \), short-circuit photocurrent \( (J_{sc}) \), fill factor \( (ff) \) and power conversion efficiency \( (\eta) \) can be evaluated from these curves and are listed in Table I.

a) The fill factor defined as ratio of the maximum output power density to the product \( (U_{oc}J_{sc}) \) gives a measure of how close the cell is to an ideal photovoltaic cell \( (ff = 1) \). The fill factors of this particular cell have different values ranging from 0.11 to 0.32. The low fill factors obtained for this cell are comparable to other organic photovoltaic cells [3, 5, 6, 10], but generally larger than of single and two-layered organic cells. This low values are due to

Fig. 6. — The absorption spectra of a) TPyP and b) CuPc layers.
Table I. — The typical parameters of ITO/CuPc/(CuPc+TPyP)/TPyP/Al cells.

<table>
<thead>
<tr>
<th>λ (nm)</th>
<th>Curve see Fig.8</th>
<th>$I'_\text{inc}$ ($\mu$W cm$^{-2}$)</th>
<th>$J_\text{sc}$ (nA cm$^{-2}$)</th>
<th>$U_\text{oc}$ (mV)</th>
<th>$f_f$ (%)</th>
<th>$\eta$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>520</td>
<td>[a]</td>
<td>3</td>
<td>107</td>
<td>75</td>
<td>0.32</td>
<td>0.09</td>
</tr>
<tr>
<td>520</td>
<td>[b]</td>
<td>12</td>
<td>410</td>
<td>220</td>
<td>0.29</td>
<td>0.22</td>
</tr>
<tr>
<td>520</td>
<td>[c]</td>
<td>30</td>
<td>916</td>
<td>405</td>
<td>0.28</td>
<td>0.35</td>
</tr>
<tr>
<td>590</td>
<td>[a]</td>
<td>6</td>
<td>136</td>
<td>87</td>
<td>0.23</td>
<td>0.05</td>
</tr>
<tr>
<td>590</td>
<td>[b]</td>
<td>30</td>
<td>528</td>
<td>277</td>
<td>0.31</td>
<td>0.15</td>
</tr>
<tr>
<td>590</td>
<td>[c]</td>
<td>72</td>
<td>1250</td>
<td>350</td>
<td>0.11</td>
<td>0.07</td>
</tr>
</tbody>
</table>

Fig. 7. — Photocurrent-photovoltage characteristics of the ITO/CuPc/(CuPc+TPyP)/TPyP/Al cell illuminated through ITO electrode with: [A] 520 nm wavelength of varying intensity as $3 \mu$W cm$^{-2}$ [a], $12 \mu$W cm$^{-2}$ [b] and $30 \mu$W cm$^{-2}$ [c]); [B] 590 nm wavelength of varying intensity as $6 \mu$W cm$^{-2}$ [a], $30 \mu$W cm$^{-2}$ [b] and $72 \mu$W cm$^{-2}$ [c]).
two separate effects, namely, the high series resistances and the field-dependent nature of the photogeneration of charge carriers [2,8]. The highest value of fill factor was obtained at 520 nm for 3 \( \mu \)W cm\(^{-2}\), and smaller values were obtained for higher light intensities at all wavelengths. 

b) The power conversion efficiency has been evaluated from the following relation

\[
\eta(\%) = \frac{ff \times J_{sc} \times U_{oc}}{P_{inc}} \times 100
\]

where \( P_{inc} \) is the light power density incident on the cell corrected for the transparency of ITO electrode. The efficiencies calculated at 520 and 590 nm, for various values of the input power are listed in Table I. The better performance of the cell are obtained for more penetrating 520 nm wavelength where the values 0.09, 0.22 and 0.35% for 3, 12 and 30 \( \mu \)W cm\(^{-2}\) input powers have been resulted. These values of \( \eta \) are about 2-3 times larger than those of a two-layer cell and are among the best reported for organic solar cells. The better performance of the cell at shorter wavelength is most probable due to the bulk ionisation of more energetic 520 nm excitons into charge carriers, mainly in TPyP layer. One can observe that for this wavelength the efficiency increases when the light intensity increases. At longer wavelength, such as 590 nm, the efficiency increases when the intensity increases up to 30 \( \mu \)W cm\(^{-2}\) and decreases when the light intensity increases further. This decrease is mostly due to the above evaluated dependence of \( J_{sc} \) on \( P_{inc} \) and can be understood in terms of a physical model involving traps and recombination centres [16]. As the light intensity is increased, the electron and hole densities increase and therefore the quasi-Fermi levels are moving toward the conduction and valence bands, respectively. As the light intensity is further increased, more and more of the trapping states are converted into recombination centres, thus reducing the lifetime of charge carriers and resulting into nonlinear variation of photocurrent with light intensity. As regarding the open-circuit photovoltage, both double- and three-layered cells showed almost the same magnitude for \( U_{oc} \), about 0.4 V as an average value. This result tells us that the built-in potential for both typed cells originates from the difference of Fermi levels between CuPc and TPyP layers. We suppose that most of built-in potential is distributed across the interlayer. Since positive charges of ionic donors in n-type TPyP and negative charges of ionic acceptors in p-type CuPc are compensated with each other, the codeposited layer behaves like an intrinsic semiconductor and then the three-layered cell resembles to a p-i-n a-Si:H based solar cell.

4. Conclusion

The new type of three-layered organic photovoltaic cells having a codeposited layer of (CuPc+TPyP), between the CuPc and TPyP films, clearly suggest an improvement, although modest, over two-layered cells. The codeposited interlayer, has been found to act as an efficient carrier photogeneration layer because: on one hand, the built-in potential drops across it and on the other hand, there is the region of maximum photogeneration rate, as a result of exciton dissociation via the exciplex of (CuPc\(^{-}\)..TPyP\(^{+}\))\(^{*}\) dyes. The fill factors of 0.11-0.32 represent an improvement over single and double-layered cells. Moreover, the power conversion efficiencies of three-layered cells, ranging from 0.07 to 0.35%, are 2-3 times greater as compared to those for double-layered cells. The thickness of about 0.4 \( \mu \)m of our three-layered cells is mainly responsible for the high internal resistance, which limits again the performances of the cells. By obtaining an optimised structure as regarding the thickness of the layers and the architecture of the cells, we are convinced that substantial improvement of the organic photovoltaic cells would be possible.
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