Design of high-performance perovskite solar cells adapted to the tandem configuration

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Abstract. Thin film based solar cells offer the added advantage of presenting a low temperature coefficient of power and are usually suggested for hot climates. Perovskite solar cells have reached today a record efficiency of 25.2%. In addition, they demonstrated a great compatibility to be used as the top cell in tandem cells, either with CIGS, CZTS, perovskite or silicon heterojunction (HIT) as the bottom cell. The purpose of this work is to design and analyse the selected planar perovskite solar cells (PSCs) reaching high power conversion efficiency and stability with layers configuration of glass substrate / FTO/ TiO2/ MAPbI(3-x)Clx/ CuSCN/ Au, using the SCAPS-1D software. The thickness of MAPbI(3-x)Clx and FTO was varied from 0.4 μm to 1.4 μm and from 0.02 μm to 0.5 μm respectively and the best results are observed at 1.1 μm of perovskite absorber and 0.05 FTO μm. The best performances of this device architecture, calculated with SCAPS software are cell efficiency of 27.79% with an open circuit of 1.2 V, a short circuit current of 26.04 mA/cm² and a fill factor of 88.85%.

1. Introduction
Perovskite solar cells are cells that can compete today with silicon cells. They have shown rapid growth since 2009 and they reach a record efficiency of 25.2%, which is close to the record efficiency of the best silicon heterojunction cell, which is on the order of 28.0% [1].

The perovskite materials have a wide range of properties and characteristics that are attractive for solar cells, namely: a direct gap that can easily be tuned between 1.30 and 2.5 eV, a low exciton binding energy (<10 meV), a large absorption coefficient (on the order of 1.5×10⁴ cm⁻¹ at 550 nm), a high carrier lifetime compared with silicon, an ambipolar conductivity and finally a good mobility of the carriers compared with the organic materials (7.5 cm² / Vs for the holes and 12.5 cm² / Vs for the electrons) [2]. Furthermore, the fabrication of this class of cells requires less sophisticated methods compared to the processes of conventional silicon cells.

A wide range of materials can be used for the different layers constituting the perovskite solar cells. This possibility offers the opportunity to get various performances that can be adapted to different applications.
The standard perovskite planar solar cell is a p-i-n type structure as showed in figure 1. The cell is composed of three main layers namely [2]: the hybrid perovskite as light absorber, an n-type semiconductor as an electron transport layer and a hole carrier layer which is a p-type semiconductor.

Perovskites refer to a family of materials with a very particular crystalline structure, of the form ABX$_3$. Hybrid perovskites are a subfamily of the ABX$_3$ perovskites and result from the combination of the organic cations A of type (R-NH$_3$), a divalent metal B of group 14, smaller than A and a halide X or combination of halides, forming a three-dimensional inorganic structure that contains the organic cation [3,4].

The current challenge is not only to improve the efficiency of the perovskite solar cell, but also to enhance its stability which depends on several factors: climatic conditions (temperature, humidity, irradiation), the nature of the perovskite absorber, the hole transport layer, the electron transport layer and the back contact [5].

The choice of the materials, the structure of the cells and the encapsulant is a good way to enhance the stability of the perovskite device. So far, the best single junction solar cell containing mixed cations and halides, reached an efficiency of 21.1% and showed a good stability of up to 1000 hours at 85°C and 85% relative humidity [6].

In this paper, we report the results of a simulation work carried out by SCAPS Software, of a of perovskite solar cell with planar structure.

2. Device modelling

2.1. SCAPS program

SCAPS (Solar Cell Capacitance Simulator) is a simulation program with seven semiconductor input layers developed by a group of solar cell researcher at the department of Electronics and Information System, University of Gent, Belgium. SCAPS uses the basic semiconductor equations such as Poisson equation, continuity equation of electrons and holes.

SCAPS calculates various profiles of the defined architectures such as steady state band diagram, carrier transport, and recombination and bulk defects.

The working procedure of the SCAPS program is shown schematically in the figure 2 below:
2.2. Device architecture

The choice of materials and structure is a crucial step in the design and optimization of PSCs and it is primordial to achieving high performances and approved stability. The device architecture is presented in the figure 3: First, we started with planar standard structure, with more stable and performant layers. The selection of the materials used in the simulation studies are based on the performances of these materials as shown in the literature.

Indeed, the model of the device was built on transparent fluorine-doped tin-oxide (FTO) coated glass substrates, followed by a TiO$_2$ compact layer is a well-known inorganic material that is used as an us (Electron Transporter layer ETL) in PSCs. This choice is based on the stat-of-the-art and the previous experiences that demonstrated efficiencies up to 22.1% of the PSCs [6, 7, 8, 4].

The mixed-halide perovskite MAPbI$_{3-x}$Cl$_x$ resulting from the substitution of Iodine in the well-known MAPbI$_3$ hybrid perovskite by the chlorine was used as the absorbing layer for several reasons. First, Lead-based perovskite are more stable than those based on tin, due to the instability of the later [2]. In addition, the substitution improves the proprieties of the hybrid perovskite absorber. Indeed, diffusion lengths for holes and for electrons increase from 100 nm for MAPbI$_3$ to more than 1μm in MAPbI$_{3-x}$Cl$_x$ [4,2]. In addition, a charge carrier mobility of up-to 33 cm$^2$V$^{-1}$S$^{-1}$ [9] was obtained in vapor-deposited MAPbI$_{3-x}$Cl$_x$ perovskite with a high gap of 1.55 eV making this material suitable as an absorber for the top cell in silicon-based the tandem solar cell configuration.
Copper thiocyanate (CuSCN) is one of the inorganic p-type semiconductors that is considered to be a good alternative HTM due to its high conductivity ($10^{-2}$–$10^{-3}$ S cm$^{-1}$), stability, and transmittance; its wide band-gap (3.6 eV) and its work function of approximately $-5.3$ eV [10]. The first remarkable work on CuSCN HTM in PSCs was reported in 2014 by Grätzel et al., where a PSC with a 12.4% efficiency was presented, compared to that of HTM-free cell displaying only 9.5% efficiency. This was mostly attributed to a significant increase in the short-circuit current and open-circuit voltage. This improvement was due to the efficient charge extraction and collection from the excited perovskite (MAPbI$_3$ in this case) to TiO$_2$ and CuSCN and to the corresponding electrodes [11].

2.3. Device simulation parameters

The main parameters that must be defined to perform the simulation with SCAPS program are: band gap (Eg), electron affinity ($\chi$), dielectric constant ($\varepsilon$), valence and conduction band density of states (NV and NC), charge carriers mobility ($\mu_n$ and $\mu_p$), acceptor and donor dopant concentration (NA, ND) and thermal velocity of charge carriers ($V_{thn}$ and $V_{thp}$).

The simulation parameters for each layer in perovskite solar cell are selected from previous research [12-15]. Table 1 summarizes the device parameters used in this study.

Table 1: Simulation parameters of the perovskite solar cell.

| Parameters                        | FTO     | TiO$_2$   | CuSCN   | MAPbI$_{3-x}$Cl$_x$ |
|-----------------------------------|---------|-----------|---------|---------------------|
| Band gap Eg (eV)                  | 3.5     | 3.2       | 3.6     | 1.55                |
| Electron affinity (eV)            | 4       | 4.2       | 1.7     | 3.9                 |
| Dielectric permittivity           | 9       | 9         | 10      | 6.5                 |
| CB effective density of states 1/cm$^3$ | $2.2 \times 10^{17}$ | $2.2 \times 10^{18}$ | $2.2 \times 10^{19}$ | $2.2 \times 10^{18}$ |
| VB effective density of states 1/cm$^3$ | $2.2 \times 10^{16}$ | $1.8 \times 10^{19}$ | $1.8 \times 10^{18}$ | $1.8 \times 10^{19}$ |
| Electron thermal velocity (cm/s)  | $1 \times 10^7$ | $1 \times 10^7$ | $1 \times 10^7$ | $1 \times 10^7$ |
| Hole thermal velocity (cm/s)      | $1 \times 10^7$ | $1 \times 10^7$ | $1 \times 10^7$ | $1 \times 10^7$ |
| Electron mobility (cm$^2$/Vs)     | 20      | 20        | 100     | 2                   |
| Hole mobility (cm$^2$/Vs)         | 10      | 10        | 25      | 2                   |
| Shallow uniform donor density ND (1/cm$^3$) | $1 \times 10^{15}$ | $1 \times 10^{16}$ | 0       | $1 \times 10^{13}$ |
| Shallow uniform acceptor density NA (1/cm$^3$) | 0       | 0         | $1 \times 10^{18}$ | $1 \times 10^{13}$ |

3. Results and discussion

3.1. Effect of the thickness of the perovskite

The thickness of the perovskite absorber is one of the parameters that affect significantly the performances of the PSC. The effect of thickness of the absorption layer on the solar cell performance is shown in figure 4. Indeed, when the thickness of the hybrid perovskite increases from 0.4 µm to 1.4 µm, the efficiency improves gradually and reaches its maximum when the perovskite thickness is 1.1 µm. Beyond this value, the performance of the cell decreases, which can be explained by the recombination of the photogenerated carriers before reaching the collecting layers.
3.2. Effect of the thickness of FTO

The thickness of the FTO layer was also varied from 0.02 µm to 0.5 µm. As shown in the figure 5 and 6 the best result was attained with 0.05 µm. However higher values of the FTO thickness lead to a significant decrease in the efficiency and the short-circuit current density, which can be explained by the parasitic absorption of the FTO layer causing a current loss of the cell.

Figure 4: The effect of the perovskite thickness on the performance of the solar cell.

Figure 5: Variation of the cell efficiency with the variation of FTO thickness.
Figure 6: variation of the current density with the variation of FTO thickness.

3.3. Effect of HTM

The effect of the HTM was also investigated by changing the HTM and keeping the other parameter of the devices are the same. The first device was with the most used HTM; spiro-Ometad and the second with inorganic CuSCN material. The results, shown in figure 7 below, demonstrate that the use of CuSCN allows higher performances (efficiency, current density, open circuit voltage, fill factor). This is attributed to the specific properties of this material.

Figure 7: variation of the HTM on the performance of the solar cell.

3.4. The optimized PCS

Based on the previous simulation results, an optimized perovskite solar cell was obtained with the configuration: glass substrate / FTO/ TiO$_2$/ MAPbI$_3$-xCl$_x$/CuSCN/Au with perovskite thickness of 1.1 µm and 0.05 µm for the FTO layer. As shown in the figure 8, we obtained for this optimized configuration an efficiency of 27.79% is, short circuit current density ($J_{sc}$) 26.12 mA/cm$^2$, open circuit voltage ($V_{oc}$) 1.19V, fill factor (FF) 88.79%.
4. Conclusions

The selection of materials plays a primordial role for the design of the perovskite solar cell. The thickness of these layers was investigated to enhance the performances of the perovskite solar cells. The best results were obtained for a perovskite absorber with double halide having a band gap of 1.55 eV and 3.9 eV electron affinity. In addition, the use of an inorganic ETL will renders the final devices more stable. The SCAPS-1D simulations resulted in the optimized perovskite solar cell with the configuration glass substrate / FTO/ TiO₂/ MAPbI₃xClₓ/ CuSCN/Au and with an efficiency of 27.79%. This finding may provide the starting point for an experimental investigation of the cell.

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Figure 8: Simulated J-V of the optimized PSC.
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