Performance Analysis of Zinc Cobaltite ($ZnCO_2O_4$) As A Hole Transport Layer (HTL) For Perovskite Solar Cell Using OghmaNano Software and Taguchi Method Optimization.

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Abstract. Perovskite solar cells (PSCs) are cost-effective and efficient photovoltaic cells that show great potential as an alternative to silicon solar cells. They possess desirable properties such as high mobility, direct bandgap, long carrier lifetime, and strong light absorption. However, the traditional materials used for the holes transport layer (HTL) in PSCs, such as PEDOT:PSS, SPIRO-OMETAD, and copper(I) iodide, have durability issues and lower carrier mobility. To overcome these challenges, Zinc Cobaltite ($ZnCO_2O_4$) with its advantages of hole transport, wide optical bandgap, and solution processability was investigated as a potential alternative HTL material. Through simulations using OghmaNano software and the Taguchi method, the device structure FTO/TiO_2/$CsPbI_3$/$ZnCO_2O_4$/Au was analyzed, and the performance was optimized by varying the thickness of the $ZnCO_2O_4$ layer. The simulation results showed a power conversion efficiency (PCE) of 32.23% with a ZnCO2O4 thickness of 300nm. ANOVA analysis revealed that the $ZnCO_2O_4$ thickness as the HTL had the most significant influence on PCE, followed by environmental temperature and the bandgap of $ZnCO_2O_4$. In particular, the $ZnCO_2O_4$ thickness had a substantial 70% impact on PCE, indicating that adjusting the thickness of $ZnCO_2O_4$ could lead to corresponding improvements in PCE.

1. Introduction

Solar cells, which are also called photovoltaic cells, operate by transforming solar energy into electrical energy. The unique characteristics of the semiconductor materials typically silicon used to create solar cells enable this method to be performed. Solar cells are made up of layers. The solar cell is made up of a p-n junction, which is a boundary between two kinds of semiconductor material with distinct electrical characteristics. The semiconductor atoms in the cell receive photons from the sunshine, which causes the atoms to discharge electrons. A voltage differential occurs across the cell as a result of the p-n junction's separation of the electrons and holes. The electrons are gathered and transported as an electrical current by electrical connections on the top and bottom of the cell. Electrical equipment can be powered by electricity right away, or it can be saved in batteries for later use.

Nowadays, the third generation of solar cells has been proven to be the future method to generate electricity using solar power. Due to their high PCE and suitability for scalable procedures, perovskite solar cells, a new 3rd generation solar cell, are featured in this newsletter and look to have a very strong possibility of helping to scale up solar energy production [1]. In no other period in the history of solar cell research has the PCE been increased at such a quick rate as it has been for perovskite solar cells, hence these solar cells merit discussion. Perovskites are considered as ideal photovoltaic materials in solar cells due to their high absorption in the visible spectrum, long carrier diffusion length, high carrier mobility, low exciton binding energy, tunable bandgaps by exchanging atomic composition, large area production and low cost owing to solution process-ability [2].

The primary role of the hole transport layer is to gather and move holes from the perovskite light-absorbing layer in order to work in tandem with the electron transport layer to facilitate the separation of the electron-hole pairs in the perovskite materials [3]. In hole transport materials, the highest occupied molecular orbit (HOMO) must match the valence band of perovskite materials for hole transport. According to the chemical composition, hole transport materials in perovskite solar cells can be divided into two types which is organic and inorganic hole transport materials [4]. Spiro-OMeTAD is the most used organic hole transport material, which shows good penetration in nanoscale perovskite and is a good match with the valence band energy of perovskite, although its hole mobility is not as high as that of other organic hole transport materials [5], [6]. Inorganic metal oxides with the merits of high carrier transport capability, low cost and superior chemical stability have largely served as the hole transport layer (HTL) in perovskite solar cells (PSCs) in recent years [2], [7]. Among them, ternary metal oxides have gradually attracted attention because of the wide tenability of the two inequivalent cations in the lattice sites that offer interesting physicochemical properties.

In this work, Zinc Cobaltite were simulated as hole transfer layer (HTM) in perovskite solar cells with varying thickness via OghmaNano software. This software was used in this work because of its capability to address device equations in both steady-state and time domains, as well as in one-dimensional (1D) or two-dimensional (2D) spatial contexts. Notably, the model used in this software adeptly resolves the electron and hole drift-diffusion equations alongside the carrier continuity equations within the spatial domain, providing a comprehensive depiction of charge movement within the device. Additionally, the thickness optimization was optimized by Taguchi method which facilitates the identification of critical factors affecting the performance of the device.

2. Device Simulation

2.1. OghmaNano Software

OghmaNano (Organic and Hybrid Material Nano Simulation Tool) is a software package that is used for simulating the performance of various types of solar cells and other optoelectronic devices. OghmaNano allows the user to model the behavior of the device under different operating conditions, and to simulate the performance of the device under a wide range of solar radiation levels, temperatures, and other environmental conditions. The software uses “Poisson equation (1), the bipolar drift diffusion equations (2,3), and the carrier continuity equations (4,5) as mentioned in [8], [9]. In addition, OghmaNano use finite element-method (FEM) for mathematical solver [10]. The AM1.5G was chosen as spectrum of the sun's spectrum.

$$\frac{d}{dx}\varepsilon_0\varepsilon_r\frac{d\varphi}{dx} = q(n-p) \tag{1}$$

$$J_n = q\mu_c n\frac{\partial \mathrm{E}_c}{\partial \mathrm{x}} + qD_n\frac{\partial n}{\partial x} \tag{2}$$

$$J_p = q\mu_n p\frac{\partial \mathrm{E}_v}{\partial \mathrm{x}} - qD_p\frac{\partial p}{\partial x} \tag{3}$$

$$\frac{\partial J_n}{\partial x} = q\left(R_n - G + \frac{\partial n}{\partial x}\right) \tag{4}$$

$$\frac{\partial J_p}{\partial x} = -q\left(R_p - G + \frac{\partial p}{\partial x}\right) \tag{5}$$

Based on equation 1 until 5, ε_0 denotes the permittivity of free space, while ε_r represents the permittivity of perovskite. The voltage profile is denoted by φ, q represents the elementary charge, p signifies the carrier density of holes, and n denotes the carrier density of electrons. The electron current density is expressed as J_n, and the hole current density is denoted as J_p. Additionally, μ_c and μ_n are utilized to denote the electron mobility and hole mobility, respectively. E_c is utilized to signify the free electron mobility edge, and E_v is employed to represent the free hole mobility edge. The free carrier generation rate is denoted by G. R_n and $R_p$represent the recombination rate of electrons and holes, respectively. The diffusion coefficients of electrons and holes are represented by D_n and D_p.

2.2. Electrical Model

The electrical simulation consists of 5 distinct layers which consist of FTO glass, ETL, perovskite, HTM and gold contact the layers are FTO/TiO_2 / $CsPbI_3$/ $ZnCO_2O_4/Au$. Figure 1 shows the simulated configuration of the device. Figure 1a show the schematic configuration of device which sun light will go to first layer of FTO, then to layer of $TiO_2, CsPbI_3, ZnCO_2O_4, Au$. Figure 1b shows the energy level diagram of whole diagram [2], [11], [12]. As shown in Figure 1b, there shown seemingly paradoxical situation where the conduction band of CsPbI3 is smaller than TiO2 and the Fermi energy of FTO results in a Type II band alignment, which could generate a barrier at the interface between CsPbI3/TiO2. An inherent electric field created by this alignment makes it easier to extract, transport, and separate charge carriers efficiently. The potential barrier functions as a selective contact despite the energy step-up, encouraging unidirectional charge flow and lowering recombination, which eventually leads to improved photocurrent and higher overall efficiency in the solar device. The distinct alignment of energy levels and the consequent electric field are essential factors in maximising the efficiency of the solar cell. The device is simulated with various of thickness of $ZnCO_2O_4$ to 50nm, 75nm, 100nm, 125nm, 150nm, 175nm, 200nm, 225nm, 250nm, 275nm, 300nm, 325nm, 350nm, 375nm, 400nm, 425nm, 450nm, 475nm and also 500nm. Table 1 show the electrical parameter used to simulate the device.

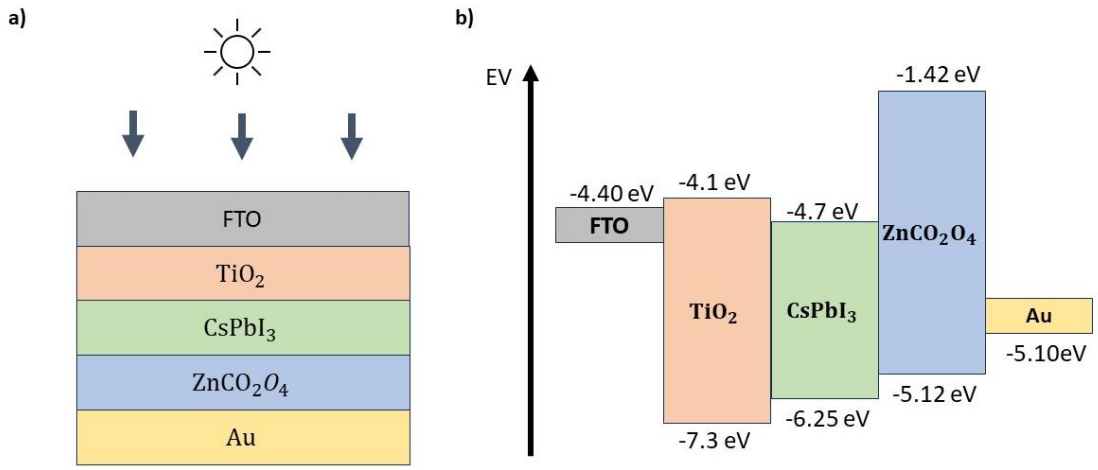

Figure 1. The simulated configuration of device. (a) The schematic configuration of device. (b) the energy level diagram of whole device.

Table 1. Electrical parameter

Parameter	TiO_2(ETL)	$CsPbI_3$(Perovskite)	$ZnCO_2O_4$ (HTL)
Eg (eV)	3.2	1.694	2.0
Xi (eV)	4.0	3.95	3.7
Relative permittivity	9	6	5
Electron mobility (m^2/Vs)	0.002	0.0025	0.00000914
Hole mobility (m^2/Vs)	0.001	0.0025	0.00000914

2.3. Taguchi Method

Taguchi's techniques have been used widely in engineering design [13].To produce a reliable process and outcome for the best product quality, the Taguchi approach includes system design, parameter design, and tolerance design techniques [14]. The key component of Taguchi's methods is the application of parameter design, an engineering method for product or process design that focuses on identifying the parameter (factor) settings that produce the best levels of a quality characteristic (performance measure) with the least amount of variation.

For optimization using the Taguchi method, 3 control factors have been used to be the determining factors in this study. The three factors are the thickness of Zinc Cobaltite as a hole transport layer, the bandgap of Zinc Cobaltite and the temperature of the environment. The Zinc Cobaltite thickness values that have been used are 100nm, 225nm and 300nm while the values for the ambient temperature are 300K, 320K and 340K. For the Zinc Cobaltite bandgap values of 1.8, 2.0 and 3.7 have been used. Table 2 and 3 shows the parameter used for the Taguchi method.

Table 2. Parameter for the control factors.

Control factors \ Levels	1	*2*	3
Thickness (nm)	100	225	300
Temperature (K)	300	320	340
Bandgap (Eg)	1.8	2.00	3.70

Table 3. Parameter for noise factors.

	Noise level			Number of levels
Noise factors \ levels	1	2	3	
Electron mobility	0.00000710	0.00000810	0.00000914	3
Hole mobility	0.00000710	0.00000810	0.00000914	3

3. Result

3.1. Effect of zinc cobaltite thickness on perovskite solar cell performance parameter.

The transport layer thickness in perovskite solar cells is crucial, as it directly affects hole extraction from the perovskite layer. An optimized thickness ensures minimal charge recombination losses and effective charge transfer. If the transport layer is too thin, hole extraction may be insufficient, leading to lower photo current and overall device performance. Conversely, an excessively thick transport layer can hinder charge extraction, resulting in higher recombination rates and reduced device efficiency. The ideal thickness for the hole transport layer depends on factors like charge extraction, conductivity, and contact resistance, and experimental optimization is often necessary to determine the best thickness for a specific perovskite solar cell design.

Table 4. PSC performance with various hole transport layer thickness.

Thickness (nm)	Voc (V)	Jsc(mA/cm^2)	FF (%)	PCE (%)
50	1.406	-29.770	69.8	29.25
75	1.384	-29.023	70.9	28.50
100	1.373	-26.701	71.6	26.28
125	1.363	-26.829	72.1	26.37
150	1.407	-31.661	69.7	31.04
175	1.415	-30.431	69.3	29.89
200	1.384	-28.763	71.0	28.25
225	1.371	-28.553	71.5	28.02
250	1.366	-26.024	72.0	25.61
275	1.372	-27.355	71.5	26.83
300	1.427	-32.887	68.7	32.23
325	1.411	-32.385	69.4	31.73
350	1.394	-31.741	70.2	31.09
375	1.399	-29.369	70.2	28.86
400	1.382	-28.720	71.0	28.21
425	1.369	-28.303	71.7	27.79
450	1.373	-25.649	71.7	25.27
475	1.358	-25.101	72.5	24.72
500	1.314	-11.482	73.5	11.09

According to the data in Table 4 and the study's overall results, it is evident that a Zinc Cobaltite thickness of 300nm yields the highest PCE of 32.23% for the perovskite solar cell. However, when the thickness exceeds 300nm, the PCE starts to decline, and at 500nm thickness, it can only achieve a PCE of 11.09%. Thus, it's clear that the optimal thickness is 300nm for the best PCE performance. Figure 2a illustrates the change in open circuit voltage (Voc) and current density (Jsc) in Figure 2b with varying thickness. Meanwhile, Figure 3a depicts the efficiency of perovskite solar cells (PCE) at different thicknesses of the hole transport layer composed of Zinc Cobaltite. The thicknesses of the perovskite and electron transport layer (ETL) are set at 400 nm and 200 nm, respectively. The PCE exhibits periodic increases and decreases in response to alterations in the hole transport layer thickness. Optimal photoconversion is achieved with a hole transport layer thickness of 300 nm, as indicated in Figure 3a. Furthermore, Figure 3b illustrates the fluctuation of fill factor (FF) with thickness.

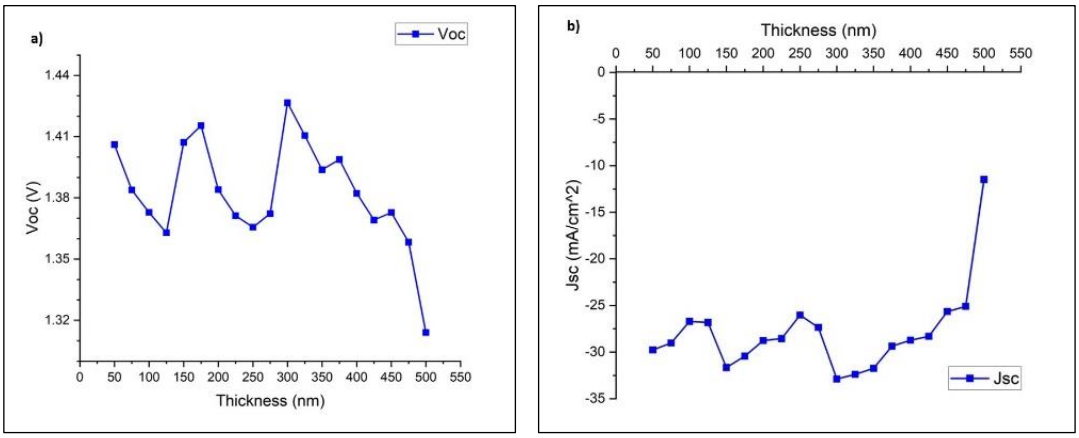

Figure 2. (a) Variation of Voc over thickness. (b) Variation current density, Jsc over thickness.

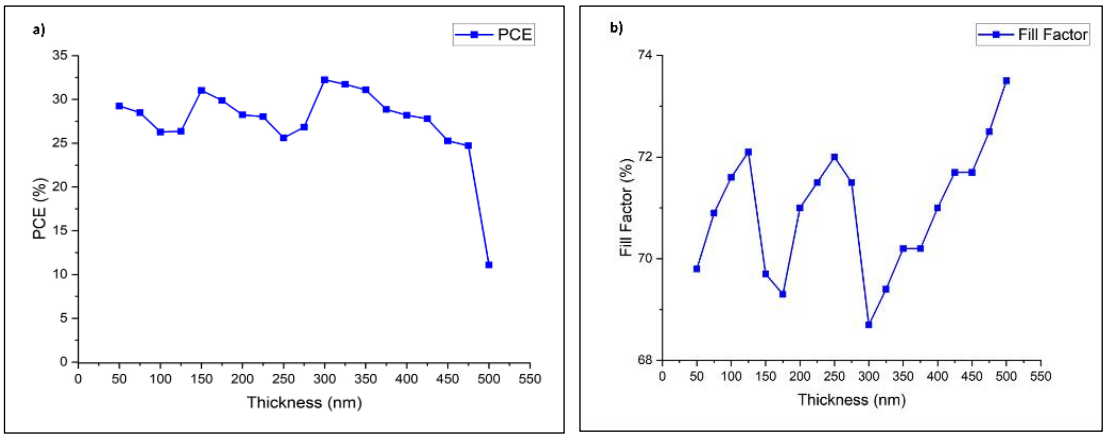

Figure 3. (a) Variation of PCE over thickness. (b) Variation of FF over thickness.

3.2. Taguchi method result

In this study, two optimization methods were used to improve the power conversion efficiency (PCE) of perovskite. The second method employed the Taguchi method, aiming to identify the key factors influencing PCE. Three control factors were considered: thickness of Zinc Cobaltite as the hole transport layer (HTL), ambient temperature, and the bandgap of Zinc Cobaltite. Additionally, two noise factors, hole mobility and electron mobility for Zinc Cobaltite, were included in the study, although they were expected to have a minor impact on PCE.

Table 5. PSC performance with various hole transport layer thickness.

PCE (%)	1	2	3	4	5	6	7	8	9
1	26.25	26.25	26.25	26.25	26.25	26.25	26.25	26.25	26.25
2	25.15	25.15	25.15	25.15	25.15	25.15	25.15	25.15	25.15
3	20.40	20.60	20.76	20.40	20.60	20.76	20.40	20.60	20.76
4	28.02	28.02	28.02	28.02	28.02	28.02	28.02	28.02	28.02
5	26.77	26.77	26.78	26.77	26.77	26.78	26.77	26.77	26.78
6	25.60	25.60	25.60	25.60	25.60	25.60	25.60	25.60	25.60
7	32.18	32.18	32.18	32.18	32.18	32.18	32.18	32.18	32.18
8	30.83	30.83	30.83	30.83	30.83	30.83	30.83	30.83	30.83
9	29.49	29.49	29.49	29.49	29.49	29.49	29.49	29.49	29.49

Table 6. Fill factor,FF result by using Taguchi method.

FF (%)	1	2	3	4	5	6	7	8	9
1	71.5	71.5	71.5	71.5	71.5	71.5	71.5	71.5	71.5
2	84.1	84.1	84.1	84.1	84.1	84.1	84.1	84.1	84.1
3	71.8	72.5	73.1	71.8	72.5	73.1	71.8	72.5	73.1
4	71.5	71.5	71.5	71.5	71.5	71.5	71.5	71.5	71.5
5	83.7	83.7	83.7	83.7	83.7	83.7	83.7	83.7	83.7
6	84.4	84.4	84.4	84.4	84.4	84.4	84.4	84.4	84.4
7	68.5	68.5	68.5	68.5	68.5	68.5	68.5	68.5	68.5
8	82.7	82.7	82.7	82.7	82.7	82.7	82.7	82.7	82.7
9	84.1	84.1	84.1	84.1	84.1	84.1	84.1	84.1	84.1

Table 7. Open circuit voltage, Voc result by using Taguchi method.

Voc (V)	1	2	3	4	5	6	7	8	9
1	1.373	1.373	1.373	1.373	1.373	1.373	1.373	1.373	1.373
2	1.120	1.120	1.120	1.120	1.120	1.120	1.120	1.120	1.120
3	1.063	1.063	1.063	1.063	1.063	1.063	1.063	1.063	1.063
4	1.371	1.371	1.371	1.371	1.371	1.371	1.371	1.371	1.371
5	1.119	1.119	1.119	1.119	1.119	1.119	1.119	1.119	1.119
6	1.062	1.062	1.062	1.062	1.062	1.062	1.062	1.062	1.062
7	1.425	1.426	1.426	1.425	1.426	1.426	1.425	1.426	1.426
8	1.133	1.133	1.133	1.133	1.133	1.133	1.133	1.133	1.133
9	1.065	1.065	1.065	1.065	1.065	1.065	1.065	1.065	1.065

Table 8. Current density, Jsc result by using Taguchi method.

Jsc (mA/cm^2)	1	2	3	4	5	6	7	8	9
1	-26.707	-26.707	-26.707	-26.707	-26.707	-26.707	-26.707	-26.707	-26.707
2	-26.709	-26.709	-26.709	-26.709	-26.709	-26.709	-26.709	-26.709	-26.709
3	-26.721	-26.721	-26.721	-26.721	-26.721	-26.721	-26.721	-26.721	-26.721
4	-28.553	-28.553	-28.553	-28.553	-28.553	-28.553	-28.553	-28.553	-28.553
5	-28.580	-28.580	-28.580	-28.580	-28.580	-28.580	-28.580	-28.580	-28.580
6	-28.549	-28.549	-28.549	-28.549	-28.549	-28.549	-28.549	-28.549	-28.549
7	-32.922	-32.921	-32.921	-32.921	-32.921	-32.921	-32.921	-32.921	-32.921
8	-32.883	-32.883	-32.883	-32.883	-32.883	-32.883	-32.883	-32.883	-32.883
9	-32.886	-32.886	-32.886	-32.886	-32.886	-32.886	-32.886	-32.886	-32.886

After conducting numerous simulations using the Taguchi method (refer to Table 5, 6, 7 and 8), it's evident that various PCE values are observed, indicating that all factors influence PCE, albeit to varying degrees. Notably, the thickness of Zinc Cobaltite as HTL emerges as a major factor affecting PCE in the study. Figure 4 shows the factor plot effect for PCE using Taguchi method and result of ANOVA for simulated PCE in Table 9.

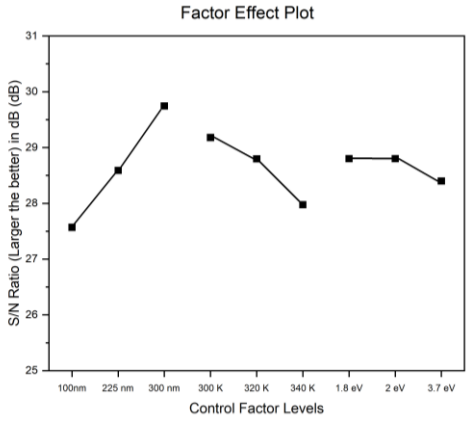

Figure 4. Factor plot effect for PCE using Taguchi method.

Table 9. Result of ANOVA for PCE.

Factor	Degrees of freedom	Sun of squares	Mean Square	Factor Effect (%)
Thickness (nm)	2	7	4	70
Temperature (K)	2	2	1	22
Bandgap (Eg)	2	0	0	4

After analysing using ANOVA, it has been identified that the thickness of Zinc Cobaltite as HTL is the factor that has the biggest impact on PCE followed by the environmental temperature and also the bandgap of Zinc Cobaltite. The thickness of Zinc Cobaltite has a 70% effect on PCE and therefore if the thickness of Zinc Cobaltite is changed then PCE will also change. Table 10 shows the best setting selection for respective parameter.

Table 10. Best setting selection for respective parameter.

Control factor parameter	Best level	Best value
Thickness (nm)	3	300
Temperature (K)	1	300
Bandgap (Eg)	2	2.0

After identifying the main factor, confirmation experiments were conducted using the best parameters for each control factor. The optimal parameters are thickness of Zinc Cobaltite as HTL at 300nm (level 3), ambient temperature at 300K (level 1), and bandgap at 2.0 (level 2). These parameters were combined to assess their impact on achieving a high PCE. Table 11 shows result after the confirmation experiment.

Table 11. Result after the confirmation experiment.

	Voc (V)	Jsc(mA/cm^2)	FF (%)	PCE (%)
1	1.426	-32.887	68.7	32.23
2	1.426	-32.886	68.7	32.23
3	1.427	-32.886	68.7	32.23
4	1.426	-32.887	68.7	32.23
5	1.426	-32.886	68.7	32.23
6	1.427	-32.886	68.7	32.23
7	1.426	-32.887	68.7	32.23
8	1.426	-32.886	68.7	32.23
9	1.427	-32.886	68.7	32.23

After conducting the confirmation experiment, the data in the table shows that all experiments resulted in a high PCE, indicating that the chosen parameters are effective for achieving high PCE values. The highest PCE achieved was 32.23%, corresponding to the following parameters: Zinc Cobaltite thickness as HTL at 300nm (level 9), ambient temperature at 300K, Zinc Cobaltite bandgap at 2.0 (level 9), and noise factor levels 3 for both hole and electron mobility (0.00000914). These findings demonstrate the successful combination of parameters for obtaining a high PCE value.

4. Conclusion

As a result of the simulation, a PCE of 32.23% was obtained with a Zinc Cobaltite (ZnCo2O4) thickness of 300nm. ANOVA analysis revealed that the thickness of Zinc Cobaltite as the hole transport layer (HTL) has the most significant influence on the perovskite solar cell efficiency (PCE), followed by environmental temperature and the bandgap of Zinc Cobaltite. Specifically, the thickness of Zinc Cobaltite has a significant 70% impact on PCE, indicating that altering the thickness of Zinc Cobaltite will lead to corresponding changes in PCE.

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