



## Impact of Al on ZnO Electron Transport Layer in Perovskite Solar Cells

Nur Syafiqah Nadiah Mohd Alias<sup>1</sup>, Faiz Arith<sup>1,\*</sup>, Ahmad Nizamuddin Mustafa<sup>2</sup>, Mohd Muzafar Ismail<sup>2</sup>, Nur Fatimah Azmi<sup>1</sup> & Mohd Saifizi Saidon<sup>3</sup>

<sup>1</sup>Faculty of Electronic and Computer Engineering, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100, Melaka, Malaysia

<sup>2</sup>Faculty of Electrical and Electronic Engineering Technology, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100, Melaka, Malaysia

<sup>3</sup>Faculty of Electrical Engineering Technology, Universiti Malaysia Perlis, 02600, Perlis, Malaysia

\*E-mail: faiz.arith@utem.edu.my

### Highlights:

- High PCE, up to 17.59%, obtained for Al-doped ZnO ETL material in a PSC.
- 1 mol% Al concentration in the ZnO ETL produced the highest PCE values after being studied in the range between 1 and 4 mol%.
- A high PCE value was achieved at a thickness of 50 nm, which is thicker than the conventional value of approximately 10 nm, resulting in a simple and cheaper fabrication process.

**Abstract.** Perovskite solar cells have shown remarkable performance and improvements in terms of solar cell efficiency. The ETL material is one of the important components in perovskite solar cells in conducting electrons to produce current. Here, ZnO was used as ETL material in a perovskite solar cell using the SCAPS 1D simulation software. The ZnO ETL showed poor cell efficiency due to its reaction with the perovskite material. A small amount of Al doped into ZnO was introduced to enhance the physiochemical properties of the ZnO against perovskite materials. Al concentrations were varied between 1 and 4 mol% to observe the effect on cell efficiency. Compared with a conventional ZnO ETL solar cell with 0 mol% Al perovskite, the Al-doped based solar cell showed better performance. Meanwhile, perovskite solar cells with 1 mol% Al-doping and appropriate layer thickness showed the best cell performance in improving the charge transport mechanism, resulting in increased cell efficiency. Thus, the parameters studied can be a guide in the fabrication process.

**Keywords:** Al-doped ZnO; ETL; perovskite solar cell; power conversion efficiency; SCAPS 1D.

## 1 Introduction

The world is witnessing an increase in the world's population every year, which causes an increase in the demand for energy. In addition, technological advances

in human daily life are also leading to increased energy consumption. Conventional energy from fossil fuels such as coal, petroleum and natural gas is non-renewable, so those sources will dwindle and disappear one day. Solar cells are among the most widely used renewable energy sources in energy production. This source is unlimited, as long as the earth receives sunlight. There are many types of solar cells that have been developed and commercialized such as silicon-based, thin-film, and organic solar cells, and many more [1-3]. Recently, perovskite solar cells (PSC) have caught the attention of researchers, with a very impressive increase in power conversion efficiency (PCE) in a relatively short period of time. A PSC consists of several main layers, i.e., an electron transport layer (ETL), an organic-inorganic perovskite layer, and a hole transport layer (HTL) [4-7]. The ETL material is an important element in improving cell performance by ensuring rapid charge transport without being hindered by charge recombination from the perovskite active layer to the electrode [4,5].

In 2020, Ulsan National Institute of Science and Technology (UNIST) in South Korea developed a PSC with a PCE as high as 25.5%. The HTL layer of Spiro-OMETAD was chemically enhanced by the fluorination method [8]. Recently, a leading company in the field of perovskite solar cells, Oxford PV, stated that a PCE value of 29.52% had been achieved, which is the highest PCE value ever recorded to date [9]. However, these conventional PSCs commonly utilize  $\text{TiO}_2$  as the ETL material.  $\text{TiO}_2$  offers high photostability and an appropriate band gap for efficient electron transport from the perovskite layer and functions well as a hole-blocker at the perovskite interface [9,10]. However, to produce a high-quality  $\text{TiO}_2$  ETL, an annealing process with high temperatures (above 500 °C) is inevitable, which causes the feasibility of flexible PSCs to be disrupted [8,11]. ZnO has been proposed as an alternative ETL material, which provides almost the same band gap and higher electron mobility compared to a  $\text{TiO}_2$  ETL as well as a low thermal budget fabrication process [4,11]. In a PSC, the electron extraction capability of the ETL is dependent on two important factors, the conductivity and the work function (WF) of the ETL. The electrical conductivity of ZnO can be improved by extrinsically doping a small amount of impurity [11,12] of Al. Since the ion radius of  $\text{Zn}^{2+}$  (0.74 Å) is larger than that of  $\text{Al}^{3+}$  (0.54 Å), a small amount of  $\text{Al}^{3+}$  can sneak in to replace  $\text{Zn}^{2+}$  at the lattice site. Then it can act as an additional dopant, resulting in enhanced conductivity of the ZnO ETL, at the same time maintaining excellent transparency in the visible light region [12].

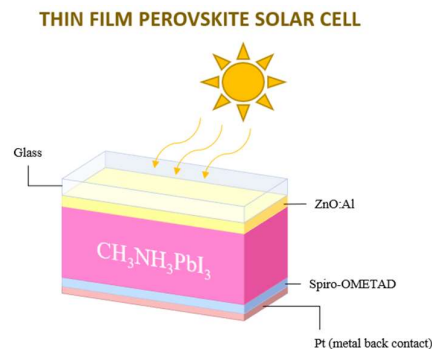
Here, Al doped ZnO was analyzed as ETL material in a PSC using the SCAPS 1D simulation software. In addition, several key parameters, i.e., layer thickness, operating temperature, doping concentration, and defect density, were varied to obtain the optimum device structure [13-16]. It was found that a small amount of Al contributed to the enhancement of the electrical conductivity in the ZnO ETL.

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A conventional ZnO ETL of PSC produced a relatively lower PCE due to the organic cation ( $\text{CH}_3\text{NH}_3^-$ ) of perovskite reacting with the ZnO compared to Al doped with ZnO [17]. 1 mol% of Al doped ZnO ETL demonstrated the best cell performance, producing PCE as high as 17.59%. This finding is supported by several previous reports, where there was a solubility limit of  $\text{Al}^{3+}$  substitution in ZnO [12,17].

### 2 Perovskite Solar Cell Simulation

In this study, the SCAPS 1D simulation software was used to study the electrical characteristics of PSC. The simulation program was designed and introduced by the Department of Electron and Information Systems (ELIS) at the University of Gent, Belgium [18]. Initially, this software was developed for cell structures of the thin film group. Nevertheless, this software has been improved in its capabilities such that it is now also applicable to crystalline solar cells (Si and GaAs family), amorphous cells (a-Si and micromorphous Si) as well as the emerging solar cells of perovskite. Figures 1 and 2 show the layer structure of the PSC that was used in the SCAPS simulation and the energy level diagram of each layer, respectively. The PSC was simulated with  $\text{CH}_3\text{NH}_3\text{PbI}_3$  as perovskite layer, Al doped-ZnO and Spiro-OMETAD as ETL and HTL, respectively. This study focused on the optimization of the Al doped-ZnO layer, where several important parameters were varied and analyzed, i.e., layer thickness, doping concentration, operating temperature, and defect density. Furthermore, a small amount of aluminum (Al) was extrinsically doped into the ZnO layer in the range between 0 and 4 mol% to obtain the optimal total concentration. Merging various material parameters into SCAPS for all aspects of the analysis resulted in changes in values of  $V_{oc}$  and  $J_{sc}$ , subsequently affecting FF and PCE. Table 1 shows a description of the parameters in the simulation and the basis of the parameters that were used in this study.



**Figure 1** Layer structure of perovskite solar cells with aluminum doped ZnO (ZnO:Al) in SCAPS.

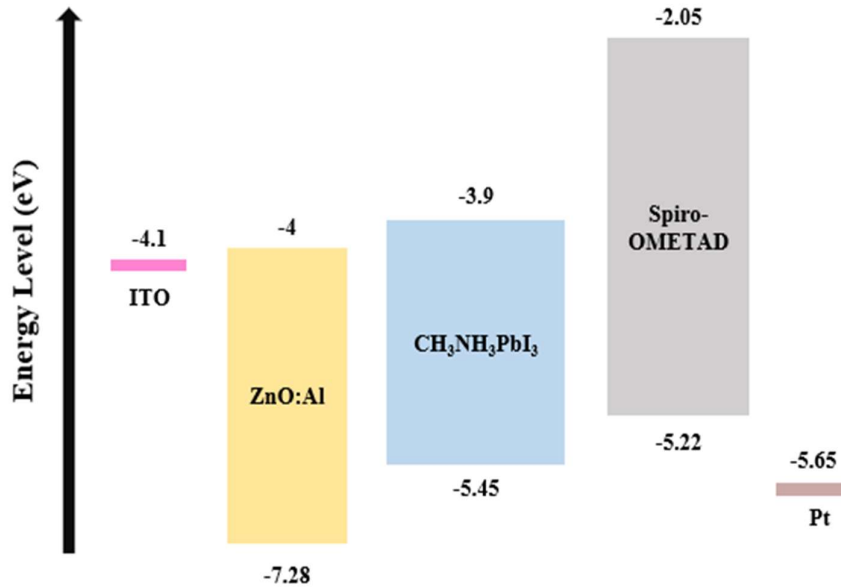


Figure 2 Energy level diagram of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite solar cells.

Table 1 Parameters for perovskite solar cell layer.

	Spiro-OMETAD [19, 20]	CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> [19, 21]	Concentration of Al (mol%) [16]				
			0	1	2	3	4
Thickness (nm)	100	400	220	220	220	220	220
E <sub>g</sub> (eV)	3.17	1.55	3.28	3.25	3.1	3.08	3.05
χ (eV)	2.05	3.9	4	4	4	4	4
ε/ε <sub>0</sub>	3	10	9	9	9	9	9
N <sub>c</sub> (cm <sup>-3</sup> )	2.2×10 <sup>18</sup>	2.75×10 <sup>18</sup>	2×10 <sup>18</sup>	2×10 <sup>18</sup>	2×10 <sup>18</sup>	2×10 <sup>18</sup>	2×10 <sup>18</sup>
N <sub>v</sub> (cm <sup>-3</sup> )	1.8×10 <sup>19</sup>	3.9×10 <sup>18</sup>	1.8×10 <sup>19</sup>	1.8×10 <sup>19</sup>	1.8×10 <sup>19</sup>	1.8×10 <sup>19</sup>	1.8×10 <sup>19</sup>
μ <sub>e</sub> (cm <sup>2</sup> /V <sub>s</sub> )	2×10 <sup>-4</sup>	10	43.00	300	13.84	13.35	15.02
μ <sub>h</sub> (cm <sup>2</sup> /V <sub>s</sub> )	2×10 <sup>-4</sup>	10	25	25	25	25	25
N <sub>D</sub> (cm <sup>-3</sup> )	0	0	2.9×10 <sup>15</sup>	7.25×10 <sup>18</sup>	1.02×10 <sup>19</sup>	1.46×10 <sup>19</sup>	1.06×10 <sup>19</sup>
N <sub>A</sub> (cm <sup>-3</sup> )	1×10 <sup>19</sup>	1×10 <sup>9</sup>	0	0	0	0	0
V <sub>e</sub> (cm s <sup>-1</sup> )	1×10 <sup>7</sup>	1×10 <sup>9</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>
V <sub>h</sub> (cm s <sup>-1</sup> )	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>	1×10 <sup>7</sup>

### 3 Results and Discussion

#### 3.1 Effects of Thickness of Al doped ZnO ETL

The thickness for each concentration of the Al doped ZnO (ZnO:Al) ETL varied in the range between 50 nm and 1000 nm. The selection of this range was appropriate considering the capabilities of cell fabrication. PCE values for all devices peaked at a thickness of 50 nm. Moreover, 1 mol% Al showed the highest PCE value, i.e., up to 17.6%, at the latter thickness as well as throughout the entire range studied, as shown in Figure 3.

The ZnO layers without Al doping showed the lowest peak value at lower thickness. In general, the PCE value decreased with increasing ETL thickness. Increasing the thickness of the ETL increases the chance of recombination, as the charge must travel longer distances for diffusion, which leads to PCE reduction. However, its efficiency decreases at a certain rate with increasing thickness, which is most likely due to the increased recombination [22,23].

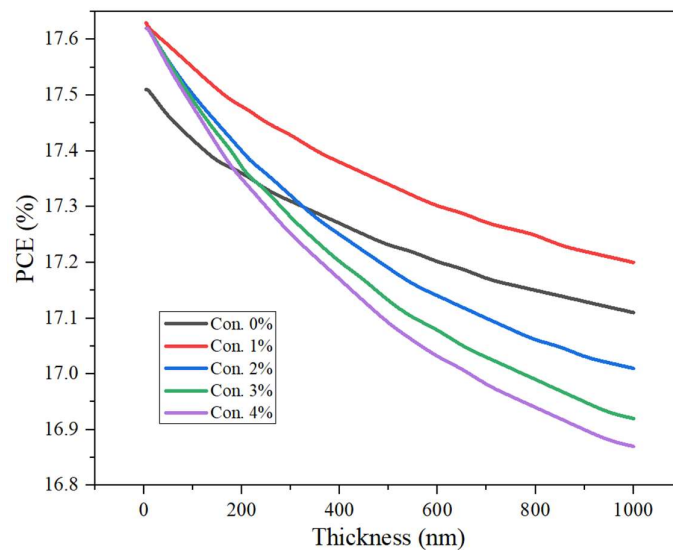


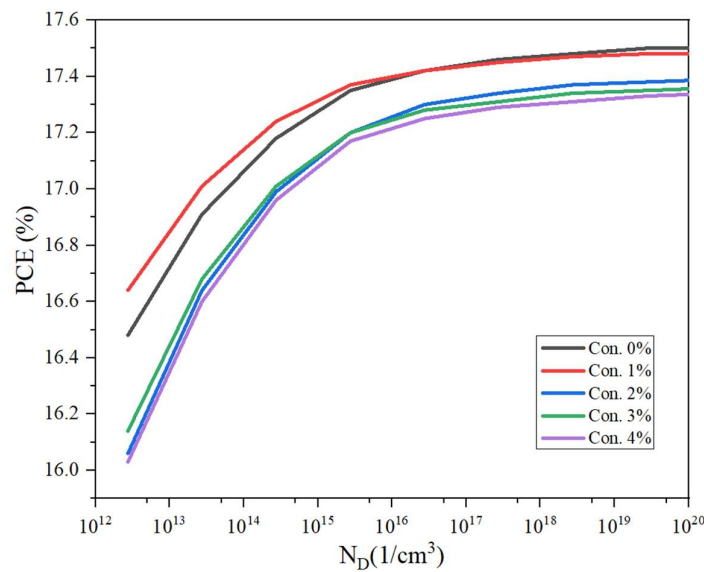
Figure 3 Cell performance with variable thickness of Al doped ZnO ETL PSC.

#### 3.2 Effects of Doping Concentration in Al doped ZnO ETL

The doping concentration in the ETL plays a vital role in accelerating the electron flow and hence the current generation. The insertion of n-type Al dopant into the ZnO ETL replaces the  $Zn^{2+}$  and effectively suppresses the ETL/perovskite

interface defects, resulting in an enhancement of the charge carrier conductivity. In addition, the Al dopants also increase the free carrier concentrations and introduce a donor level at 120 meV below the conduction band, which is promoted to the appropriate band alignment [24]. An optimal value with the lowest possible doping concentration will result in a high-performance solar cell. Here, the doping level varied from  $10^{12}$  up to  $10^{20}$   $\text{cm}^{-3}$  for each device. The other parameters, i.e., layer thickness, temperature, and defect density, were set fixed to provide a fair comparison. Figure 4 shows that the cell efficiency increased with doping concentrations up to  $1 \times 10^{16}$   $\text{cm}^{-3}$  before reaching a plateau for all devices.

It was observed that the device with 1 mol% of Al doped ZnO had the best performance at low doping concentrations and performed almost identical to the devices without Al doping with increasing doping concentration. The increment of cell efficiency is due to the increased electric field within the cell. The increase in the electric field further intensifies the charge carrier separation activity, which in turn will improve the performance of the solar cell [25].



**Figure 4** Cell performance with various doping concentrations of the Al doped ZnO ETL PSC.

### 3.3 Effects of Working Temperature

To observe cell performance in the real world, an operating temperature between 300 K to 400 K was simulated. From Figure 5, it can be seen that all devices were

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severely affected by the operating temperature. Several parameters in semiconductor devices, such as electron and hole mobility, carrier concentration, and material band gap, will react and be influenced by high temperatures. This results in a reduction of electron flow, which in turn affects the efficiency of the cell.

It can be observed that all devices showed an even downward trend with an increasing temperature of approximately  $-0.2\%/K$ . However, at lower temperatures, a relatively smaller gradient was observed for all devices. This is most probably due to the insertion of the Al dopant, which slows the bandgap narrowing mechanism. Generally, the bandgap narrows with increasing temperature, which probably promotes the recombination between charge carriers within the valence band and the conduction band. In addition, the cell efficiency decrease may also be due to the increase of the defect density inside the layers, which acts on the deformation stress [2,26,27].

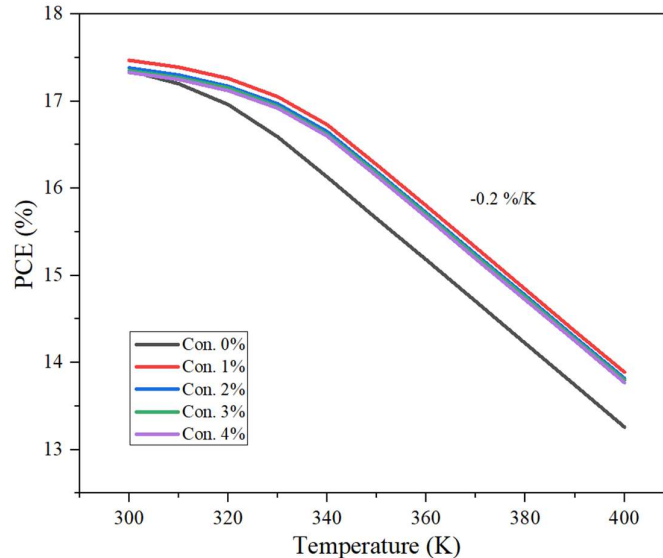


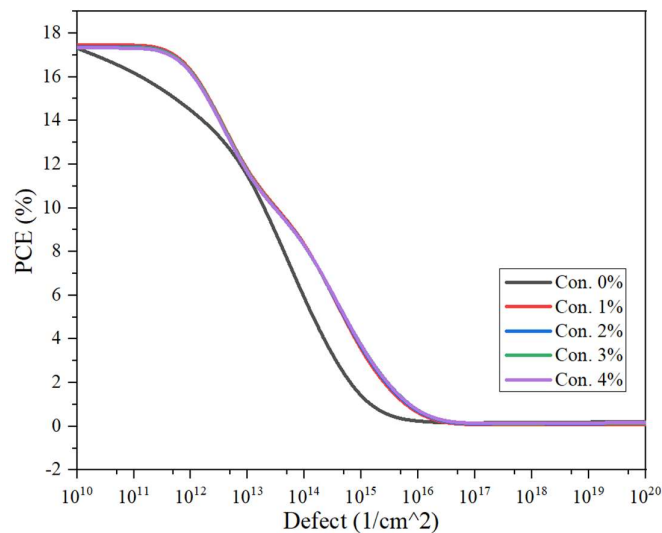
Figure 5 Effect of operating temperature to the cell efficiency for each PSC.

### 3.4 Effects of Defect Density

To observe the effect of defect density in the interlayer between the ETL and the absorbance layer of the PSC, the interface density of state ( $N_{it}$ ) was varied from  $10^{10}\text{ cm}^{-2}$  to  $10^{20}\text{ cm}^{-2}$ . At lower defect levels, cell performance was not much reduced or impaired, as shown in Figure 6. However, all cells began to show effects, where the efficiency for all cells decreased significantly with increasing

defect density. However, the PSCs without Al-doped ETL experienced a rapid reduction even at a lower defect density of  $1 \times 10^{14} \text{ cm}^{-2}$  compared to the other cells, as shown in Figure 6. The decrease in PCE at higher defect concentrations is due to the fact that the increase in interface trap density causes the recombination rate to surge, which affects the charge carrier conduction and thus deteriorates cell performance. On the other hand, at lower defect density, the carrier diffusion length is high, which leads to a lower recombination rate, resulting in better cell performance [28,29].

By optimizing all parameters, i.e., layer thickness, doping concentration, operating temperature, and defect density, the highest PCE values were obtained for each of the Al-doped ZnO ETL in PSC, as shown in Table 2. The Al-doped ZnO with 1 mol% of Al produced the highest PCE among the devices studied. The PCE decrease with Al concentrations larger than 1 mol% can be explained according to the doping rules, where theoretically the substitution of  $\text{Al}^{3+}$  at the  $\text{Zn}^{2+}$  site takes place, which occurs up to the solubility limit of Al in ZnO of 3 mol%. Thus, an increase in carrier concentration up to the solubility limit is rational, but when it exceeds the solubility limit, a new phase of  $\text{Al}_2\text{O}_3$  arises and substitution of Al is no longer as effective as before. Therefore, cell efficiency begins to decrease at an Al dopant concentration of 4 mol%. This shows that by varying the important parameters of the ETL, it is possible to further increase PCE without having to reduce the layer thickness.



**Figure 6** Effect of interface defect density concentration on the PCE performance of the Al-doped ZnO ETL PSC.



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**Table 2** PCE for different Al dopant concentrations in the ZnO ETL at optimized parameters of 50 nm thick,  $N_D$   $1 \times 10^{18}$   $\text{cm}^{-3}$ ,  $N_A$   $1 \times 10^{10}$   $\text{cm}^{-2}$  at room temperature conditions.

Concentrations of Al (mol%)	PCE (%)
0	17.46
1	17.6
2	17.56
3	17.56
4	17.55

## 4 Conclusion

This work reported the best way to increase the efficiency of PSCs by introducing Al doping into the ZnO ETL. PSCs were simulated using the SCAP 1D simulation software to achieve optimal cell parameters. In-depth analysis and correlation of layer thickness, doping, operating temperature, and interface defect density of an Al-doped ZnO photoanode were conducted. Simulated PSC cell efficiencies as high as 17.59% were obtained using a 1-mol% Al-doped ZnO photoanode with a layer thickness of 50 nm. According to previous reports, to achieve PCEs above 15%, a thickness as thin as 10 nm ZnO ETL is required, which requires the use of sophisticated and expensive control equipment. Thus, this work proved that by varying and optimizing the key parameters of the Al-doped ZnO ETL, improved performance of PSCs can be achieved and at the same time be cost-effective. Even though the cell efficiency of the optimized PSC was not significantly affected by the presence of Al-dopant, it did improve the quality of the interface layer, as evidenced by the interface defect density analysis. In addition, these parameters can also be used as a guide for the cell fabrication process.

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