On the Investigation of Interface Defects of Solar Cells: Lead-Based vs Lead-Free Perovskite

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ABSTRACT

Perovskite solar cells (PSCs) have drawn significant consideration as a competing solar cell technology because of the drastic advance in their power conversion efficiency (PCE) over the last two decades. The interfaces between the electron transport layer (ETL) and the absorber layer and between the absorber layer and the hole transport layer (HTL) have a major impact on the performance of the PSCs. In this paper, we have investigated the defect interfaces between ETL/absorber layer and absorber layer/HTL of calibrated experimental CH$_3$NH$_3$PbX$_3$ lead-based and FASnI$_3$ lead-free PSCs. The influence of the defect interfaces is studied in order to find the optimum value for the maximum possible PCE. While the PCE has not been enhanced considerably for the lead-based, it is boosted from 1.76% to 5.35% for lead-free PSCs. Also, bulk traps were found to have minor role in comparison with interface traps for the lead-free cell while they have a significant impact for the lead-based cell. The results presented in this work would shed some light on designing interface defects of various types of practical PSC structures and demonstrates the crucial impact of the interface defects on lead-free vs lead-based PSCs. All simulation studies are performed by using SCAPS-1D simulator.

INDEX TERMS

Interface defects, lead based perovskite solar cell, lead free perovskite solar cell, SCAPS-1D.

I. INTRODUCTION

Recently, an urgent demand for the substitution of fossil fuels by clean energy sources is universally considered. Solar energy is one of the most best replacement renewable energy resources [1]. Silicon solar cells demonstrate about 90% of the solar cells market [2], [3]. Silicon solar cells offer high PCEs above 25%; however, these cells are suffering from the relatively high cost of fabrication. Consequently, efficient, and low-cost solar cells (SCs) are required [2]. In this regard, perovskite solar cells (PSCs) have been introduced because of their high PCE and low-cost fabrication methods [4]. PSCs have an exceptionally electronic properties like bandgap tunability, low-cost fabrication techniques, wide absorption spectrum and long diffusion lengths of carriers.

Perovskite materials are materials with ABX$_3$ structure where ABX$_3$ is the structure of calcium titanium oxide (CaTiO$_3$). During the years 2009 to 2016, PSCs fabricated from methylammonium lead halides (CH$_3$NH$_3$PbX$_3$, where X = Iodine (I), Bromine (Br), or Chlorine (Cl)) have a fast improvement in PCE from 3.8% to more than 22% [5]–[7]. Although this high PCE of lead-based PSCs, toxicity of lead has a severe issue. Two methods are used to alleviate toxicity of lead-based PSCs. Firstly, mixing lead with other metals like tin CH$_3$NH$_3$Sn$_x$Pb$_{1-x}$ [8]. Secondly, replace lead with analogous metals. The innocuous Tin is considered the most appropriate material to replace lead because the two materials are from the same group in the periodic table. Tin-based devices suffer from instability and SnF$_2$ is added to
make the system more stable [9]. Formamidinium tin iodide (HC(NH$_3$)$_2$SnI$_3$ = FASnI$_3$) has been used to make the perovskites more solid and stable. FASnI$_3$ has an energy gap ($E_g$) of 1.41 eV and wide spectrum up to 880 nm. Lately, many methodologies have been proposed to enhance the performance of solar cells based on FASnI$_3$. Using SnF$_2$-pyrazine composite or an inverse are achieved PCE of 4.8% and 6.22%, respectively [10], [11]. The open circuit voltage is increased when using cascaded electron transport materials (ETMs) (TiO$_2$–ZnS) [12]. TiO$_2$ and Spiro-OMeTAD are the most famous ETM and hole transport material (HTM), respectively. Other materials like ZnSe, CdS, ZnS, ZnO and ZnOS are used as ETMs and CsSnI$_3$, CuI, Cu$_2$O, CuO, CuSCN and NiO are used as HTMs along with other organic materials [5], [12]–[15].

The interfaces between electron transport layer/perovskite and perovskite/hole transport layer are known as crucial factors for determining the functioning of the perovskite solar cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell. In the normal structure PSCs, electron transport layers provide paths for electron extraction and have a crucial effect on the interface cell which also affect the stability of the cell.

**II. CALIBRATION OF THE LEAD BASED AND LEAD-FREE PEROVSKITE SOLAR CELLS**

Before studying the impact of interface defects on the performance of the CH$_3$NH$_3$PbX$_3$ lead-based solar cell and FASnI$_3$ lead-free PSC, a calibration step is performed. SCAPS-1D is verified with practical lead-based and lead-free PSCs with a design of (Au) as a back contact, ZnO as ETL for the lead-based PSC, TiO$_2$ as ETL for the lead-free PSC, absorber perovskite layer, Spiro-OMeTAD as HTL and Fluorine-doped tin oxide (FTO) as a transparent conducting oxide (TCO) [30], [31]. The calibration is done by taking the given materials parameters from the practical papers and perform the simulations with the same practical parameters [30], [31]. For the other parameters, it is taken from well-known publications as cited in Tables 1 and 2. Finally, comparing the J-V curves and the output performance parameters of the simulated cells with the practical cells to ensure the simulation validity. A temperature of 300 K and air mass (AM 1.5) illumination spectrum have been used for all simulations. The CH$_3$NH$_3$PbX$_3$ absorption coefficient ($\alpha$) is extracted from practical results [32]. A neutral defect type with Gaussian distribution is chosen to achieve practical data [24]. Equation (1) is used for the calculations of the absorption coefficients ($\alpha$) of TCO, ETM, FASnI$_3$ and HTM with a pre-factor ($A_\alpha$) which is selected to be $10^7$ cm$^{-1}$ eV$^{-1/2}$ [33].

$$\alpha(E) = A_\alpha \sqrt{\frac{E}{E_g}}$$  \hspace{1cm} (1)

The main structure and the energy band diagram of the PSC different layers are demonstrated in Fig. 1(a) and Fig. 1(b), respectively. The material parameters of SCAPS-1D are listed in Table 1 while Table 2 shows the various defect properties and their parameters. FASnI$_3$ has a p-type charge carrier density [11]. The bulk defect density in FASnI$_3$ is selected to be $2 \times 10^{15}$ cm$^{-3}$ to get a carrier lifetime of 2.5 ns which match the theoretical range (1 ns- 4 ns) [46], [47]. For the lead-based cell a flat band model is utilized for the interfaces TCO/semiconductor and metal/semiconductor. The work functions of TCO and Au for the lead-free cell are set to 4 and 5.1 eV, respectively. Also, the bulk defect density is adjusted at $7 \times 10^{13}$ cm$^{-3}$ to give the best fit with experimental results. Further, the series resistance is estimated to be 5.4 $\Omega$.cm$^2$ [26]. Fig. 2 demonstrates the J-V curves of the practical PSCs [30], [31] against the simulated cells of the lead-based PSC (Fig. 2(a)) and the lead-free PSC (Fig. 2(b)). The performance parameters of the simulated cells and the practical work are listed in Table 3 showing good accuracy with absolute errors between measurements and simulations less than 5%.
III. NUMERICAL TECHNIQUE USED IN SCAPS-1D

Simulation tools are valuable methods for design, studying and expecting the efficiency of SCs. Moreover, simulation tools can help optimize various technological and geometric parameters of solar cells. SCAPS-1D is a useful program to design and study various types of SCs which has been widely verified vs experimental SCs. It can simulate SCs with up to seven layers. Most materials parameters can be assigned such as thickness, energy gap, affinity, permittivity, mobility, and concentration of doping and other physical or geometrical parameters [48]–[50].

| Parameters  | FTO | ZnO | TiO₂ | MAPbI₃ | FASnI₃ | Spiro-OmeTAD |
|-------------|-----|-----|------|--------|--------|-------------|
| Thickness   | 500 | 500 | 30   | 400    | 350    | 200         |
| Eₚ (eV)     | 3.5 | 3.3[49] | 2.2[49] | 1.5[49] | 1.4[49] | 2.0[49] |
| Electron affinity (eV) | 4 | 6[48] | 6[48] | 5.0[48] | 3.52[49] | 2.45[49] |
| Relative permittivity | 9 | 9[50] | 9[50] | 18[50] | 6.2[50] | 3[50] |
| Effective conduction band density Nₑ (cm⁻³) | 2.2×10¹⁸ | 2.2×10¹⁸ | 2×10¹⁸ | 2.2×10¹⁸ | 10¹⁸ | 2.2×10¹⁸ |
| Effective valence band density Nᵥ (cm⁻³) | 1.8×10¹⁸ | 1.9×10¹⁸ | 1.8×10¹⁸ | 1.9×10¹⁸ | 10¹⁸ | 1.9×10¹⁸ |
| Thermal velocity of electrons vₑₑ (cm/s) | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] |
| Thermal velocity of holes vᵥᵥ (cm/s) | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] | 10¹⁰[49] |
| Electron mobility µₑ (cm²/V·s⁻¹) | 20 | 10⁹[50] | 20⁹[50] | 5⁹[50] | 2⁹[50] | 2⁹[50] |
| Hole mobility µᵥ (cm²/V·s⁻¹) | 10 | 2⁹[50] | 10⁹[50] | 2⁹[50] | 2⁹[50] | 2⁹[50] |
| Donor concentration Nᵥ (cm⁻³) | 0 | 0 | 0 | 0 | 7×10¹⁸ | 10¹⁷[51, 59] |
| Acceptor concentration Nₑ (cm⁻³) | 2×10¹⁸ | 10¹⁸ | 9×10¹⁸ | 0 | 0 | 0 |

The numerical solution in SCAPS-1D solves the basic physical differential equations in steady state. The main equations are the continuity equations (Eq. 2 and 3) and Poisson equation (Eq. 4) which are solved self consistently until
convergence occurs.

\[
\frac{1}{q} \frac{dJ_n}{dx} = U_n - G(x) 
\] (2)

\[
\frac{1}{q} \frac{dJ_p}{dx} = G(x) - U_p 
\] (3)

\[
\frac{d^2\psi}{dx^2} = \frac{q}{\varepsilon} (n - p + N_A^- - N_D^+ + n_i - p_i) 
\] (4)

where \( G(x) \) is carrier generation rate, generated from the sun, at a distance \( x \) below the surface which is given by \([51],[52]\),

\[
G(x) = \int_0^\infty G(\lambda, x) \, d\lambda 
\]

Moreover, the recombination through deep traps in \( E_g \) is modeled by the Shockley Read Hall (SRH) model \([53],[54]\). The SRH recombination for bulk and interface defects is defined by bulk minority carrier lifetimes (\( \tau_n \) and \( \tau_p \)) and interface recombination velocities (\( S_n \) and \( S_p \)), respectively, utilizing the following equations,

\[
R_{RSH,bulk} = \frac{(np - n_i^2)}{\tau_p (n + n_i) + \tau_n (p + p_i)} 
\] (6)

\[
R_{RSH,interface} = \frac{n_i \left( \frac{p_i}{S_n} + \frac{n_i}{S_p} \right)}{\frac{n_i \left( \frac{p_i}{S_n} + \frac{n_i}{S_p} \right)}{\frac{1}{\tau_n} \frac{1}{\tau_p}} 
\] (7)

\[
n_1 = n_i \exp \left( \frac{E_i - E_T}{K_BT} \right) 
\] (8)

\[
p_1 = n_i \exp \left( \frac{E_T - E_i}{K_BT} \right) 
\] (9)

\[
\tau_n = \frac{1}{\sigma_n N_B v_{th}} 
\] (10)

\[
\tau_p = \frac{1}{\sigma_p N_B v_{th}} 
\] (11)

\[
S_n = \sigma_n N_B v_{th} 
\] (12)

\[
S_p = \sigma_p N_B v_{th} 
\] (13)

The symbols used in the aforementioned equations along with their definitions are summarized in Table 4.

Fig. 3 illustrates a flow chart of the numerical solution of SCAPS-1D. Firstly, the cell geometry is inserted. Then, the materials parameters are defined. Poisson equation and continuity equations are discretized into different mesh points. Discretization is executed by employing the finite difference method (FEM), and the current densities are modeled using the Scharfetter–Gummel process \([55]\). In the assigned mesh points, \( \psi, n \) and \( p \) are iteratively computed by the Gummel method \([56]\). Physical models such as \( G, U, \mu_n \) and \( \mu_p \) are enhanced after each iteration. Finally, we can get \( J-V, C-V, C-F, QE, AC, G, U \) and energy diagrams \([57]\).

In should be emphasized here that, in SCAPS-1D, an interface can be studied between any two layers. The model used for this implementation is thermionic emission. The thermal velocity in an interface is equal to the least thermal velocity of the two layers. The interface defects could be treated like bulk defects. The recombination in interface states is evaluated by the Pauwels-Vanhoutte theory \([58]\), which is an extension of the SRH theory.

The traps act as recombination centers lead to losses in the photocurrent and an increase in the dark current. Both effects contribute to the decrease in the photo conversion efficiency. The key physical parameters that ascertain how a
specific trap affects the nonradiative recombination rate are the trap density, energy level, and electron and hole capture cross sections. Regarding the density \( N_t \), its value can be technologically controlled by reducing the crystallographic defects as grain boundaries and surface defects at the interfaces throughout the fabrication processes [28].

The electron and hole capture cross sections depend mainly on the static dielectric constant of the material [59]. Also, they depend on the physical and chemical uniqueness of the trap and more prominently on the trap charge state. If the dielectric constant is high, the screening is strong, and it is less likely that a charge becomes captured. In [60], it was shown the liquid-like molecular reorientation motions in the perovskite allow the effective carrier screening. In addition, in [61], the potassium halide species at the grain boundaries was observed to lower the capture cross section.

It is reported that neutral vacancy pairs and elementary vacancy defects do not create deep energy states inside the perovskite bandgap. Only shallow traps are most likely generated which are not anticipated to act as powerful recombination centers [62]. Further, some impurities in the PbI\(_2\) precursor and traces of Au in the perovskite layer, resulting from electrode diffusion, have been reported to decrease the cell performance [63]. The trap energy levels could also be affected by whether the perovskite layer is p-type or n-type [64]. So, to avoid such impurities which may produce deep traps, careful manufacturing processes and tuning the film composition and morphology are required.

In the following sections, we will study the impact of each interface parameters to determine the most important parameters that can be tuned to get the best performance. Also, the impact of bulk traps on the performance will be investigated to give a complete picture of the traps effect and to demonstrate the dominant traps the have the most significant role in controlling the efficiency of the lead-based and lead-free PSCs.

### IV. OPTIMIZATION OF ETL/PSK AND PSK/HTL DEFECT INTERFACES OF THE LEAD BASED CELL

#### A. OPTIMIZATION OF ETL/PSK DEFECT INTERFACE

In this subsection, the influence of defect density \( N_t \) between ETL and perovskite interface is studied. It has a substantial impact on the functioning of the solar cell as the quality of the interface ETL/absorber layer has shown a pronounced influence on the performance of the PSC [65]. Consequently, the recombination rate of carriers increases which alters \( V_{oc} \) of the PSC. Fig. 4(a) illustrates the efficiency vs interface defect density where the computation is based on the Fermi level \( E_{fn} \). The performance is improved with reducing \( N_t \) with no considerable impact below \( N_t \) of \( 1 \times 10^{11} \) cm\(^{-2} \). As lowering \( N_t \) means increasing the cost of fabrication, a value of \( N_t \) of \( 1 \times 10^{11} \) cm\(^{-2} \) could be chosen as a design parameter which gives \( PCE = 16.53\% \).

In the next simulations, two distinct values of \( N_t \) are used: a low concentration value of \( 10^{11} \) cm\(^{-2} \) and a high concentration value of \( 10^{14} \) cm\(^{-2} \). The variation of the defect position \( (E_t) \) with respect to the highest \( E_{V} \) is inspected. It can be inferred from Fig. 4(b) that the change of \( E_t \) does not affect the PCE for the low value of \( N_t \). On the other hand, for the high concentration value of \( N_t \), the PCE starts with a high value of about 16% when \( E_t \) coincides with the valence band edge, \( E_v \), then it has a lower constant value of 15.41% up to the defect position, \( E_t \) of 1.3 eV. The PCE is then increased reaching about 16% for \( E_t \) matched with the conduction band edge, \( E_c \). There are two types of defects; the shallow defects with position of higher and lower than the mid of the \( E_g \) of PSK and nearer to \( E_c \) and \( E_v \), respectively which have low destructive impact on performance in comparison to deep defects which are near the mid of \( E_g \) [66].

In addition, the PCE variation vs ETL/PSK cross-section area of electrons \( (\sigma_n) \) is investigated for the same two levels of \( N_t \). As discussed herein, the capture cross-section signifies the likelihood of the trap capturing the free charge carrier. When light is irradiated to the PSC, carriers are generated in the absorber layer [36]. The electrons will be in the direction from the absorber layer to ETL and the holes will be in the direction from the absorber layer to HTL. Consequently, in ETL/PSK interface the cross-section area of electrons has no significant effect upon the performance parameters and the cross-section area of holes has a significant impact. However, in the PSK/HTL interface the cross-section area of electrons has significant influence on the performance parameters and the cross-section area of holes has no significant impact.

**FIGURE 4.** Variation in efficiency according to the variation of (a) \( N_t \) of the ETL/Perovskite interface, (b) \( E_t-E_{V\text{max}} \), (c) \( \sigma_n \) and (d) \( \sigma_p \).
So, the PCE dependence on $\sigma_n$ is the same as the previous calculations. It can be inferred from Fig. 4(c) that the change of $\sigma_n$ does not affect the PCE for the low value of $N_t$. But for the large value of $N_t$, the PCE is decreasing with increasing the capture cross section, $\sigma_n$. The PCE is about 15.45% for $\sigma_n$ greater than $1 \times 10^{-22}$ cm$^2$. For $N_t$ of $10^{11}$ cm$^{-2}$, $\sigma_n$ of $1 \times 10^{-15}$ cm$^2$ is chosen as a practical value.

Furthermore, the effect of ETL/PSK cross-section area of holes ($\sigma_p$) is investigated. Fig. 4(d) illustrates the efficiency vs $\sigma_p$. Increasing the cross-section has a significant impact on the performance. For $N_t$ of $10^{11}$ cm$^{-2}$, the PCE is decreasing for $\sigma_p$ greater than $1 \times 10^{-14}$ cm$^2$ but for $N_t$ of $10^{14}$ cm$^{-2}$, the PCE is decreasing for $\sigma_p$ greater than $1 \times 10^{-18}$ cm$^2$. So, $\sigma_p$ of $1 \times 10^{-16}$ cm$^2$ may be chosen.

**B. OPTIMIZATION OF PSK/HTL DEFECT INTERFACE**

Here, the defect density of the absorber/HTL interface is investigated. Fig. 5(a) illustrates the PCE variation vs interface defect density where the calculation is based on the Fermi level ($E_F$). The efficiency is improved with lowering $N_t$ with no major impact below $N_t$ of $1 \times 10^{11}$ cm$^{-2}$. $N_t$ of $1 \times 10^{11}$ cm$^{-2}$ is chosen and, in this case, PCE = 16.53%.

Next, two values of $N_t$ are treated, $10^{11}$ cm$^{-2}$ and $10^{14}$ cm$^{-2}$. For the low concentration value of $N_t$, the energy $E_t$ with respect to the highest $E_V$ and $\sigma_p$ of absorber/HTL interface does not have a significant impact on the performance as can be depicted in Fig. 5(b) and Fig. 5(d), respectively, while an efficiency degradation is observed when PSK/HTL cross-section area of electrons ($\sigma_n$) is increased as can be depicted in Fig. 5(b). Meanwhile, for the other high concentration value of $N_t$, the trend of the impact of trap energy position, illustrated in Fig. 5(b), is the same as that of the ETL/Perovskite interface which indicates the crucial role of designing shallow acceptor traps (near $E_F$) to enhance the cell performance for any trap density value. Furthermore, the efficiency degradation is observed when PSK/HTL cross-section area of electrons and holes are increased as can be depicted in Fig. 5(c) and Fig. 5(d), respectively. Values of $\sigma_n = 1 \times 10^{-15}$ cm$^2$ and $\sigma_p = 1 \times 10^{-15}$ cm$^2$ could be chosen to prevent this degradation.

It can be concluded here that the most vital factor that affects the performance of the cell is the trap density. If the trap density is low, the impact of $E_t$ position is neglected while there is a minor influence on the efficiency when concerning $\sigma_p$ for the ETL/Perovskite interface and $\sigma_n$ for the HTL/Perovskite interface. This is due to the rise of the electron and hole surface recombination velocity according to increasing $\sigma_n$ or $\sigma_p$ as indicated in Eq. (12) and Eq. (13). As $N_t$ increases, the impact of $N_t$ on the efficiency is more noticeable and one can see the importance of refining the design parameters in order to reach an optimum cell performance.

**V. OPTIMIZATION OF ETL/PSK AND PSK/HTL INTERFACE DEFECTS OF THE LEAD-FREE CELL**

In this section, we study the influence of the defect interfaces between ETL/PSK and PSK/HTL of the lead-free PSC. The sequence of the optimization is carried out in the same order as in the case of lead-based PSC.

**A. OPTIMIZATION OF ETL/PSK INTERFACE DEFECT**

The variation of the interface defect density of the interface ETL/absorber layer is investigated in Fig. 6(a) which illustrates the impact of the interface defect density on PCE. The performance is improved with decreasing $N_t$ with no lower limit according to the simulation. This finding is extremely important as the trend is not the same as the lead-based cells. In lead-based PSC, there is a lower limit for the trap density below which there is no significant improvement in the efficiency. On the other hand, in lead-free PSC, decreasing trap density gives a linear increase in the efficiency which is beneficial in solar cell design. But as lowering $N_t$ means increasing the cost of fabrication, a suitable value $N_t$ of around $4 \times 10^{11}$ cm$^{-2}$ could be chosen in our design which gives PCE = 4.5%.

Two values of $N_t$ are used in the investigation of the influence of trap parameters on the efficiency. A low concentration value of $4 \times 10^{11}$ cm$^{-3}$ and a high concentration value of $10^{14}$ cm$^{-3}$ are considered in the following simulations. The effect of ETL/PSK trap energy position ($E_t$) with respect to the highest $E_V$ is firstly studied as shown in Fig. 6(b). For low value of $N_t$, the change of $E_t$ does not affect the PCE substantially. When $N_t$ is $10^{14}$ cm$^{-3}$, the PCE is degraded if the trap position is near the mid-gap. So, for both cases of trap density, $E_t$ is chosen to be a shallow trap in order to boost
the performance. In addition, the impact of ETL/PSK $\sigma_n$ and $\sigma_p$ is examined and the results of the corresponding PCE is shown in Fig. 6(c) and Fig. 6(d), respectively. It is noticed the performance is degraded for low values of $\sigma_n$ and $\sigma_p$ for both values of $N_t$. So, $\sigma_n$ of $1 \times 10^{-16}$ cm$^2$ is chosen. Also, $\sigma_p = 5 \times 10^{-16}$ cm$^2$ is preferable for both trap concentrations.

### B. OPTIMIZATION OF PSK/HTL DEFECT INTERFACE

The variation of the defect density of the absorber/HTL interface is explored as illustrated in Fig. 7(a). The performance is improved with decreasing $N_t$ which is the same trend as for the absorber/ETL interface Two different values of $N_t$ are considered in the next simulations, $1 \times 10^{14}$ cm$^{-3}$ and $10^{16}$ cm$^{-3}$. Fig. 7(b) illustrates the performance variation vs PSK/HTL trap energy position ($E_t$) with respect to the highest $E_V$ (which is that of the PSK). It can be inferred from the figure that the change of $E_t$ does not affect the PCE for any $N_t$ value; so, an $E_t$ position which coincides with $E_c$ or $E_v$ is chosen. Further, Fig. 7(c) and Fig. 7(d) illustrate the dependence of PCE on PSK/HTL $\sigma_n$ and $\sigma_p$ for the two values of $N_t$. the dependence is weak as can be inferred from the figures. $\sigma_n$ of $1 \times 10^{-15}$ cm$^2$ and $\sigma_p$ of $1 \times 10^{-15}$ cm$^2$ are chosen as design parameters.

Also, it can be concluded here, for the lead-free cell, that the most critical factor that influences the cell efficiency is the trap density. If the trap density is low, the impact of $E_t$ on the efficiency is noticeable only for the ETL/Perovskite interface while it has a minor effect for the HTL/Perovskite interface. Moreover, the impact of $\sigma_n$ and $\sigma_p$ on the efficiency is more pronounced as compared to the lead-based cell. This is true for both types of solar cells.

### VI. PHYSICAL INTERPRETATION OF THE RESULTS BEFORE AND AFTER OPTIMIZATION

One of the most important differences between the lead-based and lead-free PSCs is the impact of the interface trap density. Regarding the lead-based cell, decreasing $N_t$ beyond a certain value is not beneficial as there is a saturation in the efficiency. However, the efficiency is boosted linearly when decreasing $N_t$ concerning the lead-free cell. Further, shallow defects with a position lower than the mid of $E_g$ of PSK have low destructive effect on performance, while deep defects which are near the mid of $E_g$ have strong influence [39]. This is true for both types of solar cells.

As mentioned above in the lead-based cell, in ETL/PSK interface, for low values of $N_t$, the cross-section area of electrons has no significant effect on the efficiency while the cross-section area of holes has a significant impact. On the other hand, in PSK/HTL interface the cross-section area of electrons has a more substantial impact on the performance than that of the effect of cross-section area of holes. Also, both cross-section areas of electrons and holes in ETL/PSK and
PSK/HTL defect interfaces of the lead-free cell have a little influence on the performance parameters. This difference is according to the distinction of the structure of the cells. In the lead-based, an intrinsic layer of perovskite is sandwiched between ETL and HTL and the structure is p-i-n [67]. But in the lead-free cell the structure is p⁺-n [26].

In this section, we provide a comparison that demonstrates the effectiveness of the optimization processes of the interfaces carried out in this work for both lead-based and lead-free PSCs. Firstly, the JV curves of the initial and optimized PSCs for the lead-based cell and the lead-free one are simulated and the results are shown in Fig. 8(a) and Fig. 8(b), respectively. The performance parameters of the initial design and after optimizing the defect interfaces, for both lead-based and lead-free PSCs, are listed in Table 5 for comparison. The influence of interface parameters on the lead-based is minor and very little improvement is accomplished. However, the design and tuning of the interface parameters significantly affects the performance of the lead-free PSC.

Referring to Table 5, the performance enhancement for the lead-free cell is mainly due to the improvement in the open circuit voltage ($V_{oc}$) which increases from about 0.26 V to about 0.52 V. This doubling of $V_{oc}$ is due to overcoming the recombination mechanism that arises on the interfaces and limit the reverse saturation current of the cell. For a physical justification of the cell behavior corresponding to the interfaces’ optimization, we draw the dark characteristics before and after optimization for the lead-based and lead-free cells as shown in Fig. 10(a) and Fig. 10(b), respectively. Also, the local ideality factor is illustrated. As can be inferred from Fig. 10(a), the lead-based cell has almost the same reverse saturation current for both the initial and final structures. Regarding the lead-free cell, the situation is completely different. The reverse saturation current is declined for the optimized design which indicates a higher open circuit voltage. Also, the local ideality factor is decreased which results in a higher fill factor [68], [69]. The low ideality factor results in less recombination losses which was proven experimentally in the literature [70]. So, this work shows the importance of optimization of interface defects on the performance of lead-free PSCs. Most studies are directed to lead-based cells, but their improvements are limited because of the high efficiency of the initial structure. This study is very important because it shows that inspecting the interface defects in the lead-free cells is crucial to increase the cell efficiency.

### VII. BULK TRAPS VS INTERFACE TRAPS

In the previous sections, the interface properties were investigated thoroughly to optimize and determine the most important parameters that strongly impacts the efficiency of the PSC. In this section, we incorporate the bulk traps into our study to give a complete picture about the influence of various types of defects. This analysis is important to determine the most effective competing defects, bulk vs interface traps. The bulk traps that are highly effective in the PSC are those found inside the absorber as it is the region in which the photoexcited carriers are mainly generated.

|                | $V_{oc}$ (V) | $J_{sc}$ (mA/cm²) | FF (%) | PCE (%) |
|----------------|-------------|--------------------|--------|---------|
| Initial (Lead-based) | 1.114       | 21.174             | 70.03  | 16.53   |
| Optimized (Lead-based) | 1.115       | 21.175             | 70.02  | 16.53   |
| Initial (Lead-free) | 0.2649      | 15.607             | 42.59  | 1.76    |
| Optimized (Lead-free) | 0.5259      | 16.727             | 60.84  | 5.35    |
Figure 10 displays the efficiency as a function of absorber bulk trap density \( N_B \) for both lead-based (Fig. 10(a)) and lead-free (Fig. 10(b)) PSCs. The simulation is carried out for two cases, one with the default values of the interface parameters and the other case is for optimized parameters designed here before. Generally, the trend of the efficiency is that it increases when the bulk trap density decreases as expected. As displayed in Fig. 10(a), when \( N_B \) is lower than \( 10^{11} \) cm\(^{-3} \) in the lead-based cell before optimization of the interface parameters, the efficiency nearly saturates at about 19.25% with any further decrease in \( N_B \). Also, when the interface parameters are optimized, the PCE saturates at a higher value, namely about 21.15%. The results show the crucial role of the bulk traps in lead-based cells. Also, the role of interface traps is dominant for low values of \( N_B \). So, in order to have a good design for the lead-based PSC, both bulk and interface traps should be optimized simultaneously.

Referring to Fig. 10(b), the efficiency trend, according to the variation of bulk trap density, is less pronounced for the lead-free cell. The PCE increases from 1.76% and saturates at slightly higher than 1.8% for \( N_B \) lower than \( 10^{14} \) cm\(^{-3} \). Additionally, for the optimized interface parameters, the PCE increases from 5.35% to 5.55% when \( N_B \) decreases from \( 2 \times 10^{15} \) cm\(^{-3} \) to \( 10^{14} \) cm\(^{-3} \). This little increase in the PCE suggests the minor impact of the bulk traps for the lead-free cell. These results show the importance of optimizing the interface parameters in lead-free cells to enhance the efficiency.

VIII. CONCLUSION

SCAPS-1D is utilized to simulate lead-based PSC with ZnO as ETL and Spiro-OMeTAD as HTL. The lead-based PSC is firstly verified with a practical work. The influence of the interfaces’ defects ETL/absorber and absorber/HTL is investigated. Decreasing \( N_t \) of ETL/absorber and absorber/HTL interfaces from \( 1 \times 10^{11} \) to \( 1 \times 10^{10} \) cm\(^{-2} \) increases the PCE by only 0.02%. If \( N_t \) is increased, the PCE decreases substantially. Although the interfaces’ defects could be optimized, the photovoltaic parameters do not change. In addition, we have calibrated a lead-free PSC with practical measurements. The influence of the interface defects ETL/absorber and absorber/HTL is investigated. Decreasing \( N_t \) of ETL/absorber and absorber/HTL interfaces is found to have a pivotal effect leading to increase the efficiency linearly. This impact is not seen in the lead-based cells as the efficiency is saturated beyond a certain value of the trap density. Regarding the lead-free cell, the interface defects are optimized and the PCE is enhanced from 1.76% to 5.35% at a considerably low interface trap density that is not far from experimental considerations.

Other interface parameters like the electron and hole cross-section area, and trap energy position are investigated. For lower values of \( N_t \), the influence \( \sigma_n \), \( \sigma_p \), and \( E_t \) is minor on the PCE. On the other hand, for the higher values of \( N_t \), the influence is significant, and care must be taken in order to design such factors for better performance. Finally, we investigated the variation of bulk trap density of the absorber to differentiate between its impact on the cell performance and the impact of the interface quality. It was found that the interface influence is much more crucial than the bulk traps specially for the lead-free PSC.

Moreover, the impact of the absorber bulk trap density is investigated. In the lead-based cell, optimizing the interface traps pushes the efficiency for low values of bulk traps. So, both types of traps play important role in controlling the recombination through the PSC. On the other hand, in the case of lead-free cell, the bulk traps have minor influence while the interface quality greatly impacts the performance and is a promising direction to boost the efficiency.

This study successfully addresses the impact of various parameters of interface traps on the performance of PSCs and provides some design guidelines that can help to boost the efficiency of this type of cells. Finally, the presented study sheds the light on two different materials as case studies for the lead-based and lead-free perovskite solar cells. More efforts should be directed to inspect more materials with different bandgaps to optimize their performance under the influence of interface traps.

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