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<https://doi.org/10.5109/5909089>

出版情報 : Proceedings of International Exchange and Innovation Conference on Engineering & Sciences (IEICES). 8, pp.183-188, 2022-10-20. Interdisciplinary Graduate School of Engineering Sciences, Kyushu University

バージョン :

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Numerical Simulation of Highly Efficient Lead-free MAGeI_3 Based Perovskite Solar Cell for Various ETL and HTL Layers

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Abstract: *The eco-friendly and highly stable lead-free MAGeI_3 perovskite solar cell has proved itself as a potential candidate for conventional lead-based perovskite solar cells with high efficiency. In this paper, numerical simulation has been performed over MAGeI_3 solar cell with three different Electron Transport Layer (ETL) and Hole Transport Layer (HTL) materials to obtain the best combination of ETL/perovskite/HTL layers using the solar cell simulation tool SCAPS-1D. The best performance has been found for the $\text{ITO}/\text{ZnO}/\text{MAGeI}_3/\text{NiO}_x$ structure with an efficiency of 21.19% (V_{oc} of 1.89 V, J_{sc} of 16.11 mA/cm^2 , and FF of 69.57%) due to the higher bandgap and better carrier mobility of these two ETL and HTL materials. The thickness optimization indicated that a thickness below 1000 nm is suitable for better performance which also leads to a good indication of making a lightweight solar cell. This study indicates that the use of low-cost and environment-friendly ZnO and NiO_x with non-toxic MAGeI_3 perovskite has the potential to obtain a high-efficiency eco-friendly solar cell.*

Keywords: MAGeI_3 perovskite solar cell; Electron Transport Layer (ETL); Hole Transport Layer (HTL); SCAPS-1D.

1. INTRODUCTION

The global demand for renewable and green energy for power generation in the last few decades has developed a vast field of research for new inventions with low-cost and highly efficient photovoltaic devices. The perovskite-based photovoltaic cells have also gotten the attention of researchers due to their favorable inherent characteristics like tunable direct bandgap, higher absorption coefficient, low exciton binding energy, and higher carrier mobility as well as easier and cost-effective fabrication process [1-4]. The first perovskite solar cell was introduced by Kojima et.al. in 2009 using Iodine (I^-) as the halide material which only had a conversion efficiency of 3.81% [5]. But after a decade it has advanced to 25.8% by a group of researchers who introduced an interlayer between the perovskite FAPbI_3 and charge transporting layer to reduce the defects [6]. Despite offering higher efficiency, the Lead (Pb)-based perovskite is still facing issues in the case of using the device commercially due to the toxicity of Pb leading to some technical and environmental issues [7]. So, it is necessary to find alternatives to Pb to make a highly efficient environment-friendly solar cell. Non-toxic metals such as tin (Sn), bismuth (Bi), and germanium (Ge) can work as alternatives [7-9]. Though having the possibility to be an alternative to Pb ion, both Sn^{2+} and Bi^{3+} undergo instability at the time of inclusion with the other ions of perovskite material [10]. Ge doesn't face this kind of problem which leads it to be a good candidate for the replacement of lead from the perovskite material. Ge-based absorber layer (Methylammonium Germanium tri-iodide, MAGeI_3) can provide more stability than the lead-based counterpart as the absorber layer (Methylammonium Lead tri-Iodide, MAPbI_3) in a perovskite solar cell due to the use of less degradable Ge instead of Pb [11]. Ge can provide higher stability at higher temperatures up to 150 $^{\circ}\text{C}$, making it more suitable for the fabrication of the solar cell [11]. The excellent electron and hole transport nature, and suitable

absorption in the range of 350 nm-650 nm due to the narrower bandgap of MAGeI_3 have made it a potential competitor of MAPbI_3 to make a highly efficient eco-friendly solar cell [12]. For this reason, MAGeI_3 has been chosen in this paper as the active layer material of the solar cell.

For obtaining a highly efficient perovskite solar cell, one of the major requirements is to optimize the charge transport layers (CTLs). The efficiency of the cell highly depends on the carrier mobilities of the CTL and its band alignment with the perovskite absorber. Efficient transmission of both electrons and holes throughout the device is crucial for a high-performance solar cell which can be obtained by making a good combination of ETL (Electron Transport Layer) and HTL (Hole Transport Layer) material selection. Many researchers have tried to optimize the MAGeI_3 solar cell's performance with different ETL and HTL combinations [13-14]. The most used ETL material is TiO_2 , but it suffers from lower - carrier mobility and less stability under UV illumination [15]. Other materials like ZnO, IGZO, PCBM, etc. have also been used as the ETL material [13]. For the case of HTL material, the use of D-PBTTT-14 in MAGeI_3 showed an efficiency of greater than 21% [14]. Other materials like P3HT, CuSCN, Spiro-OMeTAD, Cu_2O , etc. have also been adopted as the HTL material of MAGeI_3 perovskite solar cell [13-14].

Taking the above issues in mind, in this work, we tried to investigate the effect of different ETLs and HTLs on the performance of MAGeI_3 solar cells using the solar cell simulation software SCAPS-1D (Solar Cell Capacitance Simulator). We used three different materials ZnO, WO_3 , and SnO_2 as ETL while using NiO_x , CuI, and PEDOT:PSS as the HTL and tried to optimize the perovskite absorber layers' thickness for the best combination of ETL and HTL. The solar cell parameters open-circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF) and efficiency (η) has been calculated for optimizing the best-performing solar cell.

2. NUMERICAL MODELLING AND DEVICE STRUCTURE

The device structure used for the numerical study of the MAgel₃ perovskite solar cell in this paper is represented in Fig. 1. The structure consisted of a hole transport layer just above the glass substrate at the very bottom and is used for blocking the electrons and transporting the holes over a glass substrate that is followed by the MAgel₃ perovskite layer where most of the photons are absorbed. The next layer to the absorber is the electron transport layer for transporting the electron and blocking the holes in the electrodes. The structure is finally completed by a transparent conducting oxide layer. The light illuminates the cell through the ITO layer under the AM1.5 solar spectrum with an incident power density of 100W/m². The layer parameters for all the layers used in the simulations have been summarized in Table 1 and Table 2 which are taken from different literature [13, 16-19].

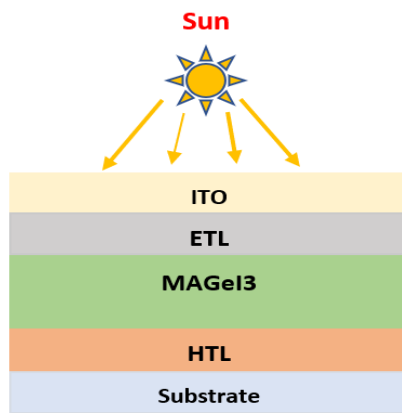


Fig. 1: The schematic of the structure of the MAgel₃ solar cell

Table 1: Layer properties of different ETL & MAgel₃

Parameters	ZnO	WO ₃	SnO ₂	MAgel ₃
E _g (eV)	3.3	2.92	3.5	1.9
χ (eV)	4.1	4.59	4	3.98
ε _r	9.0	5.76	9	10
N _c (cm ⁻³)	4×10 ¹⁸	1.96×10 ¹⁹	2.2×10 ¹⁷	1×10 ¹⁶
N _v (cm ⁻³)	1×10 ¹⁹	1.96×10 ¹⁹	2.2×10 ¹⁶	1×10 ¹⁵
μ _e (cm ² /V _s)	100	10	20	1.62×10 ¹
μ _h (cm ² /V _s)	25	10	10	1.01×10 ¹
N _D (cm ⁻³)	1×10 ¹⁸	3.68×10 ¹⁸	1×10 ¹⁷	1×10 ⁹
N _A (cm ⁻³)	0	0	0	1×10 ⁹

To obtain the values of the solar cell performance parameters, numerical solar cell simulation software SCAPS-1D has been adopted in this study. SCAPS-1D was developed at the Department of Electronics and Information System (EIS) which supports the addition of up to 7 layers for a solar cell. This software can solve the basic semiconductor equation- the Poisson equation and the continuity equations for both electrons and holes in one-dimensional steady-state conditions and measure many properties like- short-circuit current density (J_{sc}), fill factor (FF), open-circuit voltage (V_{oc}), conversion efficiency (η), quantum efficiency (QE), spectral response, generation, and recombination profile, etc.

Table 2: Layer properties of different HTL

Parameters	NiOx	CuI	PEDOT:PSS
E _g (eV)	3.75	3.5	1.6
χ (eV)	2.10	2.1	3.4
ε _r	10.70	6.5	3
N _c (cm ⁻³)	2.8×10 ¹⁹	2.8×10 ¹⁹	2.2×10 ¹⁸
N _v (cm ⁻³)	1.8×10 ¹⁹	1×10 ¹⁹	1.8×10 ¹⁹
μ _e (cm ² /V _s)	12	1.7×10 ⁻⁴	4.5×10 ⁻¹
μ _h (cm ² /V _s)	25	2×10 ⁻⁴	9.9×10 ⁻⁵
N _D (cm ⁻³)	0	0	0
N _A (cm ⁻³)	1×10 ¹⁵	1×10 ¹⁸	1×10 ¹⁶

In this work, optimization of the thickness of the MAgel₃ layer for different ETL and HTL layers have been tried to be done. During each simulation, the ETL and HTL layer thicknesses have been fixed at 50 nm while the ITO layer thickness has been maintained to be 60 nm. For the MAgel₃ layer, the thickness has been changed from 300 nm to 1500 nm with a 300 nm step thickness for finding the best thickness where the cell performs the best. Au has been used as the back contact material throughout the simulation for the optimization of perovskite layer thickness.

3. RESULT AND DISCUSSION

3.1 Effect of perovskite thickness

Solar cell efficiency largely relies on the absorbance of the solar spectrum by the absorber layer which is influenced by the thickness variation of the layer. In this section, we have observed the effect of thickness variation on the performance parameters of solar cells i.e., open-circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF), and efficiency (η) for three different ETL material- ZnO, SnO₂ and WO₃ and also for three different HTL material- NiO_x, CuI, and PEDOT: PSS. In the case of observing the variation of different ETL, NiO_x has been used as the HTL material whereas in the time of HTL variation ZnO has been used as the ETL material.

3.1.1 Simulation with different ETL

Fig. 2 shows the variation of the solar cell parameters with increasing perovskite thickness for three different ETL layers- ZnO, SnO₂, and WO₃. In all graphs, the black line represents the changes for ZnO, the blue one stands for the changes due to SnO₂, and finally, the red one is used to show the variations for the WO₃ ETL layer. It is to be noted that, for V_{oc}, FF, and efficiency curves WO₃ shows lower performance than the other two materials. This is due to their bandgap phenomena. WO₃ has a bandgap of 2.92 eV which is lower than the other two ETL materials ZnO (3.3 eV) and SnO₂ (3.5 eV). The open-circuit voltage of a solar cell is given by:

$$V_{oc} = \frac{kT}{q} \ln \left(\frac{J_{ph}}{J_0} + 1 \right) \quad (1)$$

Where kT/q defines the thermal voltage, J_{ph} and J₀ represent the light-induced current density and the diode saturation current density respectively. J₀ can be further obtained by the following equation:

$$J_0 = qn_i^2 \left(\frac{D_N}{L_N N_A} + \frac{D_P}{L_P N_D} \right) \quad (2)$$

D_N and D_P in the above equation denote the diffusion constants for electron on the p-side and holes on the n-

side of the junction correspondingly whereas, L_N and L_P stand for the electron and hole diffusion length on the p-side and n-side of the junction respectively. The net acceptor impurity concentration on the p-side and donor concentration on the n-side are denoted by N_A and N_D in the equation.

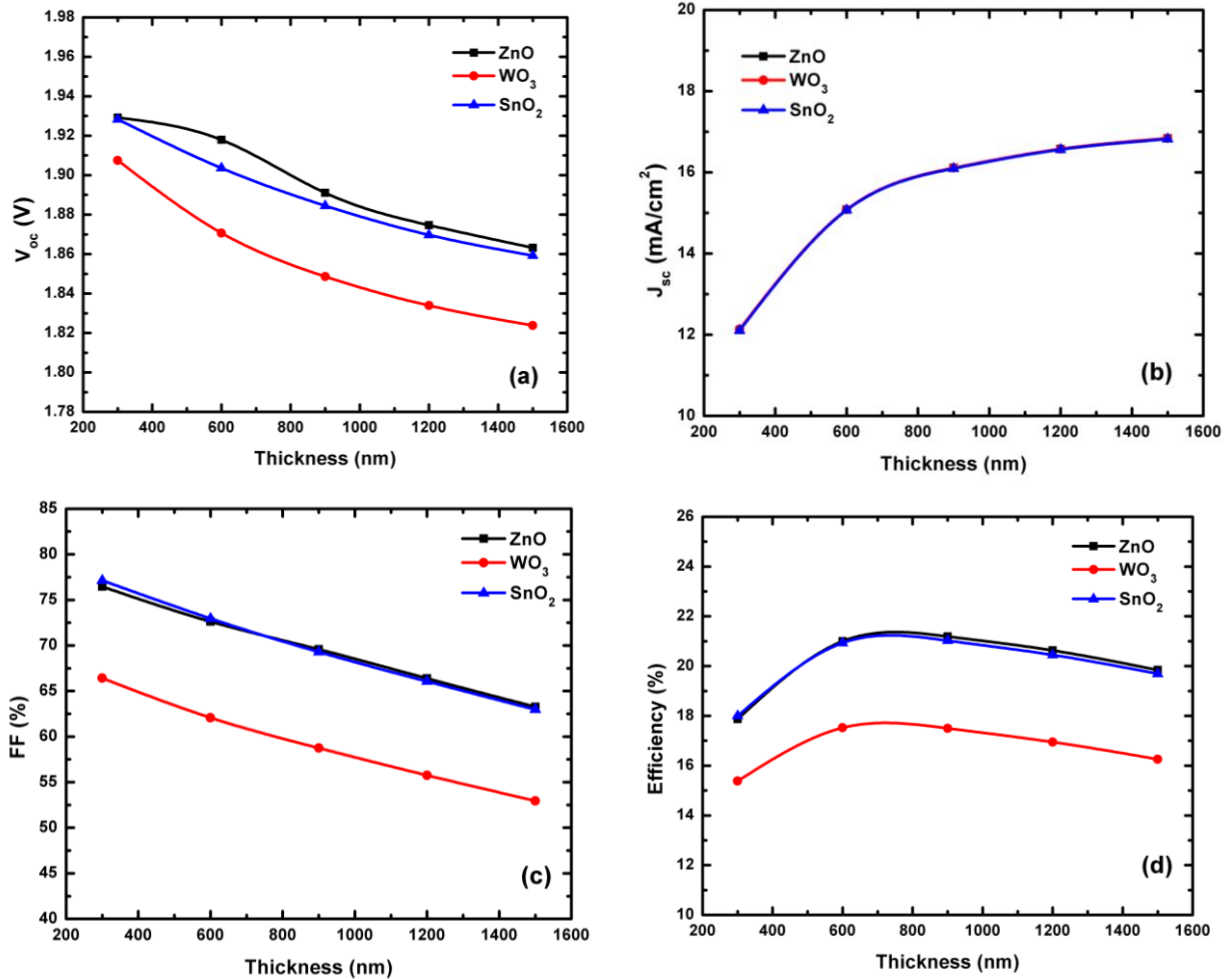


Fig. 2: Variation of (a) V_{oc} , (b) J_{sc} , (c) Fill Factor and (d) efficiency for perovskite thickness variation for different ETL

The square of the intrinsic carrier concentration, n_i , can be given by:

$$n_i^2 = N_C N_V \exp \left[-\frac{E_g}{kT} \right] \quad (3)$$

Where E_g is denoted as the bandgap of the material and N_c and N_v define the conduction band and valence band effective density of states of the charge carriers. From these above equations, it can be observed that V_{oc} is a function of the bandgap. With the increase in the bandgap, the diode saturation current density increases which in turn results in the reduction of V_{oc} [20]. For this reason, lower performance is found in the case of WO_3 which possesses a lower bandgap. This phenomenon contradicts ZnO and SnO_2 . Though SnO_2 has a higher bandgap, it has lower V_{oc} than ZnO which is due to the higher mobility of ZnO [13].

Fig. 2(a) shows the variation of V_{oc} with the perovskite $MAGeI_3$ thickness. From the figure, it can be seen that

the V_{oc} decreases with increasing perovskite thickness. As the thickness of the perovskite layer increases, the electron-hole recombination process increases which can increase the diode saturation current density, J_0 [21]. It is evident from equation (1) that an increase in J_0 will reduce the V_{oc} as they have an inversely proportional relation and so with the increase in thickness the V_{oc} value decrease. Next Fig. 2(b) shows the variation of J_{sc} with increasing perovskite thickness. As the perovskite layer becomes thicker with increasing thickness, the possibility to absorb more photons increases which in turn increases the open-circuit value of short circuit current density. Followingly, Fig. 2(c) shows the change of fill factor concerning thickness change which shows a similar kind of behavior of V_{oc} . Finally, Fig. 2(d) shows the effect of thickness variation on solar cell efficiency. At lower thickness (below 400 nm) the efficiency is lower due to the chance of electron-hole recombination before reaching the perovskite/ETL or perovskite/HTL interface. But with progress in the thickness of perovskite

more photons are absorbed which results in the generation of more electron-hole pairs. But after a certain thickness (for each ETL 900 nm) the efficiency starts to deteriorate again as the charge carriers cross their minority carrier diffusion length and the chance of electron-hole recombination increases [22]. The best efficiency has been found for ZnO/MAGeI₃ structure and it is 21.19 % with a V_{oc} of 1.89 V, J_{sc} of 16.11 mA/cm² and FF of 69.57%. The best efficiencies for all 3 ETL materials are summarized in Table 3.

The J-V characteristics and external quantum efficiency (EQE) graphs of the simulated solar cell for different ETLs have been displayed in Fig. 3 which were obtained at the optimized thickness of the MAGeI₃ layer. The S-shaped J-V characteristics curve in Fig. 3(a) has been found due to the higher recombination of charge carriers

at the thicker absorber layer. There is an almost identical shape for ZnO and SnO₂ whereas the curve for WO₃ indicates a lower performance because of its lower bandgap. The EQE curves for three ETL materials are identical shown in Fig. 3(b) which indicates zero contribution of them to the device carrier generation mechanism

Table 3: Variation for different ETL materials

ETL material	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	Efficiency (%)
ZnO	1.89	16.1075	69.57	21.19
WO ₃	1.87	15.0838	62.08	17.52
SnO ₂	1.88	16.0942	69.29	21.02

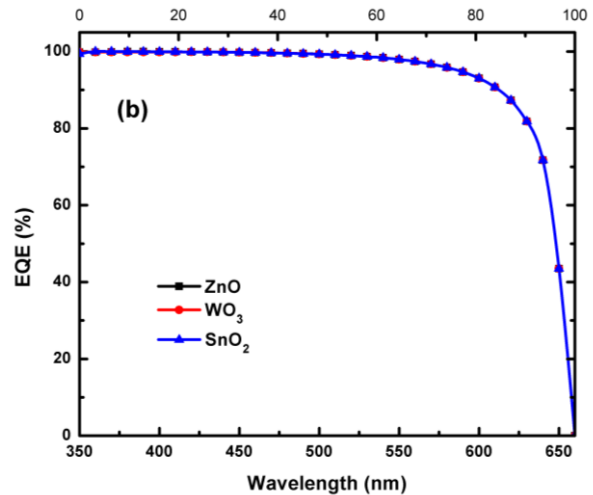
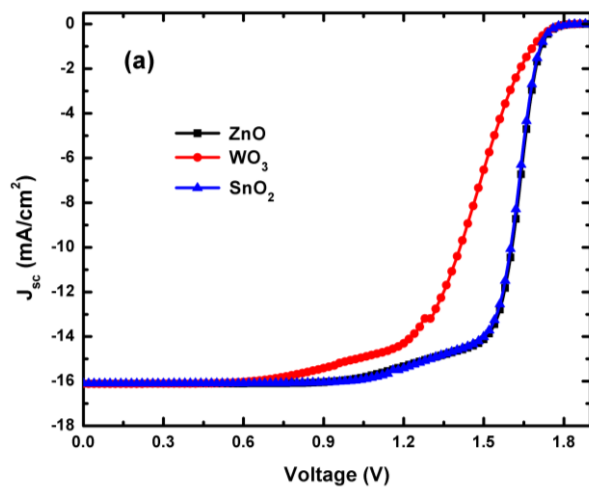


Fig. 3: (a) J-V curves and (b) EQE characteristics for different ETL in the MAGeI₃ solar cell.

3.1.2 Simulation with different HTL

Fig. 4 shows the solar cell parameter variation for different HTL layers- NiO_x, CuI, and PEDOT:PSS with the increase in perovskite layer thickness. In all graphs, black lines were used for NiO_x, red lines for CuI, and blue lines for PEDOT:PSS for indicating the changes. For the case of these three HTL materials, NiO_x has the highest bandgap (3.75 eV) than the other two materials and so it shows better performance which can be observed in Fig. 4. The reason behind this has been discussed in the previous section. Though CuI (2.98 eV) has a higher bandgap than PEDOT:PSS (2.2 eV), it shows lower performances in some cases due to its lower carrier mobility than PEDOT:PSS.

Fig. 4(a) shows the variation of V_{oc} for the increase in perovskite layer thickness. The V_{oc} value decreases almost linearly with the increase in perovskite layer thickness due to the increase of diode saturation current with increasing thickness [20]. Followingly, Fig. 4(b) shows the variation of J_{sc} with perovskite thickness where for all the cases value of J_{sc} increases with the increasing perovskite layer. But for the case of PEDOT:PSS the value is higher than the other two materials which may be higher absorption of photons due to the bandgap variation of the materials. Next, Fig. 4(c) shows the variation of the fill factor. For all three materials, FF shows a decreasing trend when the perovskite layer

thickness increases. Finally, Fig. 4(d) shows the effect on solar cell efficiency. At lower thickness, the efficiency value is lower which then increases up to a certain thickness and after that starts to decrease with increasing thickness. The reason behind this phenomenon has been discussed in the previous section. Among the three structures, the best efficiency has been found for NiO_x/MAGeI₃ structure and the value is 21.19 % with a V_{oc} of 1.89 V, J_{sc} of 16.11 mA/cm² and FF of 69.57%. The best efficiencies for all 3 HTL materials are concise in Table 4.

Table 4: Variation for different HTL materials

HTL material	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	Efficiency (%)
NiO _x	1.89	16.1075	69.57	21.19
CuI	1.88	16.1074	54.55	16.60
PEDOT:PSS	1.58	16.5862	65.44	17.21

Fig. 5 shows the J-V characteristics and EQE curves for a variety of HTL materials. S-shaped J-V curves for NiO_x and CuI have been displayed in Fig. 5(a) while for PEDOT:PSS, the value of J_{sc} concerning V just rises after 1.3 V. The EQE curves for the three HTLs have been displayed in Fig. 5(b) showing almost the same look. The EQE for CuI just shows a higher value from 600 nm to 650 nm rather than the two others for its lower mobility.

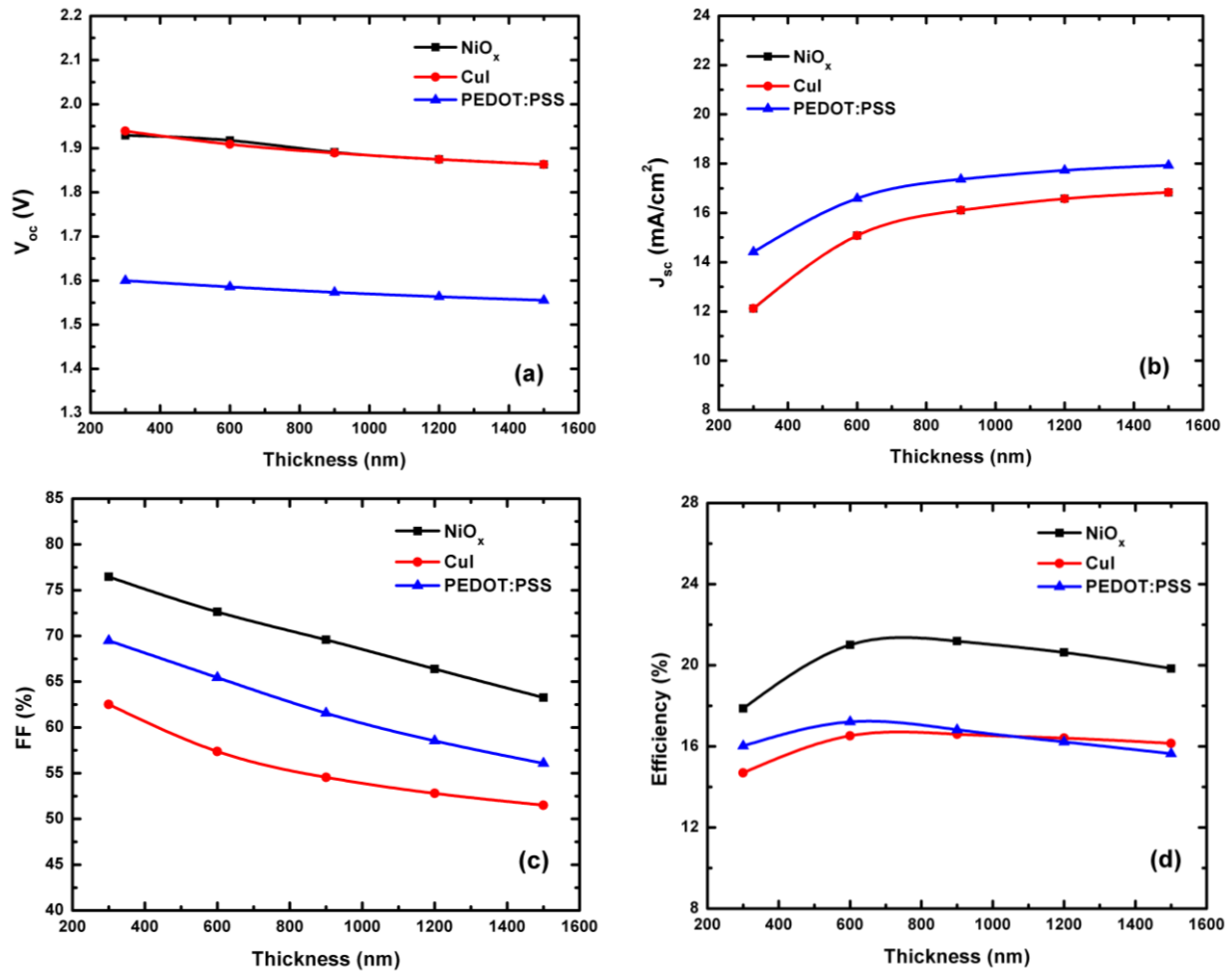


Fig. 4: Variation of (a) V_{oc} , (b) J_{sc} , (c) Fill Factor and (d) efficiency for perovskite thickness variation for different HTL

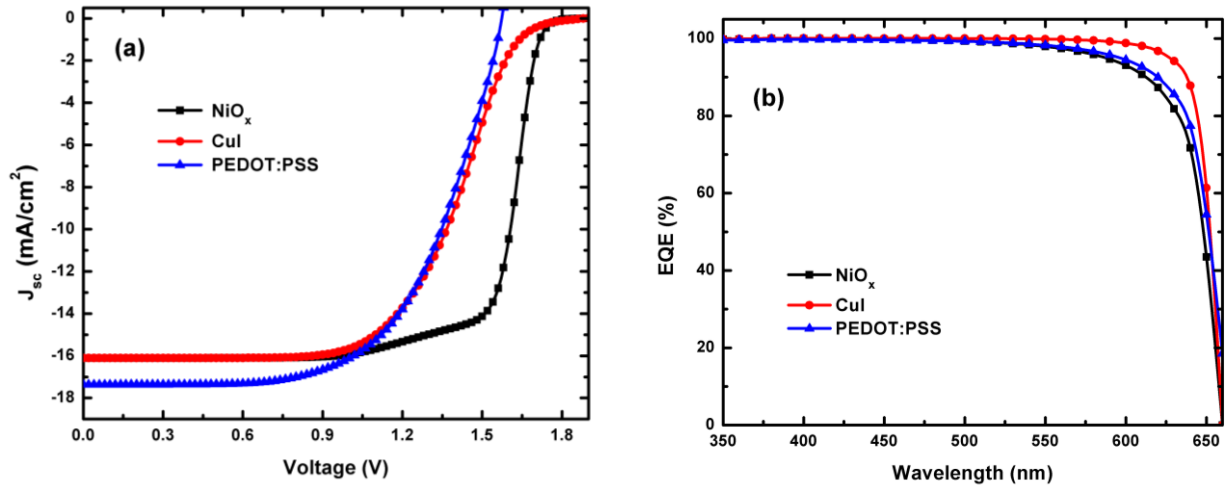


Fig. 5: (a) J-V curves and (b) EQE characteristics for different HTL in the MAgel₃ solar cell.

4. CONCLUSION

In this paper, the performance analysis of MAgel₃ solar cell with three different ETL and HTL layers has been done through numerical simulation using SCAPS-1D. From the thickness variation analysis of the perovskite layer, it has been found that the thickness of the perovskite layer should be kept between 600 nm-900 nm to obtain the best performance of the cell. Below this thickness range, there will be a limited number of photogenerated carriers to help to boost the efficiency

and above the thickness range, the recombination process will be dominant to suppress the efficiency. The best performing solar cell has been found with the combination of ZnO ETL layer and NiO_x HTL layer with an efficiency of 21.19%. As both ZnO and NiO_x are environment-friendly and cost-effective materials, their combination with MAgel₃ perovskite absorber can provide an eco-friendly low-cost solar cell with good efficiency.

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