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Computational Study of Highly Efficient SnO_2 ETL-based Inorganic Perovskite Solar Cell

***Abstract**. The growing demand for high Power Conversion Efficiency (PCE) in Perovskite Solar Cells (PSCs) has increased the need for the introduction of new material combinations. The conventional TiO_2 Electron Transport Layer (ETL) has reached its maturity limit, and no further progress can be made. Therefore, the replacement with the SnO_2 layer is seen as very reasonable. Furthermore, the crystallization of TiO_2 requires a high-temperature annealing process, thus obliterating its use in flexible applications, and increasing fabrication costs. This study numerically elaborated on the potential of SnO_2 as an ETL in PSCs using the Solar Cell Capacitance Simulator (SCAPS-1D). The combination of SnO_2 ETL with solid-state CUSCN HTL is also deliberated to provide an alternative to the use of all inorganic PSCs, where the cell consists of ITO/ SnO_2 /$CH_3NH_3PbI_3$/CuSCN/Au. Numerous key parameters of SnO_2 have influenced cell performance, including operating temperature, layer thickness, dopant density and defect density state. The highest PCE has been recorded reaching up to 24.14% with FF of 85.99%, VOC of 1.15V, and JSC of 24.42 mA/ cm^2 from the optimized cell structure. However, it can be seen that the impact of high defect density has had a profound effect on PCE performance, thus illuminating the comprehensive concern required in containment. This may provide a guideline prior to the future fabrication of PSCs utilizing the SnO_2 as ETL.*

Streszczenie.** Rosnące zapotrzebowanie na wysoką wydajność konwersji energii (PCE) w perowskitowych ogniwach słonecznych (PSC) zwiększyło potrzebę wprowadzenia nowych kombinacji materiałów. Konwencjonalna warstwa transportu elektronów TiO_2 (ETL) osiągnęła swój limit dojrzałości i nie można poczynić dalszych postępów. Dlatego też zastąpienie warstwą SnO_2 wydaje się bardzo rozsądne. Co więcej, krystalizacja TiO2 wymaga procesu wyżarzania w wysokiej temperaturze, co eliminuje jego zastosowanie w elastycznych zastosowaniach i zwiększa koszty produkcji. W tym badaniu opracowano numerycznie potencjał SnO_2 jako ETL w PSC przy użyciu symulatora pojemności ogniw słonecznych (SCAPS-1D). Rozważa się również połączenie SnO_2 ETL z półprzewodnikowym CUSCN HTL, aby zapewnić alternatywę dla stosowania wszystkich nieorganicznych PSC, gdzie ogniwo składa się z ITO/SnO2/CH3NH3PbI3/CuSCN/Au. Na wydajność ogniwa wpływa wiele kluczowych parametrów SnO_2, w tym temperatura robocza, grubość warstwy, gęstość domieszki i stan gęstości defektów. Odnotowano najwyższy współczynnik PCE sięgający 24,14% przy FF wynoszącym 85,99%, VOC wynoszącym 1,15 V i JSC wynoszącym 24,42 mA/cm^2 ze zoptymalizowanej struktury komórkowej. Można jednak zauważyć, że wpływ dużej gęstości defektów miał głęboki wpływ na wydajność PCE, rzucając w ten sposób światło na wszechstronną troskę wymaganą w zakresie powstrzymywania. Może to stanowić wytyczne przed przyszłym wytwarzaniem PSC wykorzystujących SnO_2 jako ETL. (Badanie obliczeniowe wysoce wydajnej nieorganicznej ogniwa słonecznego z perowskitu na bazie SnO2 E**TL)

Keywords: SnO_2, perovskite solar cell, SCAPS-1D, ETL.
Słowa kluczowe: SnO_2, perowskitowe ogniwo słoneczne, SCAPS-1D, ETL.

Introduction

Perovskite Solar Cell (PSC) is an emerging solar cell with excessive progress in the last two decades, making it a promising candidate as a future energy source [1]. Perovskite was discovered in 2009 and named after Lev Perovski, who discovered it as the third generation of solar cell technology. Typically, PSC has three important layers, which are the ETL (SnO_2), the HTL (CuSCN), and the absorber layer (Perovskite). These three layers are sandwiched with each other to produce a solar panel [2], [3].

ETL material is one of the most significant aspects of a solar cell. It plays a crucial role in extracting and transporting photogenerated electron carriers and serves as a hole-blocking layer by suppressing charge recombination [4]. TiO_2 has been widely used as an ETL in PSC. However, it is not very efficient for charge extraction at the interface, especially in planar structures [5], [6]. Meanwhile, other ETL materials such as SiO_2, ZrO_2, SnO_2, and ZnO have been suggested to overcome the limitations of TiO_2. According to the research that has been conducted, SnO_2 has stood out the most compared to the others. It has excellent chemical stability, U-V resistance, high charge extraction, and greater band alignment than other ETLs [7], [8].

SnO_2 material is chosen as the ETL in this project because of its favourable advantages. A wide bandgap that SnO_2 has is higher than other ETL materials, allowing it to have a superior light transmittance of 90% in glass [9], [10]. It enables SnO_2 to absorb less UV light while offering better device stability. Other than that, it has up to 240 cm^2 $(Vs)^{-1}$ mobility of electrons, which is considered 100 times higher than TiO_2 [9], [11]. High electron mobility helps it to extract electrons efficiently. SnO_2 is easily processed at low temperatures, making it suitable for flexible solar cells and tandem solar cells [11].

Maciej łuszczek et al., used SCAPS-1D software to simulate their analysis on perovskite solar cells with three different ETLs, which are TiO_2, SnO_2, and ZnO. In this study, SnO_2 has the best performance and the highest PCE compared to the other ETLs. This is due to certain attributes that SnO_2 owns, such as better band alignment and stability under ultraviolet illumination [12]. The PCE achieved is 17.08%, 18.33%, and 17.53% by TiO_2, SnO_2, and ZnO, respectively.

The electron mobility of SnO_2 was also investigated by Peng Zhao et al., [13]. They claim that the device performs optimal performance with a PCE of 20.43% when the SnO_2 electron mobility goes beyond 10^{-3} $cm^2.(Vs)^{-1}$[13]. They reported that when the electron mobility is less than 10^{-4} $cm^2.(Vs)^{-1}$, the recombination rate increases and causes the JSC and FF to decrease. The overall PCE has been obtained with an efficiency of 20.43%, JSC 21.5 mA/ cm2, and 81.12% FF.

Another simulation analysis is conducted by Yassine Raoui et al., where the perovskite performance is observed by using different ETL and HTL layers. The final results concluded that SnO_2 achieved the highest PCE compared to the other ETL (TiO_2 and ZnO). The device is structured as FTO/ETL/$MAPbI_3$/Spiro-OMeTAD/Au [14]. The thickness of the ETL layers is equal to 90nm. SnO_2 demonstrated greater current density due to the increased light absorption by the perovskite layer, which is also reflected in the External Quantum Efficiency (EQE) rise in the 300 – 450 nm spectral range [14]. Raoui et al., also found that the

increases in the thickness of ETL lead to attenuation of V_{OC}, J_{SC}, and P_{CE} of both TiO_2 and ZnO. On the contrary, the SnO_2 performance parameter has no obvious variations in its value.

Later, in 2020, Elham and Seyed analyzed the performance of SnO_2 on perovskite solar cells using SCAPS-1D and AMPS simulation software. It was analyzed with the FTO/SnO_2/$CH_3NH_3PbI_3$/NiO/Au structure. The optimum thickness of SnO_2 and its electron density concentration is 140nm to 160 nm and 1018 cm^{-3}, respectively [15]. It also stated that the V_{OC} is reduced in this research when the operating temperature increased from 300K to 440K. This is due to the temperature's dependency on the reverse saturation current.

Furthermore, when the bandgap energy was decreased to some amount, the recombination between the valance band and the conduction band was improved. The majority of free electrons are generated in the conduction band energy, but high temperatures can cause the recombination of holes and electrons, making the bandgap energy unstable [16]. As a result, J_{SC} is decreasing. The drop in V_{OC} and J_{SC} concentrations caused by the increase in working temperature results in a loss in performance.

Methodology - Device simulation

In this study, the PSC device modelling framework comprises five layers. The layers included ITO as the front contact, $CH_3NH_3PbI_3$ (an absorber), SnO_2 and CuSCN as an ETL and HTL, respectively, and Gold (Au) as the back contact (Fig. 1). The PSC efficiency has been optimized by considering a few characteristics focused on SnO_2. For example, SnO_2 thickness, SnO_2 doping density, the operating temperature of photovoltaic cells, and a defect interface are some of the factors listed. The first step of the analysis is carried out by performing a numerical simulation on SCAPS-1D software. The parameter of each structured layer is studied and optimized to achieve the best efficiency. In this simulation, the solar cell's efficiency value (PCE), open-circuit voltage (V_{OC}), short circuit current (J_{SC}), and fill factor (FF) were considered to be recorded and studied from the J-V curve obtained.

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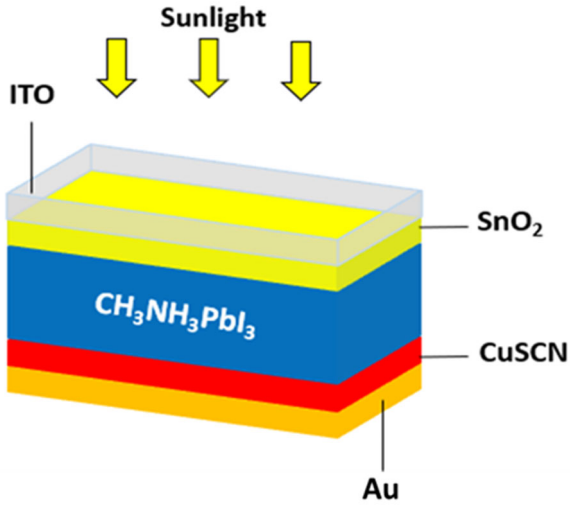


Fig. 1. The structure of PSC for simulation analysis

Table 1. Input parameter of numerical analysis for PSC structure

Layer	CuSCN [17]	ITO[18]	SnO_2 [17]	**$CH_3NH_3PbI_3$** [17]
Layer thickness, d (μm)	350	200	Varied	600
Bandgap energy, E_g (eV)	3.4	3.6	3.6	1.55
Electron affinity, (eV)	2.1	4.1	4	3.93
Dielectric permittivity, ε (relative)	10	10	9	30
Effective density of conduction band, Nc (cm^{-3})	2.2×10^{21}	2×10^{18}	2.2×10^{18}	2.2×10^{18}
Effective density of valence band, Nv (cm^{-3})	1.8×10^{21}	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}
Thermal velocity of electrons, V_e ($cm\ s^{-1}$)	100	0.001	100	2
Thermal velocity of holes, V_h ($cm\ s^{-1}$)	25	0.1	25	2
Electron mobility, μ_e (cm^2/V_s)	1×10^7	1×10^7	1×10^7	1×10^7
Hole mobility, μ_h (cm^2/V_s)	1×10^7	1×10^7	1×10^7	1×10^7
Density of donors, N_D (cm^{-3})	NA	1×10^{19}	Varied	NA

Analysis of working temperature

Working temperature is one of the critical aspects that can greatly impact the photovoltaic performance of the PSCs. In this simulation, the temperature values varied are in a range of 300 K to 500 K with steps of 20 K. The graph shows a downward trend in efficiency as the operating temperature rises with an inclination of -0.0417 %/K, as shown in Fig. 2. All the parameters except J_{SC} decline monotonically as the temperature rises, except for a very slight increase in J_{SC} value. This is due to the fact that the material's carrier concentration, charge mobility, resistance, and bandgap will all be significantly impacted by higher temperatures, resulting in a shift in the PV's key properties [16][19]. In addition, the increased carrier concentration in the semiconductor is generated by a higher operating temperature, particularly the rate of internal carrier recombination. V_{OC} decreases as a result of a rise in reverse saturation current

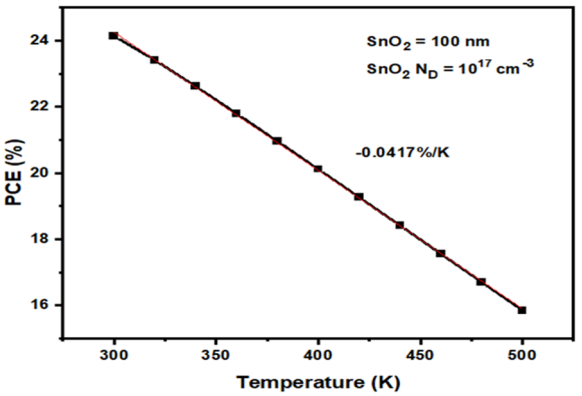


Fig. 2. The analysis of efficiency is based on the variation of working temperature.

Analysis of the variation of SnO2 thickness

The thickness analysis for ETL SnO_2 is performed by altering the layer thicknesses in the same structure. For this study, only the ETL layers are altered for each simulation. The thickness of SnO_2 is varied from 0.005 µm to 1 µm. The P_{CE} is affected by the increasing thickness of ETL (Fig. 3). It causes attenuation of P_{CE}. It also reduces the cell's current and voltage, but the value is insignificant. The P_{CE} constantly decreased until, at a thickness of 0.20 µm, the P_{CE} stopped to decrease, this may be because it has reached the minimum efficiency. The attenuation of P_{CE} due to a thick ETL can have a multitude of impacts. For example, the thicker the ETL may result in the distance for the electron to reach ITO, which is farther and raises the device's series resistance and lowers the fill factor, resulting in the efficiency of the solar cells dropping [20], [21].

Moreover, the electron may tend to enhance the recombination rate and reduce the V_{OC}. According to Xiong et al., [20], highly efficient PSCs have a preference for thin compact layers. It is plausible to believe that a thin compact layer can facilitate electron extraction and reduce charge carrier combination due to its short diffusion length [20]. This is why a thinner ETL is preferable, where more photons can pass through the ETL layer into the absorber layer and be absorbed there. The thickness of 0.5 µm has the highest P_{CE}, which is 25.83%. However, it is not a suitable thickness for the fabrication process. Therefore, in this analysis, the thickness of 0.1 µm is selected as an optimum thickness with a P_{CE} of 24.14%.

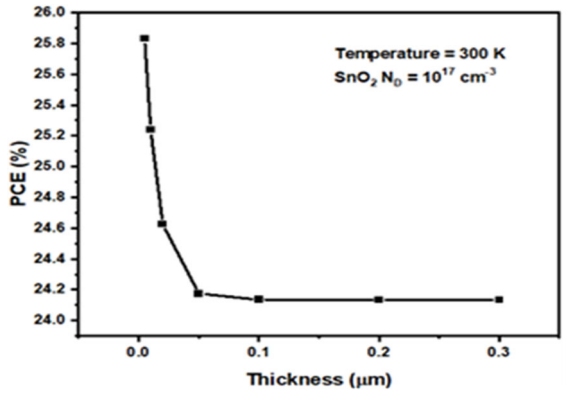


Fig. 3. The analysis of efficiency is based on the variation of SnO_2 thickness.

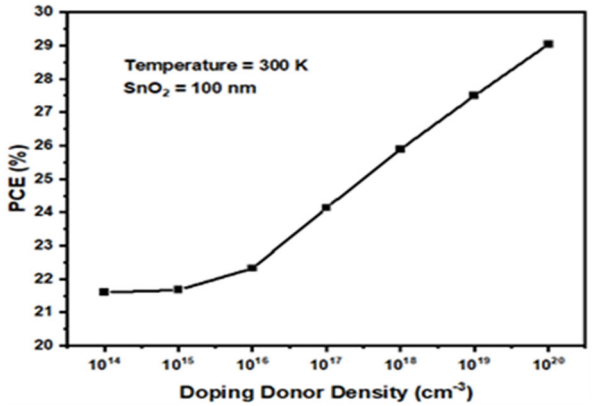


Fig. 4. The analysis of efficiency based on the variation of doping donor density.

Analysis of the variation of doping donor density

Next, the analysis of doping donor density varied from 10^{15} cm^{-3} to 10^{20} cm^{-3}. The efficiency rises with doping donor density (Fig. 4). Increasing doping is equivalent to increasing the number of impurities in the semiconductor to modulate its electrical, optical, and structural properties [19][22]. Therefore, as the amount of impurity in the semiconductor increases, more free charge will be created in the semiconductor. When the free charge increases, it indicates that there is more current flowing through the system at once, causing the series resistance to fall [23]. As a result, efficiency has been boosted. The optimum doping donor density obtained is 10^{17} cm^{-3}. As for the FF, it is greatly affected by the increase in doping density. The FF increased drastically from 10^{15} to 10^{18} cm^{-3}, gradually increasing as doping exceeded 10^{18}.

Analysis of the variation of doping acceptor density

Following that, the analysis of doping acceptor density of CuSCN is varied from 10^{14} cm^{-3} to 10^{20} cm^{-3}. The graph illustrates the efficiency of the device increase as the doping acceptor density increase (Fig. 5). Increasing doping is comparable to increasing the number of impurities in the semiconductor to vary its electrical, optical, and structural properties [18][24]. Doping SnO_2 can boost its characteristics, hence improving the performance of PSCs [20]. Therefore, when the amount of impurity in the semiconductor grows, more free charge will be produced in the semiconductor. When the free charge increases, it implies that more current flows through the system at once, causing the series resistance to falls. As a result, the efficiency has been enhanced.

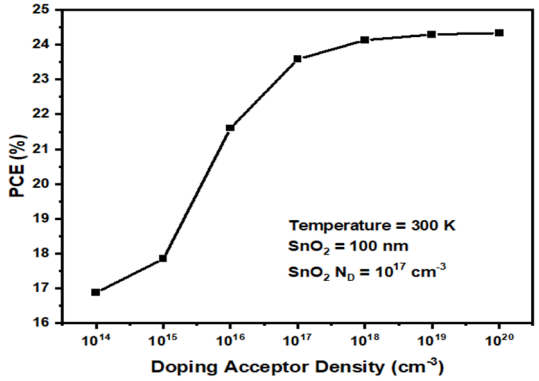


Fig. 5. The analysis of efficiency based on the variation of doping acceptor density.

The variation of recombination rate is analyzed with two types of recombination rates. The first one is the recombination rate of the defect density at the interface of SnO_2 and $CH_3NH_3PbI_3$ layers. This analysis aims to observe the effect of recombination rate on the variation of defect density. In this study, the analysis is further to analyze the recombination rate that occurred at the ETL bulk.

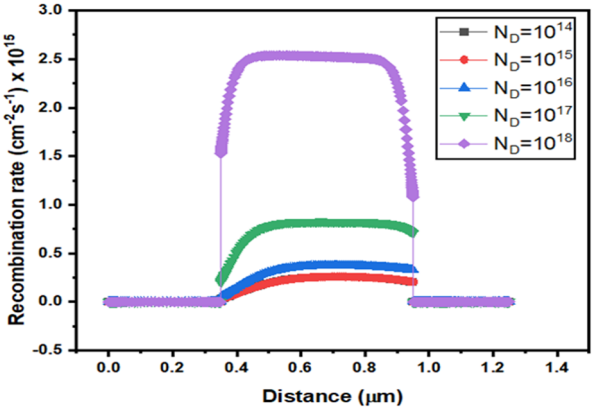


Fig. 6. The recombination rate with the variation of doping density at SnO2 bulk.

A noticeable change is shown as the doping density increases and the recombination rate decreases (Fig. 6). Specifically, ETLs in PSCs gather and transport charge carriers after the perovskite active layer injects electrons, more critically, ensuring effective charge separation and minimizing charge carrier recombination. An inefficient charge transport could damage the ETL/perovskite interface, which could lead to inhomogeneous charge accumulation and significant interfacial recombination [25], [26]. This means that high-quality ETLs must be designed and fabricated to provide efficient charge conduction and, consequently excellent photovoltaic performance.

Analysis of the variation of defect at the interface

Only one sort of defect density interface is analyzed, which is neutral. One faulty interface layer proposed is the defect at the interface between the SnO_2 and $CH_3NH_3PbI_3$ layers. The total density of the interface defect was determined to be in the range of 10^{10} cm^{-2} to 10^{16} cm^{-2}. When the defect density increased in the solar cell, efficiency decreased (Fig. 7). The performance of the PSC increased as a result of reduced carrier loss owing to recombination as a result of low defect density. Higher defect densities, on the other hand, lead to shorter diffusion lengths, which promotes charge carrier recombination [27]. Consequently, there is a considerable decline in PSC performance.

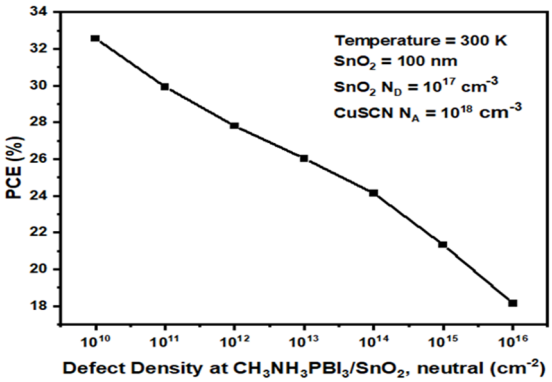
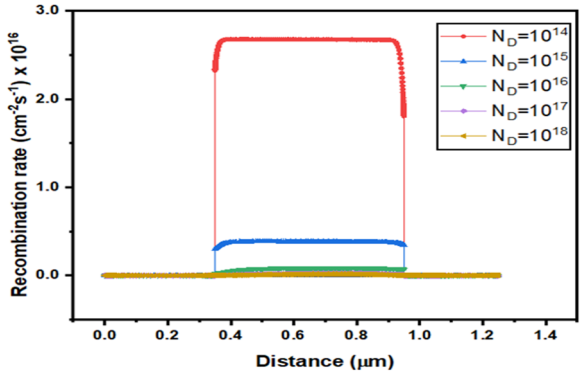


Fig. 7. The variation of defect density at $CH_3NH_3PbI_3/SnO_2$



Fig, 8. The recombination rate with the variation of defect density at $CH_3NH_3PbI_3/SnO_2$

Next, the recombination rate at the interface of $CH_3NH_3PbI_3/SnO_2$. This is performed by changing the defect density value at the interface with a variation range of 10^{14} cm^{-2} to 10^{18} cm^{-2}. This investigation is then carried out to investigate further the impact of defect density at the tin oxide-perovskite interface. The open-circuit losses in the PSCs are caused mainly by the recombination that occurs in the bulk layer of the Perovskite and at either the perovskite transport layer interfaces or the transport layer [25]. Theoretically, the defect density is correlated with the recombination rate, which causes the solar cell efficiency to fall. This is the first-ever study reported on the recombination rate for defect density at $CH_3NH_3PbI_3/SnO_2$. As shown in the figure, the recombination rate decrease as the defect density increases (Fig. 8). The result of this recombination rate is classified as a special instance that will be explored further because it is not a common occurrence.

Analysis of the variation based on optimum value for all parameters

The optimum values for all parameters achieved from the simulation analysis are listed in the table. The simulation findings demonstrate that SnO_2 produced the optimized efficiency by replacing all parameters with the optimal value. The PCE achieved is 24.14%, which is higher than the previous study conducted by Raoui and his team [14] where the highest P_{CE} obtained is below 20%. This demonstrates the need to optimize the ETL layer parameter to attain a higher P_{CE}. Figure 9 displays the optimized PSC structure's J-V curve using SnO_2 as an ETL. FF of 85.99%, V_{OC} of 1.15V, and J_{SC} of 24.42 mA/cm^2 resulted in a P_{CE} of 24.14%.

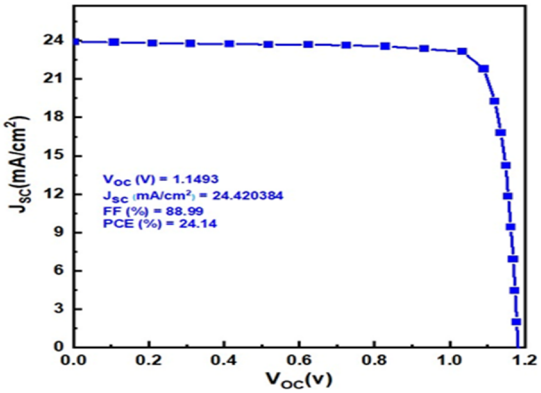


Fig 9. The optimized PSC structure's J-V curve

Table 2. Optimum parameters for SnO_2 as ETL in inorganic perovskite solar cell based on computational analysis.

Parameter	SnO_2
Layer thickness, d (µm)	100
Bandgap energy, E_g (eV)	3.6
Electron affinity (eV)	4
Dielectric permittivity, ε (relative)	9
Effective density of conduction band, N_C (cm^{-3})	2.2×10^{18}
Effective density of valence band, N_V (cm^{-3})	1.8×10^{19}
Thermal velocity of electrons, V_e (cm/s)	100
Thermal velocity of holes, V_h (cm/s)	25
Electron mobility, μ_e (cm^2/V_S)	1×10^7
Hole mobility, μ_h (cm^2/V_S)	1×10^7
Density of donors, N_D (cm^{-3})	1×10^{17}
Density of acceptors, N_A (cm^{-3})	NA

Conclusion

Perovskite solar cell is categorized as the third generation of the solar cell. It consists of 3 main components: the ETL, absorber, and HTL. Various analyses and investigations have been conducted in both simulation and fabrication to improve the efficiency of PSC. SnO_2 is a promising ETL with favorable attributes such as a wide optical band gap (up to 4.0 eV), good transmittance feature, and can be processed by the low-temperature method.

In this study, a PSC structure of ITO/SnO_2/$CH_3NH_3PbI_3$/CuSCN has been analyzed by using the SCAPS-1D simulator. The main purpose of this analysis is to design a PSC structure by utilizing SnO_2 as an ETL. The

parameters involved in designing the finest structure include the working temperature of a solar cell, the thickness of ETL, the dopant density of both ETL and HTL, and the defect density at the ETL and absorber interface. The optimum efficiency obtained is 24.14% at 300 K working temperature, 0.10 µm thickness of SnO_2, 10^{17} cm^{-3} doping donor density, 10^{18} cm^{-3} doping acceptor density, and 10^{14} cm^{-2} defect density.

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