

# *Analysis of the stability of organic photovoltaic devices through external quantum efficiency (EQE)*

## *Análise da estabilidade de dispositivos fotovoltaicos orgânicos através da eficiência quântica externa (EQE)*

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### **Abstract**

The use of solar panels for the purpose of converting solar energy into electrical energy has been increasingly common in the community, where the vast majority of these panels are produced from inorganic materials, especially silicon (Si). In recent decades, many researchers have been analyzing organic and hybrid materials in order to apply them to photovoltaic devices. The use of organic and hybrid materials in devices is advantageous due to some factors, such as: low production cost, variety of materials available, production of flexible devices. A fundamental analysis for any photovoltaic device is the external quantum efficiency measure (EQE). This technique correlates the number of incident photons with the number of electrons generated, making it possible to know in which region of the electromagnetic spectrum the photovoltaic device is more efficient. This work aims to apply the external quantum efficiency characterization technique for the evaluation of different types of photovoltaic devices in terms of their stability.

**Keywords:** External quantum efficiency; photovoltaic devices; hybrid materials; organic materials.

### **Resumo**

A utilização de painéis solares com a finalidade de converter energia solar em energia elétrica tem sido cada vez mais comum na comunidade, onde a grande maioria desses painéis é produzida a partir de materiais inorgânicos, principalmente o silício (Si). Nas últimas décadas, muitos pesquisadores vêm analisando materiais orgânicos e híbridos para aplicá-los em dispositivos fotovoltaicos. A utilização de materiais orgânicos e híbridos em dispositivos é vantajosa devido a alguns fatores, como: baixo custo de produção, variedade de materiais disponíveis, produção de dispositivos flexíveis. Uma análise fundamental para qualquer dispositivo fotovoltaico é a medida de eficiência quântica externa (EQE). Essa técnica correlaciona o número de fótons incidentes com o número de elétrons gerados, possibilitando saber em qual região do espectro eletromagnético o dispositivo fotovoltaico é mais eficiente. Este trabalho tem como objetivo aplicar a técnica de caracterização de eficiência quântica externa para a avaliação de diferentes tipos de dispositivos fotovoltaicos em termos de estabilidade.

**Palavras-chave:** Eficiência quântica externa; dispositivos fotovoltaicos; materiais híbridos; materiais orgânicos.

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## Introduction

The solar energy received at the Earth's surface per year is approximately 120,000 TW, which is approximately 6 times more than the current global energy consumption (TAI; YAN, 2017). Recently, the rapid development of organic solar cells (OPVs) has increased an energy conversion efficiency (PCE) of less than 3% to more than 18% (CHAKRABORTY *et al.*, 2014; XIONG *et al.*, 2020). Meanwhile, OPVs have great advantages of light weight, easy fabrication, low cost, and less polluting compared to traditional silicon-based solar cells (GAO *et al.*, 2020). Many efforts have been made to improve the efficiency and stability of OPVs, such as the adoption of solvent additives, using several active layers of heat treatments, employing ternary strategy, and inverted structure cells (HU *et al.*, 2020).

When targeting the performance of a photovoltaic (PV) device a good indicator is the external quantum efficiency (EQE), also known as incident photon conversion efficiency (IPCE), referring to the ratio of generated electrons to the number of photons incidents of a certain wavelength. In practical terms, it is a measure of the spectral response of the photovoltaic cell to different wavelengths of light (BONDARENKO *et al.*, 2018; LI *et al.*, 2014).

An ideal solar cell would demonstrate an EQE of 100% over the entire absorption spectrum, with all photons generating respective electrons at any wavelengths up to the corresponding gap of the absorbing material, considering that at the longest wavelengths of this limit, photons will lack enough energy to lift electrons across gap bands from HOMO (highest occupied molecular orbital) to LUMO (lowest unoccupied molecular orbital). However, even in the range of wavelengths corresponding to a specific gap, in a real solar cell there will be a reduction in the EQE response due to various effects. A typical situation is in the blue response (short wavelength), reduced due to absorption by the optical window layer, e.g. ITO glass (JIANG *et al.*, 2020; KLEEBE; TURQUAT; SORARÙ, 2001), as well as the longer wavelength response, reduced due to the decrease in the absorption coefficient beyond the bandgap level. Also, there are some general problems such as charge decoupling, interphase problems, and thin film defects.

In this context, EQE measurements provide important information about the electrical current that a cell will produce for a given wavelength. If the EQE is integrated into the incident spectrum (which depends on the energy

source - sun, lamp, etc.), the photocurrent that the cell will produce in response to this light source can even be predicted. Incidentally, it is also an important tool to understand the effectiveness of three main processes: the optical absorption of light creating the charge carriers; the electrical transport of those carriers that contribute to the general current; and the capture of excess carriers generated by the defects, leading to capture or recombination (BARNES *et al.*, 2009; BRINKMANN *et al.*, 2021; ZHANG *et al.*, 2020).

The aim of this work was to carry out a study on the photostability of different organic photovoltaic devices and to present comparative results when inserting an additive (1,8-octanedithiol) in the active layer of the particular PCPDTBT:PCBM device. Regarding the monitoring of the photo-stability of the devices, this occurred through measurements of the EQE curves over the time of light exposure, whose methodological details and results achieved are mentioned later. And as another objective, present an alternative for groups that already work with characterization of materials and have monochromators to perform characterization of photovoltaic devices without the immediate acquisition of a solar simulator.

## Materials and methods

For the UV-Vis (ultraviolet-visible) absorption measurements of the active layers of organic photovoltaic devices, a Shimadzu UV-3600 spectrophotometer was used.

Cell devices were prepared on pre-patterned ITO glass slides with a surface resistivity of 16  $\Omega$ /square. The ITO slides were cleaned using deionized water, acetone, and isopropyl alcohol (IPA) and blown dry using nitrogen. The slides were then placed in an oxygen plasma for 10 min. A layer of the conductive polymer PEDOT:PSS mixture was spin coated (in air) onto the substrate at 4500 rpm for 30 s, before a 60 min heat at 60 °C on a hotplate to remove any residual H<sub>2</sub>O.

Devices were fabricated and tested in a glovebox, which maintained H<sub>2</sub>O and O<sub>2</sub> concentrations to typically less than 0.1 part per million. Active layers of PCPDTBT:PCBM with weight ratios of 1:3 were prepared and dissolved in chlorobenzene solvent with a concentration of 30 mg/ml, using the same manner as previous papers by this group on PCPDTBT (CHANG *et al.*, 2012; KETTLE *et al.*, 2011; KETTLE *et al.*, 2012), where the optimization of polymer: fullerene blend ratio,

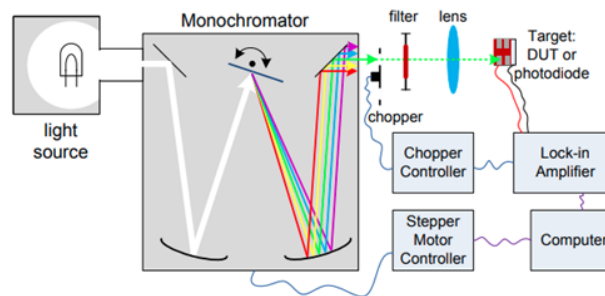
film thickness and annealing conditions have already been rigorously studied. For devices with alkanethiol additive, 1,8-octanedithiol (ODT), Sigma-Aldrich (03605), was mixed with the initial blend with a concentration of 3% by weight. The solutions were then allowed to equilibrate for 24 h on a hot plate stirrer at 60 °C and then filtered using a 0.45  $\mu\text{m}$  PTFE filter. Active layers were applied by spin-casting in a glovebox from a 60 °C heated solution (2200 rpm for 60 s for devices without ODT, 2500 rpm for devices with ODT). It is important to note that ODT possesses a high boiling point of 269 °C and is unlikely to be lost from the active layer. The active layer of P3HT:PC61BM, in a proportion of 17:13 in mass, was dissolved in chlorobenzene, resulting in a solution of 30 mg/ml of concentration. The solution was stirred for 24 h and then it was deposited by spin coating on the substrates. A rotation of 1500 rpm for 60 s was used. After that, the samples were heated at 150 °C for half an hour. Aluminium electrodes were deposited by thermal evaporation. All the reactants were obtained from Sigma Aldrich.

Finally, a 5 nm layer of Calcium (Ca) and an 80 nm layer of Aluminium (Al) were deposited on each sample by thermal evaporation through a shadow mask to form a composite cathode electrode, which defined six active areas of 0.2  $\text{cm}^2$ .

Measurements of the External Quantum Efficiency (EQE) were conducted periodically to follow the trend as light soaking proceeded. They were made with a monochromator with nm resolution (Bentham) and an SRS lock-in amplifier. A Xenon quartz light source (Osram HLX64623) with an output spectrum from 350 nm to 1000 nm was used and measurements were taken every 2 nm over a range of 375–900 nm. A silicon photodiode (Thorlabs SM1PD1A) was used to make the reference measurement. In Figure 1, an experimental scheme of the EQE measurement is shown.

Two order sorting filters were used to ensure that the correct wavelength was being used to illuminate the devices. The monochromator did not separate out a single frequency of light but also allowed harmonics of this frequency through. This was found when the output was checked using an Ocean Optics HR4000 spectrometer. This is a reasonably common occurrence in older monochromator systems as a result of diffraction gratings. For EQE measurements, only one device was used and was light soaked in between measurements.

**Figure 1** – EQE measurement system schematic.



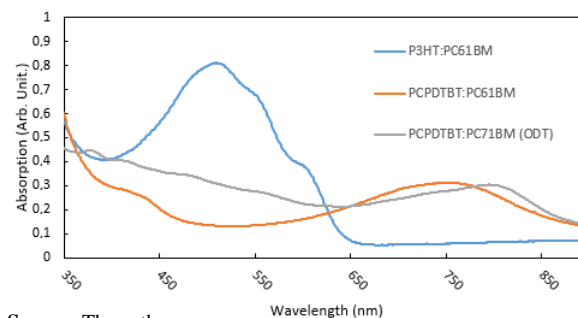
Source: Bristow (2017).

The devices were tested using a Newport solar simulator with  $100\text{mWcm}^2$  AM1.5 output calibrated using a silicon reference cell verified by (RERA, Netherlands). Subsequent light soaking measurements were provided from a halogen light soaker (GB-Sol Ltd, Cardiff, UK). This was conducted in accordance with ISOS-L-2 standards (REESE *et al.*, 2011). The device temperature was not controlled but remained at  $50\text{ °C} \pm 5\text{ °C}$  during the measurements. Both devices were measured for 601 h.

## Results and discussions

Figure 2 shows the absorbance spectra of the materials used as the active layer of the organic photovoltaic devices analyzed in this study.

**Figure 2** – Absorbance spectra of thin films used as active layer of organic photovoltaic devices.



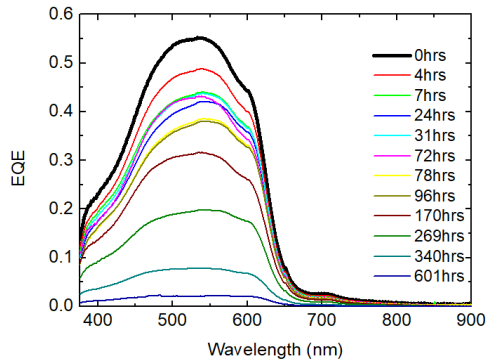
Source: The authors.

From the spectra shown in Figure 2, it is possible to state that the active layers containing the PCPDTBT polymer have a wider absorbance (350 nm – 900 nm) when compared to the layers containing P3HT (350 nm – 620 nm). In practical terms, a greater absorption range for photovoltaic devices is favorable, as it covers a larger region of the solar spectrum, making it possible to maximize the decoupling of charges and, consequently, increase the photocurrent of the device.

In this consideration, it is also verified that the insertion of the ODT generates an increase in the absorption in the region of (400 nm – 600 nm).

Figure 3 shows a variation of the EQE curve, here obtained from an organic photovoltaic device of P3HT:PCBM exposed to the irradiation of a solar simulator.

**Figure 3** – EQE curve of the P3HT device after different lighting exposure times of the solar simulator.



Source: The authors.

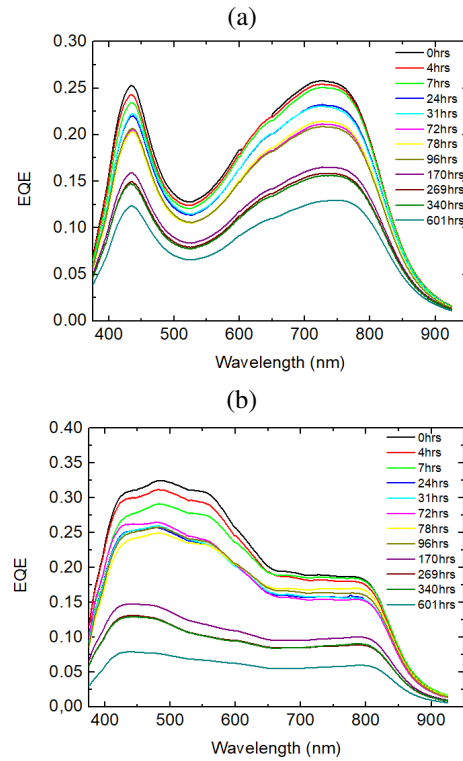
Figure 3 indicates that the EQE curve of the P3HT device suffers an intensity drop with the time of exposure to irradiation, a result expected in the consideration of the device not being encapsulated, that is, exposed to atmospheric conditions (CHANDER; SINGH; IYER, 2017).

Figure 4 shows a variation of the EQE curve of a photovoltaic device with an active layer of PCPDTBT:PCBM at different times of exposure to irradiation from a solar simulator, where Figure 4(a) represents the variation of the EQE curve of the device without ODT and, Figure 4(b), the variation of the EQE curve of the device with ODT.

Due to the addition of ODT in the active layer of the photovoltaic device, as noted in Figure 4, a change in the line shape of the EQE curve of the device is obtained, thus verified by comparing Figures 4(a) and 4(b), prevailing compliance with the literature (KETTLE *et al.*, 2015; WATERS *et al.*, 2014). Both Figures 4a and 4b show the effect of the low band polymer causing absorption up to about 850 nm. However, due to the incorporation of ODT (COATES *et al.*, 2008; HORIE *et al.*, 2010), it is also observed that the EQE curve increases strongly in the region between 450 nm and 600 nm.

In order to analyze and compare the stability of these devices, it is appropriate to use the relation given by equation (1)

**Figure 4** – EQE curves at different exposure times to lighting of photovoltaic devices with active layer of PCPDTBT:PCBM (a) without ODT and (b) with ODT.



Source: The authors.

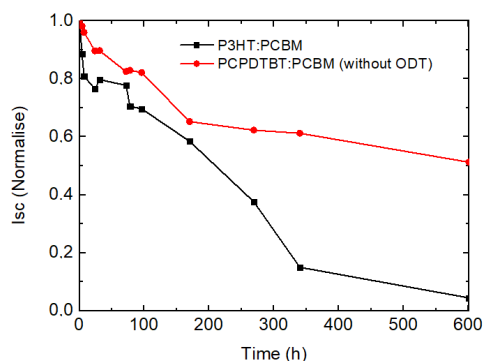
$$I_{sc} \propto \int_{\lambda_i}^{\lambda_f} (EQE)_{\lambda} d\lambda \quad (1)$$

Relation, equation (1), shows that the short circuit current  $I_{sc}$  is proportional to the integral of the EQE curve of the device (BAI *et al.*, 2021; QIAN *et al.*, 2021). To disregard the initial efficiency of each device, the EQE curve integral was calculated for the different measured times and normalized by the initial value for each device.

Initially, a comparison was made based on the short circuit current (calculated and normalized by the initial EQE curve) at different exposure times. The first devices compared were those with different active layers (P3HT:PCBM and PCPDTBT:PCBM), analyzing the results obtained in Figure 5.

Figure 5 shows that the photovoltaic device, whose active layer is PCPDTBT:PCBM, has a higher stability when compared to the active layer device of P3HT:PCBM. This sharper drop in P3HT:PCBM devices is associated with oxygen-induced degradation, which causes a strong reduction in the charge recombination coefficient, which formally corresponds to a reduction in charge carrier mobility.

**Figure 5** – Variation of the short circuit current over the exposure time of the different P3HT (black curve) and PCPDTBT (red curve) devices.

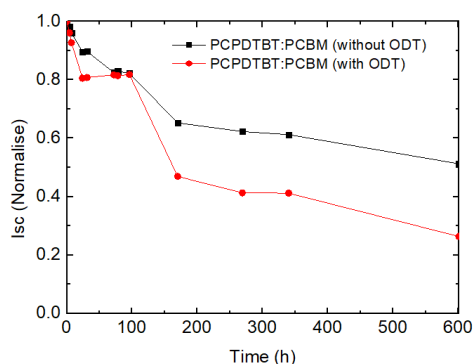


Source: The authors.

This is due to the formation of PCBM dimers when the device is under illumination (ALVES, 2017; YAN *et al.*, 2017). Another result verified in accordance with the literature is the fact that P3HT has a lower stability when exposed to light compared to PCPDTBT (TROMHOLT *et al.*, 2011).

Then, devices of the same active layer (PCPDTBT:PCBM) were compared, differentiating or not the insertion of the additive. The next Figure 6 characterizes the variation of the short circuit current obtained by equation 1 of the EQE curves for the different exposure times and normalized by the initial value.

**Figure 6** – Short-circuit current variation over time of exposure of PCPDTBT devices without ODT (black curve) and PCPDTBT with ODT (red curve).



Source: The authors.

The drop in I<sub>sc</sub>, seen in Figure 6, is different for devices produced with and without ODT. With this, it is possible to affirm that the degradation in the electrodes is not the only reason for the decrease in the performance of the photocell. It is reported in the literature that the insertion of ODT promotes greater change in morphology,

which explains the worse performance of the PCPDTBT solar cell with ODT (KETTLE *et al.*, 2015). It is also reported that the decrease in I<sub>sc</sub> may be due to a number of mechanisms; however, this is likely to be due to chemical degradation in PCPDTBT and PCBM (WATERS *et al.*, 2014). The inclusion of ODT leads to enrichment of PCPDTBT on the upper surface with aging, thus leading to efficient orifice transport from device to electrode (KETTLE *et al.*, 2015).

## Conclusion

The results indicate that the insertion of the ODT showed a significant improvement in the EQE curve for the PCPDTBT:PCBM devices. However, regarding the stability of these devices in the face of such insertion, the analysis of that curve allows us to conclude that, for PCPDTBT:PCBM devices without encapsulation, the insertion of ODT does not become attractive to practical interests. It was also found that P3HT:PCBM devices are more sensitive to photobleaching under ambient conditions when compared to PCPDTBT:PCBM devices. Finally, the results also conclude a corroboration of the feasibility of the EQE technique to carry out studies on the stability of photovoltaic devices, which may serve as an alternative for groups that already work with characterization and have monochromators not needing to immediately acquire a solar simulator.

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