

Analysis of Inverted Planar Perovskite Solar Cells with Graphene Oxide as HTL using L9 OA Taguchi Method

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ABSTRACT

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In this work, the Taguchi Method approach is used to optimize graphene oxide (GO) as the hole transport layer (HTL) in inverted perovskite solar cells (IPSC). By using this method, the data from the numerical modelling Solar Cell Capacitance Simulator-One Dimensional (SCAPS-1D) was optimized. While it has distinct parameter results and diverse causes, it also takes a long time to complete the analysis process. The Taguchi method is reported to be able to find the most significant factor and reduce the parameter variations in less time. The Taguchi algorithm is used in this experiment because it is based on orthogonal array (OA) experiments, which provide a substantially smaller variance for the experiment with optimal control parameter values. SCAPS-1D software was used to simulate the IPSC with GO as HTL. The results obtained with the software are then analysed and compared with the performance of the solar cell. The final results show that the Taguchi Method optimized IPSC with GO as HTL achieved better Power Conversion Efficiency (PCE) compared to previous researchers, with the efficiency increasing from 18.53% to 23.408%.

1. Introduction

Renewable energy sources, as environmentally friendly energy resources, will become even more important in the future, because they are unlimited and provide additional energy forms along with the existing conventional power plants [1]. The common source of renewable energy used worldwide is solar energy. It is a renewable energy source capable of providing sufficient power to households. Solar energy is an infinite renewable energy source generated from the sun's electromagnetic radiation. Solar PV technology is a promising way to harness solar power as it generates electrical power on-site directly from solar radiation through the photovoltaic effect of the employed solar

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cells [2]. When a PV cell is exposed to sunlight, solar energy is transformed into electricity by the photovoltaic effect. Photovoltaic effect plays a key role in producing electricity from solar radiation.

Perovskite has the outstanding capabilities for use as light harvesters in solar cells due to the ability to adjust its optical properties. The perovskite materials can be used not only as light-absorbing layer, but also as an electron/hole transport layer due to the advantages of its high extinction coefficient, high charge mobility, long carrier lifetime, and long carrier diffusion distance [3-5]. It also acts as a charge carrying material. Perovskite solar cells have two structure which is n-i-p (regular) and p-i-n (inverted). The different between these two structures is the position of HTL and ETL as shown in Figure 1. For the perovskite solar cells with inverted structure (IPSC) the HTL layer place on top of TCO (transparent conducting oxide) substrate while perovskite solar cells with regular structure (PSC) the HTL layer place between metal and perovskite substrate.

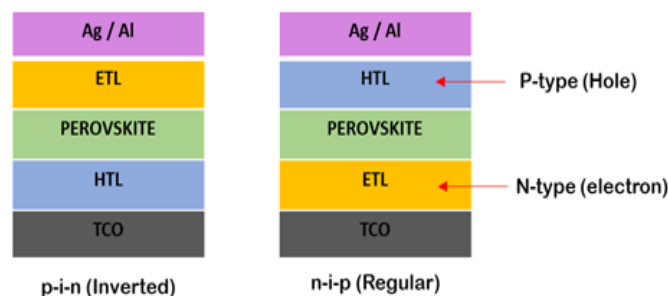


Fig. 1. Inverted and regular planar structure of perovskite solar cells

The ETL and HTL layers offers a driving force for the carrier transport. These two layers may also provide protection of the perovskite layer from moisture and metal diffusion from connection. Both of it can reduce the cell's PV performance. After that, TCO substrate is needed for perovskite solar cells. It should have excellent transmission, conductivity, and adherence to the deposited layers. The purpose of the Taguchi method is to find factors that are most important in achieving useful goals in a manufacturing process. These factors are varied over two or more levels in a systematic manner. The experiments are designed according to an Orthogonal Array to show the effects of each potential primary factor. The presence of Orthogonal Array (OA) is one of the major advantages of the Taguchi method. Taguchi method also helps in minimizing the number of experiments required in optimization purpose [6-11]. The important characteristics of HTL is high conductivity, high transparency, favorable process solution and stability, high WF and good hole mobility [12].

In this experiment, IPSC devices can improve the stability of solar cell compared to Planar Perovskite Solar Cells (PSC). Next, OPV cells have a substantially lower efficiency than inorganic-based devices, which is a major flaw at the moment. Organic semiconductors have a bigger band gap than inorganic semiconductors. Nevertheless, OPV use Spiro-OMeTAD which is dopants in that material show strong water absorbency, stability issues arising from photochemical degradation (exposure to moisture or humidity) which seriously threatens the service life of PSCs. The costs of organic materials such as spiro-OMeTAD, PEDOT: PSS, PTAA and P3HT are all prohibitively high for large-scale applications. The industrial growth and market potential of photovoltaic solar cells (PSCs) is constrained by their high cost and instability in water, heat, and light, despite the fact that all of these materials provide higher open-circuit voltages and higher efficiencies [13].

1.1 Hole Transport Layer

Hole transport material is an important component in Inverted Perovskite Solar Cell (IPSC). It has the function of adjusting the energy compatibility, optimize the interface and to gain higher PCE. The hole transport layers can be divided into three categories, which are organic HTL, Polymeric HTL and inorganic HTL [14]. HTL is inorganic p-type due to higher mobility, chemical stability and increased transparency in the visible region. Due to its good transparency, it will significantly increase the efficiency [15]. According to Salim *et al.*, [16], the transportation layers utilized in the device architecture determine the stability and performance because it serve numerous features in PSCs. Where it acts as energetic barrier between ETL and perovskite layer by blocking the electron transfer. It can also avoid the deterioration and corrosion that can occur at the metal-perovskite interface without an HTL [17]. According to several research results, inorganic materials that usually used as HTL are CuSCN, NiOx, CuO and GOx. For example, Nickel oxide (NiOx) as HTL with expected stability because it has good optical transparency, prevents electron leakage and has appropriate energy levels. An excellent HTL must have the optimum energy level for the perovskite material, as well as high electrical conductivity, optical transparency, and chemical stability [16-18].

1.2 Graphene Oxide (GO)

Graphene has a hexagonal structure and composes of sp² hybridized carbon atoms. From previous research, GO is known as an excellent interfacial material [19]. GO has sparked a lot of interest due to its remarkable characteristics, reliability, low processing cost, large-scale production possibilities, and good dispersibility in a variety of solvents [20]. Due to its high charge mobility, the GO can provide energy compatibility by providing a sufficient exciton breakup the path and charge transport with the photoactive layer, either as transparent conductive electrodes, ETL, or HTL [21]. According to Cho *et al.*, [22], GO and r-GO, which have a two-dimensional sheet structure consisting of carbon atom monolayers, are considered to have a high potential for use in PSCs when compared to carbon materials because it has good electrical conductivity and a large specific surface area. Graphene derivatives GO and r-GO have thus been used as a substitute for spiro-OMeTAD as HTL in PSCs due to the aforementioned features. Due to the device's mobility and stability, this could be a viable solution both economically and technically [13].

Replacing organic HTL with GO could improve efficiency, and r-GO could be a viable candidate for boosting cell stability. PEDOT: PSS has been modified with GO materials possess several advantages. The advantages that are suitable for OPVs application is high surface area, conductivity, and mechanical elasticity. Besides, GO wide bandgap (≈ 3.6 eV) can enhance the potential for utilization as an electron blocking layer [23]. GO as HTL in inverted PSC shows that GO has a suitable WF of - 4.9 eV which accumulates efficient hole extractions from perovskite to GO. Extractions from perovskite to GO; thus, forming homogeneous big domains with enhanced surface coverage and appropriate vertical resistivity is facilitated. As a result, the PCE of the PVSC with GO HTL increases [15]. This GO modified PEDOT: PSS maintained 83.5% of its initial PCE value, indicating that using the GO-modified PEDOT: PSS as HTL rather than the unmodified PEDOT as HTL appears to be a good strategy for enhancing the effectiveness and stability of PSCs [24].

1.3 Taguchi Method

Taguchi method is a method used to improve the quality of the analysed processes and products. This method, which is based on a predictive model, specifically the analysis of S/N ratios, permits the

control of many components in order to limit parameter changes and alter the experimental process under the same experiment circumstances, resulting in improved response analysis and physical attributes [25]. Indeed, the Taguchi method has been effectively implemented for a variety of systems and materials with a high level of complexity in a variety of scientific, engineering, and industrial fields, and it has been proven to be a reliable and reputable methodology with high reliability and product quality [26-28].

The Taguchi technique is used to identify the most essential aspects in accomplishing beneficial outcomes in a manufacturing process. These variables are systematically altered at two or more levels. According to H. Absike, described in this paper the development and characterization of copper oxide thin films using the Taguchi method to optimize the structural and optical properties of films formed under optimum conditions. Three parameters, namely Cu^{2+} content, preheating temperature TP, and final heat-treatment temperature, were chosen [29]. The signal to noise ratio (S/N) was determined to examine the experimental data and determine the elements that influenced the response. According to Taguchi method, the best values for each chosen experience were determined by comparing the signal-to-noise (S/N) ratio in order to allow "the greater the better" response and make the device superior performance [30].

2. Methodology

2.1 Simulation of IPSC Device

Figure 2 shows the inverted perovskite solar cells structure. In this experiment, there are five layers in the IPSC device. First layer is FTO function as front contact where the metal work function for FTO is 4.4eV. Then followed by GO as the HTL, the active layer, which is perovskite, $\text{CH}_3\text{NH}_3\text{PbI}_3$, TiO_2 as ETL and Ag as back contact where the metal work function for Ag is range from 4.26-4.73eV.

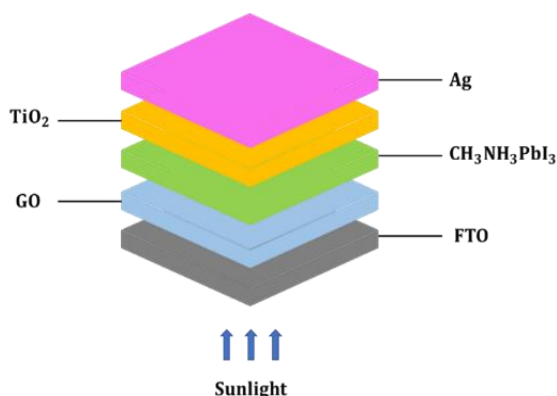


Fig. 2. Inverted Perovskite solar cells structure

There are 4 parameters have been optimized to obtain optimal effectiveness of IPSC device based on the presence of GO as HTL. The parameters are the thickness of GO, Graphene Oxide doping thickness, a deformity interface at each layer and a solar cell's operating temperature. Table 1 shows the numerical analysis input parameter for each component mentioned in the previous research [20]. Finally, after simulation, the I-V curve will be analyzed and see the performance of IPSC through PCE. From I-V curve it shows the V_{OC} , J_{SC} , FF and PCE. In any case, if the results do not match the ideal result, the method should be reviewed until the ideal result is achieved.

Table 1
 SCAPS-1D input parameter of numerical for each layer in IPSC devices [20]

Material properties	FTO	TiO ₂	CH ₃ NH ₃ PBI ₃	GO
Thickness, d (um)	0.5	0.05	1	0.1
Bandgap, Eg(ev)	3.5	3.2	1.58	2.48
Electron affinity χ (ev)	4	4	3.9	2.3
Dielectric permittivity, ϵ_r (relative)	9	9	10	10
CB effective density of states, N_C (cm ⁻³)	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}
VB effective density of states, N_V (cm ⁻³)	1.8×10^{19}	1.8×10^{19}	1.0×10^{19}	1.8×10^{19}
Electron thermal velocity (cms ⁻¹)	1.0×10^7	1.0×10^7	1.0×10^7	5.2×10^7
Hole thermal velocity (cms ⁻¹)	1.0×10^7	1.0×10^7	1.0×10^7	5.0×10^7
Electron mobility, μ_n (cm ² . V ⁻¹ s ⁻¹)	20	20	2.2	26
Hole mobility, μ_p (cm ² . V ⁻¹ s ⁻¹)	10	10	2.2	123
Donor density, N_D (cm ⁻³)	2.0×10^{19}	2.0×10^{18}	1.0×10^{13}	-
Acceptor density, N_A (cm ⁻³)	-	-	1.0×10^{12}	2.0×10^{18}
Defect GO/CH ₃ NH ₃ PBI ₃	-	-	-	1×10^{10}

2.2 Optimization Approach

This project utilized the Taguchi method to determine the optimal solution for IPSC devices. Taguchi method emphasizes the design and execution of experiments that can determine the effect of input process parameters on output responses. By analyzing the impact of different components, the optimal combination of factors can be identified. In this experiment, need to identify the input process parameter which is V_{oc} , J_{sc} , FF and PCE. Taguchi method also can improve the quality of the analyzed processes and products. The increment the number of process parameters, many experiments need to execute.

2.2.1 Selection of orthogonal array

The Taguchi method employs a unique design of orthogonal array (OA) to address this issue and explore the whole process parameter space with just a few experiments. OA can reduce the number of experimental. In order to choose an appropriate OA, it depends on the total of parameter. In these experiments, L₉ OA was used. It has 9 experiments with 4 control factors. Taguchi L₉ OA greatly can reduce the number of tests and increase the efficiency [30,31]. The experimental layout for the process parameters employing an orthogonal array of L₉ (3⁴) elements. The L₉ orthogonal array is used to comprehend the effect of four control factors whose levels were altered throughout nine rows of experiments.

2.2.2 Performance specification

The IPSC parameter such as V_{oc} , J_{sc} , FF and PCE should meet the performance specification which is larger-the-better. In this experiment consist of 4 control factors and L₉ orthogonal array with 9 experiments was used. The target is to maximize the respond. The bigger the S/N, the better it is calculated to be. Since our aim is to maximize strength, the compressive strength in the current study should be higher. Eq. (1) shows the η for the quality characteristics of higher-the-better. The number of tests and experimental value of the obtained respond characteristics being represented as n and Y_n respectively [25,31].

$$\eta = -10 \log_{10} \left[\frac{1}{n} \sum \left(\frac{1}{Y_1^2} + \frac{1}{Y_2^2} + \dots + \frac{1}{Y_n^2} \right) \right] \quad (1)$$

Experimentation can be utilized, for instance, to examine the influence of Factor A at level 3 (A_3). Level A_3 of factor A was observed in experiments 7, 8, and 9. The average S/N ratio for these experiments, denoted by, m_{A_3} is calculated as follows [25,31]:

$$m_{A_3} = \frac{1}{n} (\eta_7 + \eta_8 + \eta_9) \quad (2)$$

2.2.3 Confirmation experiment

The final step for the design of experiment process is the confirmation experiment. The aim of the confirmation experiment is to validate the analysis phase's conclusions [30]. Once the optimal level of process parameters has been determined, a confirmation test or final simulation is conducted to confirm the accuracy of the Taguchi Method prediction. The confirmatory test is unnecessary if the optimal combination of parameters and their levels coincides with one of the experiments in the orthogonal array.

In this experiment, initially 5 control factors were selected and L_8 orthogonal array was used. It has 8 experiments with 5 control factors. However, the multiple result showed that only 4 control factors gave higher significant to S/N ratio compared to temperature always pooled or neutral in every process parameter. So L_9 orthogonal array with 9 experiments and 4 control factors was selected [25,31].

$$Y_{opt} = m + \sum_{i=1}^n (m_{iopt} - m) \quad (3)$$

where m is average performance, n represents the number of process parameters and m_{iopt} is average process parameter at the optimal level.

3. Result and Discussion

Different parameter has been analyzed using Taguchi method L_9 (OA) and the results will be discussed to obtain a better performance of the device. Then, all the parameters will be related with theory and discussed regarding problem faces during an experiment.

3.1 Analysis of Graphene Oxide (GO) Thickness

Initially, the thickness of GO has been varied from 10 nm to 100 nm. The trend of a varied thickness of GO layer for four parameters is illustrated in Figure 3. It can be observed that as the thickness of GO was varied from 10 nm to 100nm, the power conversion efficiency of IPSC decreases from 19.133% to 18.532%, FF slightly raised from 81.360% to 81.481% and J_{sc} is slightly decreased from 25.125 mA cm⁻² to 24.174 mA cm⁻² while V_{oc} value is slightly increased from 0.935V to 0.942V.

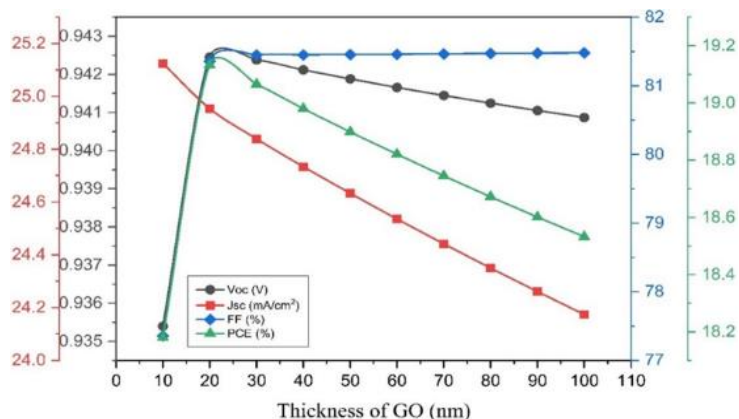


Fig. 3. The variation of GO thickness on PCE, FF, J_{sc} and V_{oc}

The thickness of GO cannot be too thin or too thick based on previous study. From previous research, HTL was applied to transport holes from the perovskite layer to the electrode by creating a barrier between the two substances. If HTL is excessively thick, the series resistance rises, and it becomes more difficult to transfer holes to the electrode. If HTL is too thin, it may not offer sufficient space between these layers. As a result, recombination will occur at the interface of the perovskite layer and electrode which is can affect the performance of solar cells [20].

3.2 Analysis of Interface at GO/Perovskite Layer

The interface between GO/Perovskite are very important in order to improve the performance of PCE. The higher the total density the lower the PCE. The defect density was varied from $1 \times 10^7 \text{ cm}^{-2}$ to $1 \times 10^{18} \text{ cm}^{-2}$, cell parameters, the efficiency (PCE), voltage open circuit (V_{oc}), current short circuit (J_{sc}) and fill factor (FF) are significantly reduced [20]. The defect density parameters are illustrated in graph in Figure 4.

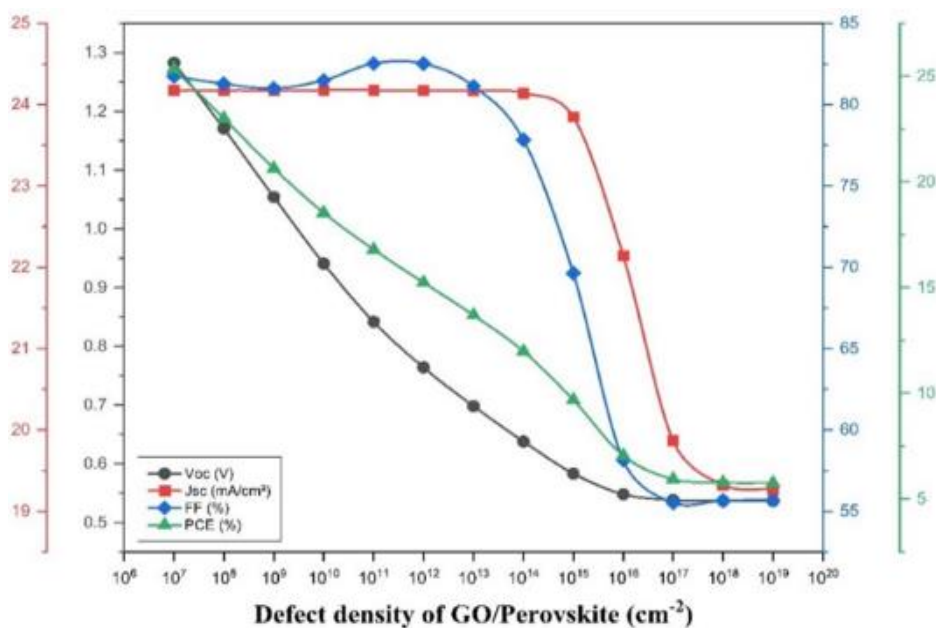


Fig. 4. The variation of defect density of GO/Perovskite on PCE, FF, J_{sc} and V_{oc}

The influence of defect density of GO/Perovskite layer on the IPSC has been changed from $1 \times 10^7 \text{ cm}^{-2}$ to $1 \times 10^{19} \text{ cm}^{-2}$ while the other variables were kept constant. It is seen in Figure 4, the increase in the defect density results in a slight decrease in the PCE, FF, J_{sc} and V_{oc} . It can also be noticed that the PCE decrease from maximum efficiency by about 77.270% and FF decreased from 81.729% by around 31.880% and J_{sc} decrement was less than 20.300% while V_{oc} decreased by around 58.130%.

The defect at the interfaces also can be called as recombination centers and it can affect the recombination process. If total density in interface GO/perovskites increased at the two interfaces, it will cause trapping and recombination which is it can reduce the performance of PSC. For PSC to obtain a good result, the defect density of GO/Perovskite must be less than or equal to $1 \times 10^{12} \text{ cm}^{-2}$. The interface of GO/Perovskite influence the performance of IPSCs compared to interface between Perovskite/ TiO_2 [20]. The reaction between GO with perovskite surface can enhanced surface coverage and caused the films smoothness with fewer pinholes [32,33]. For an example, with using GO as an amphiphilic modifier, the photovoltaic performance of PSCs can be improved, and it can enhance the interface contact between perovskite and the hole transport layer [34]. Solar cell performance decreases slightly due to the interface showing as defects when the defect density is varied over the test range.

3.3 Analysis of Capture Cross-Section of Electrons

Analyzed the value of capture cross-sections of electrons because it affects IPSC efficiency. Firstly, the value of capture cross-sections of electrons has been varied from $1 \times 10^{-19} \text{ cm}^2$ to $1 \times 10^{-5} \text{ cm}^2$. The capture cross-sections represent the probability of the trap capturing the free carrier. As shown in Figure 5, if the value of capture cross-sections of electrons less than $1 \times 10^{-9} \text{ cm}^2$ is almost constant whereas, if the value of capture cross-section increases from $1 \times 10^{-19} \text{ cm}^2$ and above, the IPSC efficiency will decrease slightly from 18.532% to 9.121% where FF slightly decrease to $1 \times 10^{-19} \text{ cm}^2$, however it was drastically reduced from 81.481% to 50.162%. The degradation of J_{sc} and V_{oc} is $23.567 \text{ mA cm}^{-2}$ and 0.772V respectively.

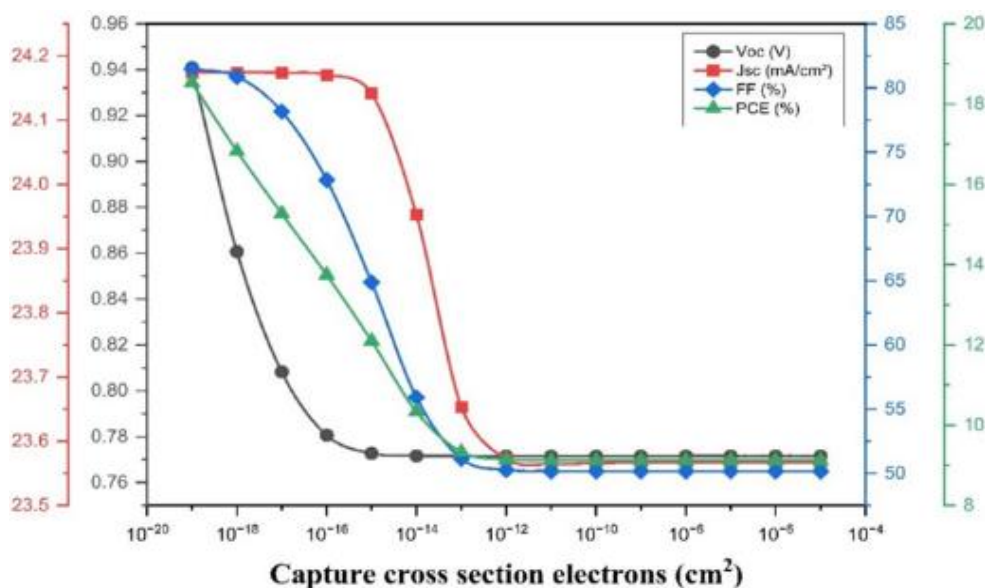


Fig. 5. The variation of capture cross-sections of electrons on PCE, FF, J_{sc} and V_{oc}

3.4 Analysis of Capture Cross-Section of Holes

The capture cross-section of holes values under $1 \times 10^{-19} \text{ cm}^2$, the efficiency drops 18.532% to 18.509%. This is because the most significant processes in defect-assisted recombination losses. Defect with negatively charged will captured by holes due to the Coulomb interactions [35]. From Figure 6, increasing the value of capture cross-section influenced the FF where the value of FF increased slightly up to 81.481 % to 81.909 %. This is because the value of J_{sc} is almost constant at $24.174 \text{ mA cm}^{-2}$ compared to value of V_{oc} drops slightly from 0.941 V to 0.935 V.

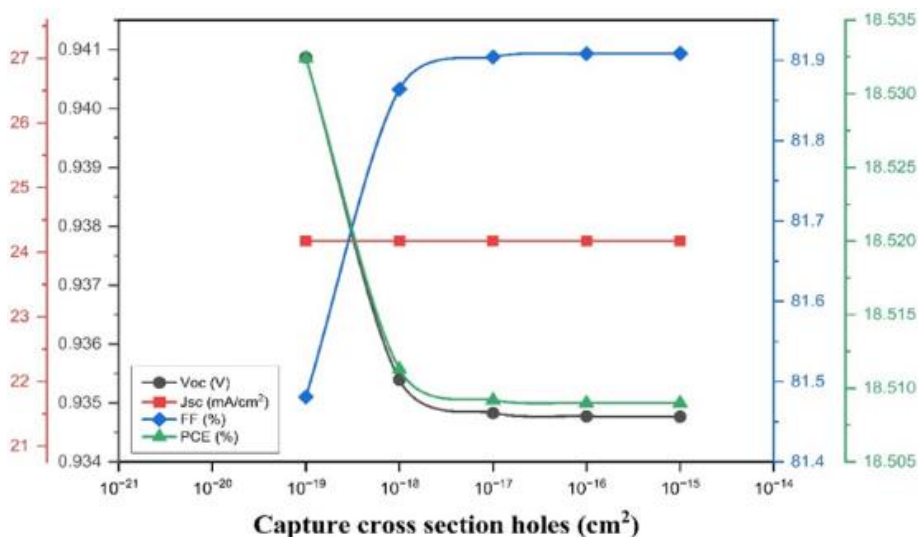


Fig. 6. The variation of capture cross-sections of holes on PCE, FF, J_{sc} and V_{oc}

3.5 Analysis of Working Temperature

Based on Figure 7, it can be analyzed that as the working temperature increase, the efficiency of the solar cell will decrease from 18.532% to 17.465%. This is because up to 300 K and above freely moving electrons will move so rapidly. When free electrons move so rapidly, it will not pass through into PSC and current cannot flow. The benchmark between 300 K to 390 K can reduce the number of PCE, FF and V_{oc} . Meanwhile, the J_{sc} increase from $24.174 \text{ mA cm}^{-2}$ to $24.186 \text{ mA cm}^{-2}$ due to the increase in temperature.

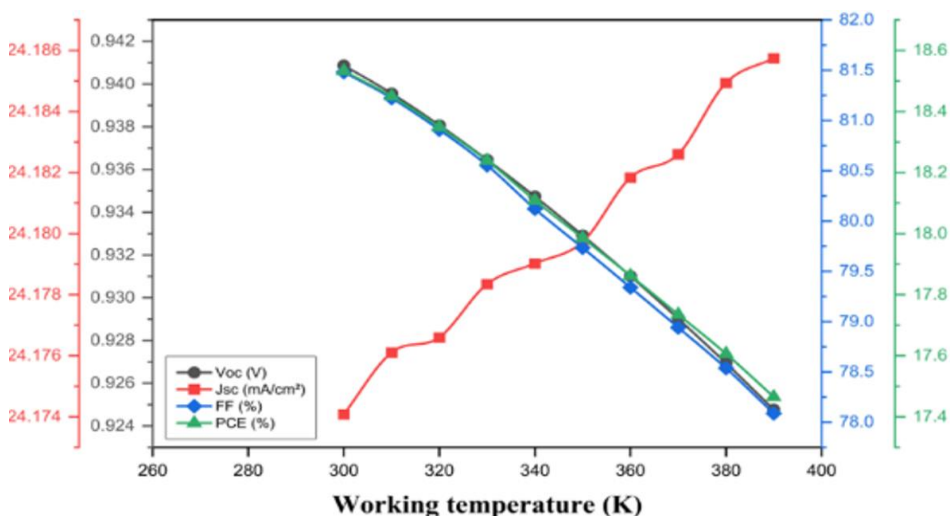


Fig. 7. The variation of working temperature of PCE, FF, J_{sc} and V_{oc} of IPSC

For instance, the increase in temperature has an impact on J_{sc} to increase. In general, the reduction in PCE is from the increase of probabilities of recombination [36]. Due to the considerable energy absorption by the electrons at high temperatures, the performance of solar cells decreases as the temperature rises. A state of instability will then be reached by the electron. As a result, the rate of recombination before reaching the depletion region will rise and the efficiency of a solar cell would decrease. Hence, the optimum working temperature of the device for IPSC with GO as HTL is 300 K where the efficiency of IPSC that has been achieved is 18.532%.

3.6 Multiple Optimization

Based on individual results from PCE, FF, J_{sc} and V_{oc} , the average performance analysis from each parameter was recorded. Each parameter shows the optimum level was chosen because of the higher S/N ratio. The percent effect on the S/N ratio indicates the dominant factor to the process parameter. From Table 2, it shows the multiple optimization results.

Table 2

The level obtained for multiple optimizations in IPSC devices

Parameters	A	B	C	D	Average
PCE (%)	1 (0%)	1 (80%)	1 (0%)	1 (19%)	23.407
FF (%)	1 (24%)	3 (36%)	3 (32%)	1 (8%)	81.781
J_{sc} (mA/cm ²)	1 (100%)	1 (0%)	1 (0%)	1 (0%)	24.571
V_{oc} (V)	3(0%)	1 (8%)	1 (1%)	1 (16%)	1.171
Multiple optimization	1	1	1	1	

From Table 3, it shows each parameter that has been used before and after optimization. The GO thickness value after optimization is 80 nm thinner than before optimization due to the thicker HTL, the series resistance, R_s increases and the transfer of holes to the electrode becomes more difficult. Next, to obtain a higher PCE in IPSC devices above 25%, the GO/Perovskite defect density value should be considered. The cross-sectional capture of electrons and holes does not require any changes as $1 \times 10^{-19} \text{ cm}^2$ is commonly used from other research [20].

Table 3

Parameter before and after optimization

Parameters	Initial IPSCs	Optimized IPSCs
Thickness of GO (nm)	100	80
Defect Density of GO/Perovskite	1×10^{10}	1×10^8
Capture cross section electrons (cm ²)	1×10^{-19}	1×10^{-19}
Capture cross section holes (cm ²)	1×10^{-19}	1×10^{-19}

As illustrated from Table 4 and Figure 8, the results obtained after optimization were improved compared to before optimization. The PCE has achieved the optimum efficiency which is at 23.41%. Furthermore, the J_{sc} and V_{oc} were improved after optimization to 24.571 mA/cm² and 1.172 V respectively. This is because modification of process parameters can increase quality, and the optimal process parameters found using the Taguchi method are insensitive to ambient variables and other noise factors [30,31]. Fill factor, FF were slightly decrease from 81.48% to 81.295% due to the certain control factors have been selected according to the PCE. In terms of the environment, a solar cell is well-known as the cleanest energy source. In contrast to fossil-fuel-generated electricity, a solar cell is very effective at maximizing the production of electricity and minimizing carbon emissions.

Table 4
 Comparison of values obtained before and after optimization

Parameters	Before optimization	After optimization	
		Individual	Multiple
PCE (%)	18.530	23.407	23.408
FF (%)	81.480	81.782	81.295
J_{sc} (mA/cm ²)	24.174	24.572	24.571
V_{oc} (V)	0.941	1.171	1.172

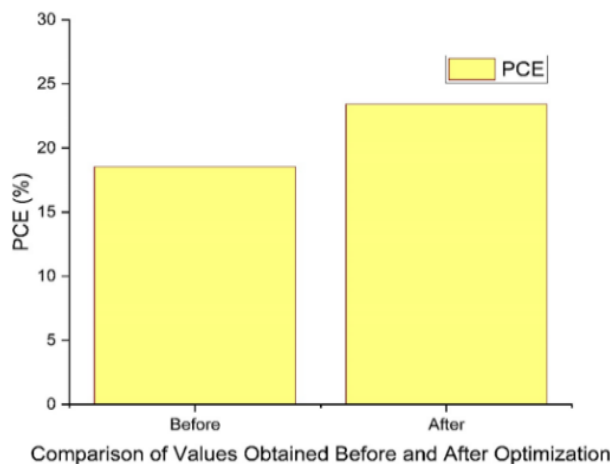


Fig. 8. Comparison of values obtained before and after optimization

4. Conclusions

The simulation was implemented by employing a complete simulated device structure composed of FTO/GO/ CH₃NH₃PbI₃/ TiO₂/Ag. Besides, the analysis of this project which is to optimize GO as HTL also was successfully performed. Several key parameters of HTL have been analyzed to obtain the optimum performance for IPSC as well as the influence of back contact. The simulation results showed that GO as HTL in IPSC has produced an efficiency 18.53% compared to previous researcher of methylammonium lead triiodide perovskite solar cell (PSC) containing graphene oxide (GO) as HTL has achieved an optimal PCE of 16.51% using SCAPS-1D simulation. In additionally, after optimization using Taguchi Method L₉ OA, the efficiency increased to 23.408 %. This is shows that the optimum solution in achieving the desired efficiency in IPSC devices was successfully predicted by using Taguchi Method. Overall, the project was a success. The efficiency can be improved by using GO as HTL in IPSC devices and optimizing it using Taguchi Method.

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