

Design of a p-i-n type inverted perovskite solar cell using SiO_x as down-conversion material to improve PCE: Simulation and optimization in SCAPS-1D

Diseño de una celda solar de perovskita tipo p-i-n utilizando SiO_x como material de energía de conversión descendente para mejorar el PCE: Simulación y optimización en SCAPS-1D

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Abstract

In this research work, an inverted p-i-n type perovskite solar cell: ITO/PEDOT: PSS/CH₃NH₃PbI₃/PCBM/Au has been simulated and optimized in SCAPS-1D. The optimized parameters in SCAPS-1D that improved solar performance were: perovskite thickness, the total defect density of perovskite, the total defect density of interfaces, series and shunt resistances, and device operating temperature. As a result, the efficiency (PCE) increased to 18.33%. Subsequently, when the silicon-rich oxide (SiO_x) material was implemented in the simulation as down-conversion energy material on the outside of the cell, a power conversion efficiency (PCE) of 23.7% was obtained. The SiO_x film obtained experimentally by sputtering obtained good photoluminescence, absorption coefficient, band gap, and transmittance characteristics before and after thermal annealing. These characteristics have been considered for the proposed device. It is indicated that the inverted perovskite solar cell of type p-i-n: SiO_x/ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Au has better J-V output values and EQ quantum efficiency than the perovskite solar cell without SiO_x.

Perovskite, Efficiency, Simulated

Resumen

En este trabajo de investigación, una celda solar de perovskita tipo p-i-n invertida: ITO/PEDOT: PSS/CH₃NH₃PbI₃/PCBM/Au ha sido simulada y optimizada en el software SCAPS-1D. Los parámetros optimizados en SCAPS-1D que mejoraron el rendimiento solar fueron: el grosor de la perovskita, la densidad total de defectos de la perovskita, la densidad total de defectos de las interfaces, las resistencias en serie y derivación y la temperatura de operación del dispositivo. Como resultado de esto, la eficiencia (PCE) incrementó a 18.33%. Posteriormente, cuando se implementó en la simulación el óxido rico en silicio (SiO_x) como material de energía de conversión descendente en la parte externa de la celda, se obtuvo una eficiencia (PCE) de 23.7%. La película de SiO_x obtenida experimentalmente por sputtering obtuvo buenas características de fotoluminiscencia, coeficiente de absorción, band gap y transmitancia antes y después del recocido térmico. Estas características fueron consideradas en el dispositivo propuesto. Se concluye que la celda solar de perovskita: SiO_x/ITO/PEDOT: PSS/CH₃NH₃PbI₃/PCBM/Au tiene mejores valores de salida J-V y eficiencia cuántica EQ que la celda solar sin SiO_x.

Perovskita, Eficiencia, Simulado

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Introduction

Perovskite solar cells (PSCs) have become a principal source of research since their emergence in 2009. They are currently the most promising devices of the future for use as a renewable energy source. In just a few years, they have made remarkable progress as their efficiency has been certified up to 25.5 %, comparable to silicon (Si), gallium arsenide (GaAs), and cadmium telluride (CdTe) technology, according to Chao *et al.*, (2021). Until now, most perovskite solar cells (PSCs) have been fabricated with the traditional vertical or n-i-p type structure. Notwithstanding, these devices suffer from severe hysteresis effects and have poor stability. In contrast, the inverted or p-i-n type cell has good, fewer defective states between interfaces it is possible to fabricate devices at low temperatures (Yang *et al.*, 2022).

Notwithstanding, metal-organic perovskite ($\text{CH}_3\text{NH}_3\text{PbI}_3$) remains one of the materials used in perovskite solar cells due to its simple synthesis process, low cost, and high performance (Yin *et al.*, 2022). Nevertheless, its instability and easy degradation due to environmental factors are still an obstacle to its commercialization (Mazumdar *et al.*, 2021). Perovskite $\text{CH}_3\text{NH}_3\text{PbI}_3$ has many advantages as a high absorption coefficient, tunable band gap, high charge carrier mobility, low trap density, long carrier diffusion length, and small exciton binding energy (Mohanty *et al.*, 2019).

Researchers are exploring a lot in the field of perovskites as the use of different structures in solar cells, the design of new hole and electron transport materials, the improvement of perovskite materials, and the implementation of interlayers to achieve higher efficiency and improved spectral response. Improving the utilization of the incident solar spectrum may be one of the approaches to increase the performance of these devices because currently, perovskite solar cells can only utilize a fraction of the incident solar photons, ranging from the visible to the near infrared (Datt, Bishnoi, Lee, *et al.*, 2022). That is ultraviolet (UV) regions are not used and are considered harmful.

According to the literature, a down-conversion energy material absorbs the short wavelength photons of the solar spectrum, which the solar cell does not use efficiently, and re-emits them at longer wavelengths (Hosseini & Ghanbari, 2018). The main characteristics of a down-conversion energy material are high photoluminescence quantum efficiency (PLQE), photochemical and environmental stability, broadband absorption in the region where the spectral response of the solar cell is low, high absorption coefficient in low wavelength region, high transmittance and broadband emission, particularly in the region where the device response is high, sufficient Stokes shift to minimize the self-absorption energy losses due to the spectral overlap between the absorption and emission bands, low cost, easy to process and can be deposited by the large scalable method, and low film roughness (Datt, Bishnoi, Hughes, *et al.*, 2022).

An alternative to solve spectral mismatch losses, improve stability, and increase efficiency is to apply a silicon-rich oxide (SiO_x) layer on the surface of a perovskite solar cell. This material is a silicon oxide out of stoichiometry that contains small silicon islands embedded in a silicon dioxide matrix and stands out for its high photoluminescence intensity of red light when illuminated with ultraviolet UV radiation (Vivaldo *et al.*, 2016).

In this work, the photovoltaic characteristics of a SiO_x film deposited by co-sputtering have been reviewed for implementation in perovskite solar cells. Subsequently, an inverted perovskite solar cell type p-i-n: ITO/PEDOT: PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PCBM/Au was numerically simulated. Photovoltaic parameters such as perovskite thickness, total defect densities, series and shunt resistances, and operating temperature were optimized for this device, reaching an efficiency of 18.33 %. The J-V output values were compared with those reported experimentally. Finally, a new device model was proposed: SiO_x /ITO/PEDOT: PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PCBM/Au, which includes a down-conversion energy material (SiO_x) on the outside, and whose efficiency increased to 23.74 %. In the latter simulation, the J-V output characteristics and QE quantum efficiency were better than those of the device without SiO_x .

Numerical methodology

Solar cell structure

The simulated inverted perovskite solar cell has the configuration ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Au, where indium doped tin oxide (ITO) is the front contact; poly (3,4- ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) is the hole transport layer; CH₃NH₃PbI₃ is the absorber layer; Phenyl-C61-butyric acid and methyl ester (PCBM) is the electron transport layer; and gold (Au) is the back metal contact. In addition to the proposed model SiO_x/ITO/PEDOT/CH₃NH₃PbI₃/PCBM/Au is a device that includes an outer layer of silicon-rich oxide (SiO_x), which works as a down-conversion energy material. The two solar structures are represented in Figure 1 (a, b) and the energy band diagram is shown in Figure 1 (c).

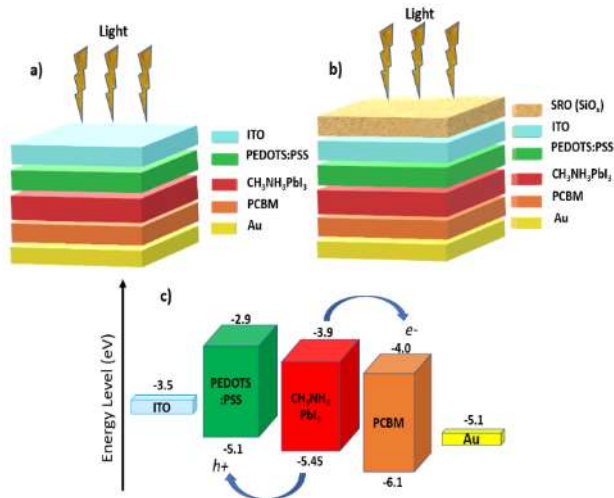


Figure 1 (a) Schematic diagram of perovskite solar cell; (b) Schematic diagram of perovskite solar cell with SiO_x; (c) Energy band gap diagram

Source: Own elaboration, Paint 3D

Numerical method

The simulation was performed in SCAPS-1D software version 3.3.10 under AM1.5G illumination at 300 K ambient temperature. The SCAPS-1D software, developed at the Department of Electronics and Information Systems (ELIS) at the University of Ghent in Belgium, has been used to model and simulate solar cells (Burgelman *et al.*, 2000). The software solves the semiconductor equations in one dimension in a steady state (Decock *et al.*, 2013) and (Burgelman *et al.*, 2013).

The main equations it solves are the Poisson equation (1), the electron continuity equation (2), the hole continuity equation (3), the electron charge transport equation (4), the hole charge transport equation (5), and the absorption coefficient (6), which are solved until convergence occurs (Verschraegen & Burgelman, 2007).

Poisson equation:

$$\frac{dE}{dx} = \frac{q}{\epsilon_0 \epsilon_r} [p(x) - n(x) + N_D^+(x) - N_A^-(x) + p_t(x) - n_t(x)] \quad (1)$$

Where E is the electric field, q is electronic charge, ϵ_0 is permittivity of vacuume, ϵ_r is relative permittivity, N_D^+ is the shallow donor impurity density, N_A^+ is the shallow acceptor impurity density, $n(x)$ and $p(x)$ are the densities of electrons and holes. Similarly $p_t(x)$ y $n_t(x)$ represent the trapped holes and electrons as a function of x respectively.

Continuity equations:

$$\frac{dJ_n}{dx} = G - U_n \quad (2)$$

$$\frac{dJ_p}{dx} = G - U_p \quad (3)$$

Where J_n and J_p are the electron and hole current densities, G is the generation rate, U_n and U_p are the recombination rates for electrons and holes respectively.

Charge transport equation:

$$J_n = D_n \frac{dn}{dx} + \mu_n n \frac{d\phi}{dx} \quad (4)$$

$$J_p = D_p \frac{dp}{dx} + \mu_p p \frac{d\phi}{dx} \quad (5)$$

Where D_n and D_p are the electron and hole diffusion coefficients respectively, μ_n and μ_p are the electron and hole mobility respectively.

Absorption coefficient equation:

$$\alpha(\lambda) = \left(A + \frac{B}{h\nu}\right) \sqrt{h\nu - E_g} \quad (6)$$

Where A and B are constants, h is Planck constant, ν is frequency of photon and E_g is the band gap of the absorber layer.

Simulation parameters

The physical, optical, and electrical parameters were obtained from the scientific literature and summarized in Table 1 (Basyoni *et al.*, 2021; A. Ghosh *et al.*, 2020; B. Ghosh *et al.*, 2022; Jamal *et al.*, 2019; Sabbah *et al.*, 2022). The parameters used for the silicon-rich oxide SiO_x correspond to a SiO_2 film. Nevertheless, the absorption coefficient, the photoluminescence, and the band gap are the results of the characterization of the deposits obtained by co-sputtering for this work. In addition, the work function for the metal contact (Au) was 5.1 eV.

Photovoltaic Parameters	SiO_x	ITO	PEDOTS: PSS	$\text{CH}_3\text{HN}_3\text{PbI}_3$	PCBM
Thickness (nm)	90	100	40	300	80
Energy Band gap, E_g (eV)	4.5	3.5	2.2	1.57	2.1
Electron affinity, χ (eV)	0.950	2.3	2.9	3.9	4.1
Dielectric permittivity, ϵ_r	3.9	9	2.3	18	4
Effective Density of states at conduction band, N_c ($1/\text{cm}^3$)	2.8×10^{18}	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.5×10^{19}
Effective Density of states at valence band, N_v ($1/\text{cm}^3$)	2.6×10^{19}	1.8×10^{19}	2.8×10^{18}	1.9×10^{19}	2.5×10^{19}
Electron thermal velocity, V_h (cm/s)	1×10^7	1×10^7	1×10^7	1×10^7	1×10^7
Hole thermal velocity, V_e (cm/s)	1×10^7	1×10^7	1×10^7	1×10^7	1×10^7
Hole mobility, μ_n (cm^2/Vs)	1.5×10^3	31	0.0002	3	0.01
Electron Mobility, μ_p (cm^2/Vs)	4.5×10^3	50	0.02	17	0.01
Uniform Shallow Donor Dopping, N_D ($1/\text{cm}^3$)	0	0	0	0	5×10^{17}
Uniform Shallow Acceptor Dopping, N_A ($1/\text{cm}^3$)	2.0×10^{18}	2.0×10^{16}	1×10^{18}	0	0
Defect density, N_t ($1/\text{cm}^3$)	10^{15}	10^{15}	10^{15}	10^{15}	10^{15}

Table 1 Optical and electrical parameters for simulation
Source: Own elaboration, summary of bibliography

Figure 2 shows the definition and editing panel in the SCAPS-1D software. In this section, you can enter and edit the electrical and optical parameters of the layers, the direction of light incidence, and the interface parameters.

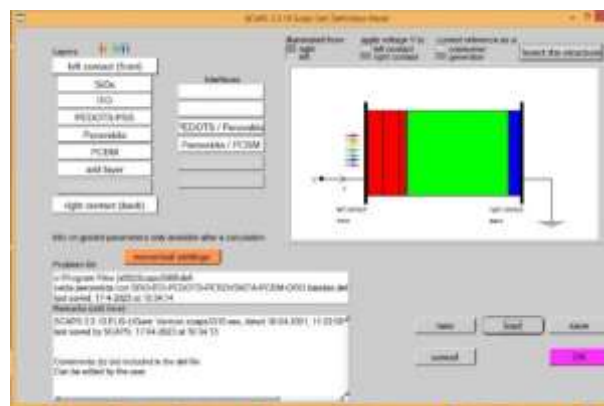


Figure 2 Definition and editing panel in the SCAPS-1D simulator

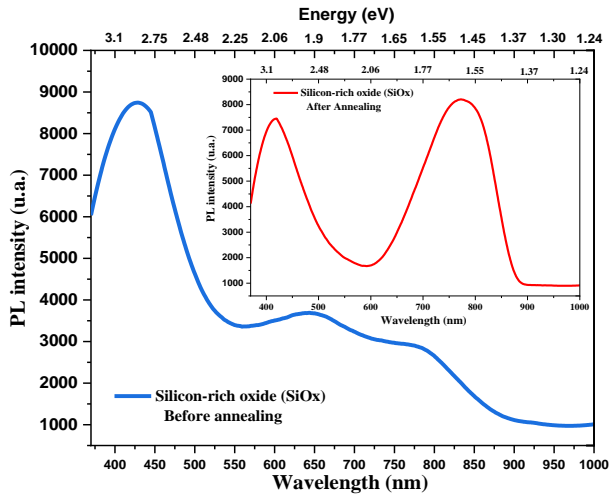
Source: SCAPS-1D Software version 3.3.10

Results and discusión

Photoluminescence, absorption coefficient, band gap, and transmittance of SiO_x

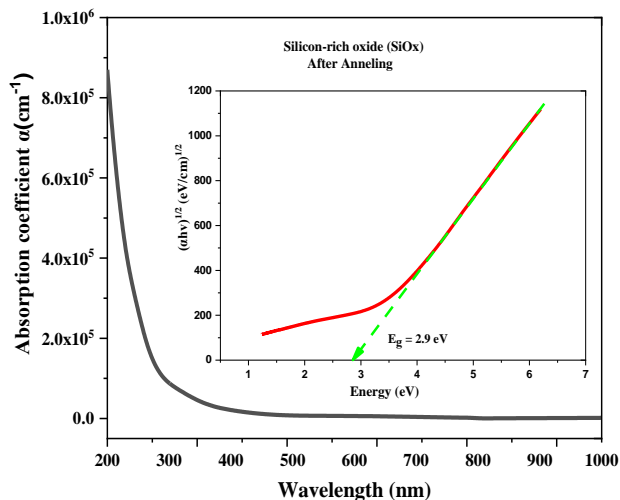
The silicon-rich oxide (SiO_x) film was deposited by co-sputtering on 1" x 1" x 1 mm quartz substrates, obtaining a thickness of 90 nm with low roughness. The SiO_x film was deposited by simultaneous sputtering of Si and SiO_2 targets. The equipment used for the deposition was a magnetron sputtering system. The deposition parameters were estimated to reach an average silicon excess (5.9 at. %) to achieve a maximum photoluminescence emission in the red region, according to Coyopol *et al.*, (2018).

Graph 1 shows the PL photoluminescence spectrum, where it is observed that SiO_x has PL emission in the red-blue region before and after thermal annealing. SiO_x before annealing shows a dominant emission peak at 423 nm, with two smaller peaks at 640 nm and 780 nm after deconvolution. After thermal annealing at 1100°C for 2 hours, the film obtained two emission peaks, one in the red region (775 nm) and another of lower intensity in the blue region (418 nm). The PL emission peaks are associated with the complete activation of Si=O defects, favored by the average size of silicon nanocrystals embedded in a silicon-rich oxide dielectric matrix (Coyopol *et al.*, 2016).



Graph 1 Photoluminescence spectra of silicon-rich oxide (SiO_x) before and after thermal annealing
Source: Own elaboration, OriginPro 2018

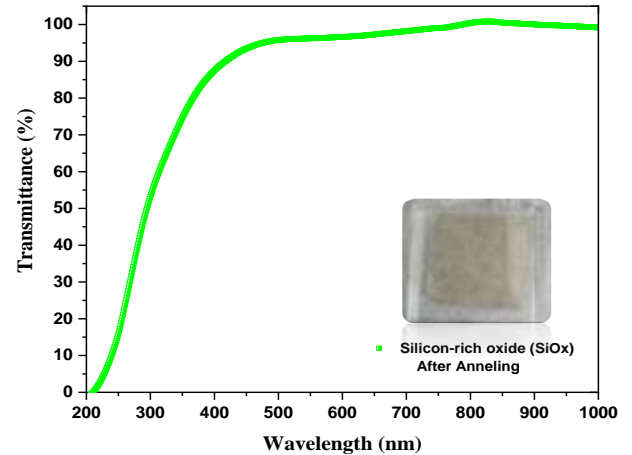
Graph 2 shows the absorption coefficient and band gap obtained from the SiO_x film after thermal annealing. The graph shows that the film has a high absorption coefficient in the low wavelength region (ultraviolet UV). In addition, the film has a low absorption coefficient in the range from 350 nm to 800 nm, which corresponds to the visible spectrum. The band gap energy was estimated using the Tauc relation and the Beer Lambert equation. This absorption coefficient and the band gap have been used in the SCAPS-1D simulator.



Graph 2 Absorption coefficient of silicon-rich oxide (SiO_x)
Source: Own elaboration, OriginPro 2018

Graph 3 shows the transmittance spectrum of the SiO_x film after thermal annealing. The graph shows that the film has a transmittance above 90% in the range from 400 nm to 800 nm, which is the visible spectrum.

In addition, the film shows a progressive decrease in transmittance in the 200 nm to 400 nm range, which corresponds to the ultraviolet spectrum. These results agree with the absorption coefficient, concluding that the spectral response of the SiO_x film is concentrated in the ultraviolet region.



Graph 3 Transmittance spectrum of silicon-rich oxide (SiO_x)
Source: Own elaboration, OriginPro 2018

J-V current-voltage curve, EQ quantum efficiency, and band diagram

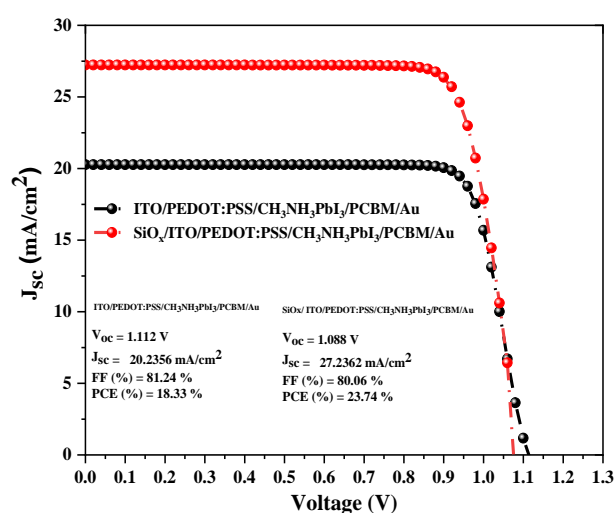
This section shows the J-V plot, QE quantum efficiency plot, and energy band diagram of the perovskite solar cell: ITO/PEDOT:PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PCBM/Au and the proposed perovskite solar cell: SiO_x /ITO/PEDOT:PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PCBM/Au. Furthermore, we have considered the optimum values obtained from the SCAPS-1D simulation. Table 2 shows the optimal values for perovskite thickness, total defect density, series and shunt resistance, and solar cell operating temperature. The parameters were varied in the simulator, keeping the other parameters constant.

Parameters	Variation range	Optimal value
Perovskite thickness	50-1000 nm	500 nm
Perovskite defect density	10^{10} - 10^{20} cm^{-3}	10^{13} cm^{-3}
Defect density of PEDOT:PSS/Perovskite interface	10^{10} - 10^{20} cm^{-3}	10^{13} cm^{-3}
Defect density of Perovskite/PCBM interface	10^{10} - 10^{20} cm^{-3}	10^{13} cm^{-3}
Series Resistance	10-100 $\Omega\cdot\text{cm}^2$	10 $\Omega\cdot\text{cm}^2$
Shunt Resistance	500-5000 $\Omega\cdot\text{cm}^2$	5000 $\Omega\cdot\text{cm}^2$
Operating temperature of the solar cell	300-390 K	300 K

Table 2 Optimal values obtained from SCAPS-1D
Source: Own Elaboration

Simulation of the perovskite solar cell: ITO/PEDOT: PSS/CH₃NH₃PbI₃/PCBM/Au produced output values of Voc = 1.112 V, Jsc = 20.236 mA/cm², FF = 81.24 % and PCE = 18.33 %, similar to those reported experimentally, according to Heo *et al.*, (2015).

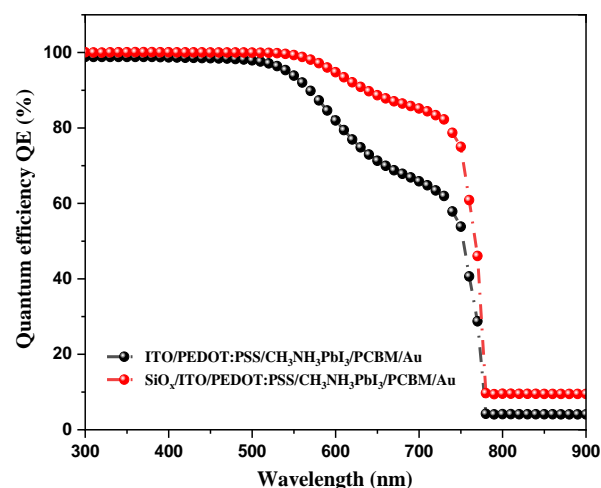
Notwithstanding, the proposed perovskite solar cell: SiO_x/ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Au, containing a SiO_x monolayer film, produced output values of Voc = 1.088 V, Jsc = 27.236 mA/cm², FF = 80.06 % and PCE = 23.74 %. Graph 4 shows the J-V curves with the output values of both devices.



Graph 4 J-V curve of perovskite solar cell with and without SiO_x

Source: Own Elaboration, OriginPro 2018

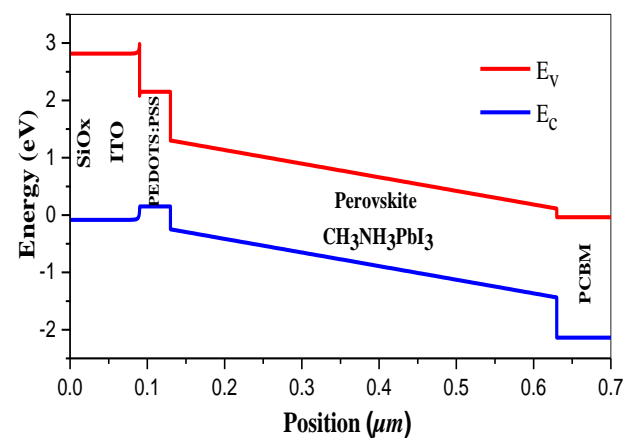
Graph 5 shows the QE quantum efficiency for both devices. An improvement in the QE quantum efficiency is observed for the solar cell that includes the SiO_x film compared to the one that does not. The range of enhancement is observed mainly in the wavelength range from 500 to 780 nm. This increase determines that the structure with the SiO_x film has a better spectral response.



Graph 5 QE Quantum efficiency of perovskite solar cell with and without SiO_x

Source: Own Elaboration, OriginPro 2018

Graph 6 shows the band diagram of the perovskite solar cell with the optimized parameters and the SiO_x down-conversion material. It is observed that there is a splitting of the valence band and conduction band. In addition, the energy band diagram indicates that both the PEDOTS: PSS material and the PCBM material have higher energy bandwidths compared to the absorber layer (CH₃NH₃PbI₃), so there is a band alignment, favoring the passage of electrons and holes generated in the perovskite. Meanwhile, the energy barriers at the interfaces ensure the diffusion of carriers generated in the perovskite into the hole and electron transport layers. Finally, the transparent SiO_x material filters visible light due to its wide band gap and, at the same time, absorbs ultraviolet light, which contributes to increasing the photocurrent.



Graph 6 Energy band diagram of perovskite solar cell with SiO_x

Source: Own Elaboration, OriginPro 2018 and SCAPS-1D

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Conclusions

A p-i-n type inverted perovskite solar cell: ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Au has been simulated and optimized in SCAPS-1D software.

The optimal values obtained from the simulation indicate that a perovskite layer thickness of 500 nm, a total defect density at the perovskite and interfaces of 10¹³ cm⁻³, a series and shunt resistance of 10 Ω.cm² and 5000 Ω.cm² respectively, and an operating temperature of 300 K improve the performance of the solar cell.

In addition, the experimentally obtained silicon-rich oxide (SiOx) material has the main characteristics of an energy down-conversion material for application in perovskite solar cells. In conclusion, the solar device: SiOx/ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Au has the potential to generate a higher photocurrent, as the simulation results in an improved efficiency of 23.74 %, while the solar device without the SiOx film: ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Au produces an efficiency of 18.33 %, slightly higher than the experimental data, i.e. 18.1 %. The perovskite solar cell with SiOx is considered efficient, as it has a simple architecture in addition to the fact that it can easily obtain experimental verification.

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