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Interface Modification by Using an Ultrathin P3HT Layer in a Custom Perovskite Solar Cell Through SCAPS-1D Simulation

Seyyedreza HOSSEİNİ[1], Mahsa BAHRAMGOUR[1], Nagihan DELİBAŞ[2], Aligholi NİAİE*[1]

Abstract

Recently, renewable energy sources such as solar energy have gained much attention for electricity generation because of their easy access and infinite resources. Solar cells are a good choice for this goal. Among the various solar cells that have already been studied, perovskite solar cells (PSCs) have recently become an interesting issue for researchers due to their tremendous improvement in system performance and efficiency. This type of solar cell is divided into several layers, each of which has its role in the structure of the cell. "Front Contact/Electron transporting material/Absorber (perovskite)/Hole transporting layer/Back Contact". The overall structure of these cells has shown a maximum efficiency of about 22% which is good efficiency for solar cells. However, this type of solar cell suffers from stability problems, especially at the junction point between the hole transporting layer (HTM) and the perovskite (absorber) layer, despite its cost-effectiveness advantages. To solve this problem, recent studies have been transferred to a study called interface engineering. In this study, the mentioned interfaces are modified by some materials that have regular and stable structures such as polymers. Many polymeric modifiers have been studied in recent years. Among them, P3HT (Poly(3-hexylthiophene-2,5-diyl)) has provided the best results. In this paper, first, the effect of different layer properties such as their thickness and charge carrier density were investigated and optimal parameters were obtained for each one using SCAPS-1D (Solar Cell Capacitance Simulator) software. Then we simulated the structure of a perovskite solar cell using a polymeric modifier in its structure. The results showed that by adding an ultrathin polymeric film as an interface between HTM and perovskite, the performance of the device was improved and its efficiency was enhanced. The final efficiency of the device with the optimal parameters was obtained about 26.5%.

Keywords: solar cells, perovskite, interface, modifiers, efficiency

1. INTRODUCTION

Electricity energy and the method of electricity generation have become a global issue and interest over the years. Nonrenewable energies like fossil fuels are the largest sources of energy generation because of their high efficiency and good performance. Nowadays, the shortage of nonrenewable energy resources is an important problem because of their limited content and their

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environmental impacts like air pollution and releasing greenhouse gases. Despite these disadvantages, fossil fuels are still the common sources of energy generation; but usage of renewable energies like solar systems that have infinite sources, have progressed rapidly in recent decades. It was predicted that these energy resources will be responsible for most of the electricity generation since 2050 [1].

Solar cells are one of the hopeful resources of electricity generation because of their unique properties like global and overall distribution of sunlight and its low environmental impacts [1]. Generally, solar cells are divided into three common generations. Crystalline silicon was the first type of material that was used for solar cells because of their regular structures and high stability. Despite these good properties, their fabrication demanded high costs because of their relatively high thicknesses. Therefore, solar cell fabrication developed to a new generation that had low cost because of their thin layers. But their efficiency was not as well as the first generation. The final generation of solar cells has progressed to the third generation. Some common types of them are dye-sensitized solar cells (DSSC), Quantum dot solar cells, and perovskite solar cells (PSC).

Perovskite solar cells have become an interesting topic for researchers during the last decade because of their rapid growth in device performance over few years. They have some tremendous features such as tunable band gaps, low fabrication costs, easy fabrication process (for example they don't need high temperatures for their depositions), and acceptable efficiencies. The first study about these solar cells belongs to Miyasaka et al. in 2009 [2] that reached 3.8% efficiency utilizing $CH_3NH_3PbI_3$ as an absorber layer with electrolytes used for charge-transporting layers. However, this was not the desired result and indicated low stability because corrosion occurred between absorber and electrolytes. Although, researches about perovskite solar cells continued and reached a structure with solid and relatively stable charge transporting materials in 2012. This study was done by Kim et al. [3] that reached an efficiency of 9.7%. Over the years, studies about these cells continued and researchers have reached efficiencies of about 22%.

Perovskite solar cells structure generally consists of three types of layers. The most important layer is named absorber layer that is a perovskite material having a general formula of MAX_3 that M, A, X is referred to as monovalent cation, a divalent cation, and a halide ion, respectively. According to carriers of these ions, perovskite is a generally inert material. This layer absorbs photon or light's energy and generates electrons and holes due to the concept of solid-state band theory and the electron-hole pair generation process in semiconductors. Other layers of PSC's structure are electron transporting material (ETM) and hole-transporting materials (HTM) that carry electrons and holes and transport them to counter electrodes, respectively. The total structure of these cells is shown in figure 1.

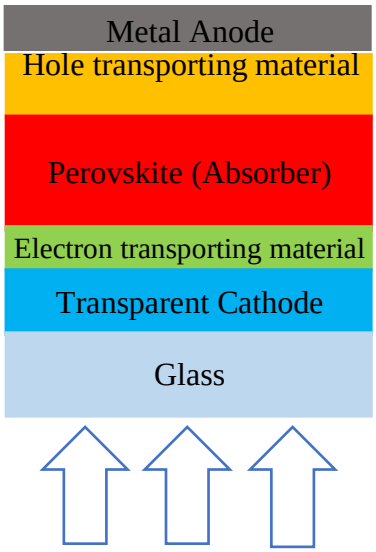


Figure 1 Perovskite solar cell's general structure [1]

Although perovskite solar cells have many advantages as mentioned above, they have some problems with stability issues. Charge recombination and corrosion at the interfaces of these cells especially at the interface between HTM and absorber layer leads to the unstable structure during the time. There have been some approaches for solving this problem in recent years. Some of them belong to a science named interface engineering that utilizes a material with more stable and regular structures like polymers to modify the surface and interfaces. Some

experimental studies have been performed to investigate the effect of the ultrathin polymeric layer in the interface between the absorber layer and charge transporting materials in recent years. All of them represented that the addition of polymeric modifiers led to an increment in both stability and efficiency indexes. Cai et al. in 2018 [4] investigated the effect of three different polymers including PMMA[2], MEH-PPV[3] and PEG[4] on the PSC with TiO_2 layer; results indicated that by addition of these polymers not only stability improved but also efficiency increased from 15.49% to 17.15%, 17.09%, 16.96% for PMMA, MEH-PPV, PEG modifiers, respectively. In 2018 Du et al. [5] studied the effect of the addition of PTAA[5] to the PSC structure based on NiOx; results dedicated that the stability improved and efficiency increased from 13.2% for NiO_x-based to 16.7% for PTAA/NiO_x based PSC. Another empirical study was done by Hunag et al. in 2018 [6] that investigated the effect of utilizing P3HT[6], PTAA, MEH-PPV, poly-TPD[7], PBDTTT-CT[8] as the polymeric modifiers on the TiO_2/perovskite/Spiro-OMeTAD structure; results indicated that the best performance was for the P3HT as the modifier that it improved the device efficiency from 15% for the simple structure to 19% for the P3HT-modified structure. In this study, we will investigate the effect of P3HT on the conventional TiO_2/perovskite/Spiro-OMeTAD[9] structure in the simulation environment for the first time. Before this investigation, the effect of PSC's different layer properties and the optimum amount of each one will be investigated to improve the cell's efficiency.

2. GENERAL REQUIREMENTS

The simulation tool that was used in this work is software for solar cells named Solar Cell Capacitance Simulator in optical-electrical 1-dimensional physics abbreviated as SCAPS-1D. This simulation tool was developed by Marc Burgelman et al. at the Department of Electronics and Information Systems (ELIS) of the University of Gent, Belgium. It can simulate up to seven layers (in addition to electrodes) with each one's properties like bandgap, electron affinity, defects, etc. SCAPS-1D can give us information about energy diagrams, recombination plots, and I-V (current-voltage) characteristics including VOC[10], ISC[11], FF[12], PCE[13] (also called eta) and I-V curve. This software simulates the cell's structure and gives results by solving Poisson's equation and electron and hole continuity equations simultaneously [7]. The equations are as below:

$$\vec{\nabla}.\varepsilon\vec{\nabla}\varphi = -q(p - n + N_D + -N_{A^-}) \quad (1)$$

$$\vec{\nabla}.\overrightarrow{J_n} = q(R - G) + q\frac{\partial n}{\partial t} \quad (2)$$

$$-\vec{\nabla}.\overrightarrow{J_p} = q(R - G) + q\frac{\partial p}{\partial t} \quad (3)$$

In these equations, φ, ε,q, $p, n, N_D, N_{A^-}, \overrightarrow{J_p}, \overrightarrow{J_n}, G, R$ refer to electrostatic potential, dielectric permittivity, electric charge, hole density, electron density, the dopant concentration of donor, dopant concentration of acceptor, hole current density, electron current density, charge generation rate, recombination rate, respectively.

In this work, two modified and simple optimized perovskite solar cell structures were simulated by SCAPS-1D. First, the Au/ Spiro-OMeTAD/ $CH_3NH_3PbI_3$/ TiO_2/ FTO[14] was investigated and its performance parameters were obtained. Then a modified Au/ Spiro-OMeTAD/ P3HT/ $CH_3NH_3PbI_3$/ TiO_2/ FTO with the ultrathin P3HT polymeric layer was simulated and its effect on

[2]Poly (methyl methacrylate)
[3]Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene]
[4]Polyethylene glycol
[5]Poly(triarylamine)
[6]Poly(3-hexylthiophene-2,5-diyl)
[7]Poly(4-butyl-N,N-diphenylaniline)
[8]Poly[[4,8-bis[5-(2-ethylhexyl)-2-thienyl]benzo[1,2-b:4,5-b′]dithiophene-2,6-diyl][2-(2-ethyl-1 oxohexyl)thieno[3,4-b]thiophenediyl]]
[9]N2,N2,N2′,N2′,N7,N7,N7′,N7′-octakis(4-methoxyphenyl)-9,9′-spirobi[9H-fluorene]-2,2′,7,7′-tetramine
[10] Open-circuit voltage
[11] Short-circuit current
[12] Fill Factor
[13] Power conversion efficiency
[14] Fluoridized tin-oxide

the efficiency of the system was investigated. To simulate the aforementioned structures, optical-electrical properties and characteristics of each layer are required. These properties are obtained from other researches and studies. Here, these properties were given in Table 1 [8-10]. The work function of the electrodes including gold (back contact) and FTO glass (front contact) were set in 5.1eV and 4.4eV, respectively. (Simulation was performed in the temperature of T=300 K, A.M.1.5 spectrum, and light power of 1000 W/m^2)

Table 1 Material parameters used in the cells [8-10]

Properties	TiO_2	$CH_3NH_3PbI_3$	P3HT	Spiro-OMeTAD
Thickness (nm)	100	450	20	200
Bandgap (eV)	3.2	1.5	1.05	3.06
Electron Affinity (eV)	3.9	3.9	3.9	2.05
Dielectric Permittivity (Relative)	9	30	3	3
CB Effective Density of States (1/cm^3)	1.00 E+19	2.50E+20	1.00E +20	2.80E +19
VB Effective Density of States (1/cm^3)	1.00 E+19	2.50E+20	1.00E +20	1.00E +19
Thermal Velocity of Electron (cm/s)	1.00 E+7	1.00E+7	1.00E +7	1.00E +7
Thermal Velocity of Hole (cm/s)	1.00 E+7	1.00E+7	1.00E +7	1.00E +7
Mobility of Electron (cm²/Vs)	2.00 E+1	5.00E+1	1.00E -4	1.00E-4
Mobility of Hole (cm²/Vs)	1.00 E+1	5.00E+1	1.00E -4	2.00E-4
Dopant Concentration of Donor N_D (1/cm^3)	1.00 E+16	0	0	0
Dopant Concentration of Acceptor N_A (1/cm^3)	0	1.00E+17	1.00E +16	1.00E +18
Defect Density Nt (1/cm^3)	0	1.00E+13	0	0

3. RESULTS AND DISCUSSION

The simulation was performed in two different structures including modified and simple structures. Figure 2 shows the structures of these two cells. Total results indicated that using a polymeric modifier improves the cell's efficiency. Before this investigation, for the goal of having an optimized cell, the impact of important layer properties containing thickness and charge density was investigated too. We will discuss these properties in the sections below.

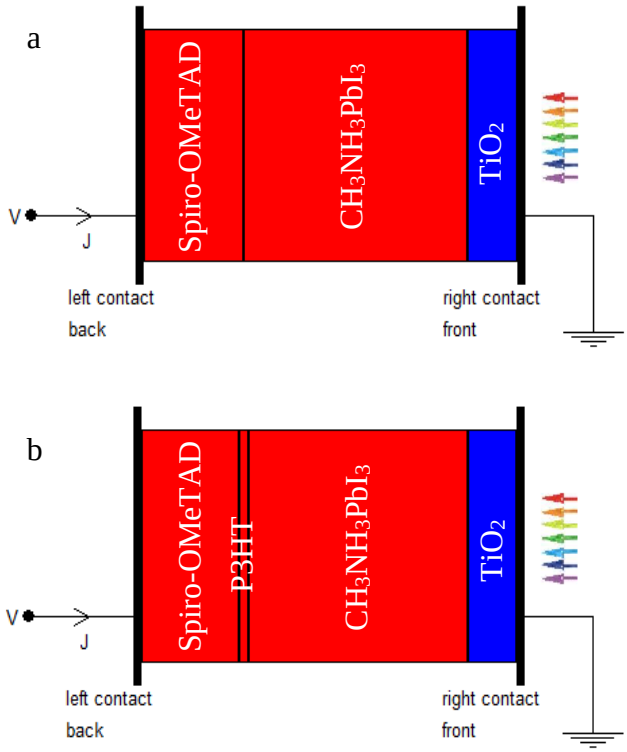


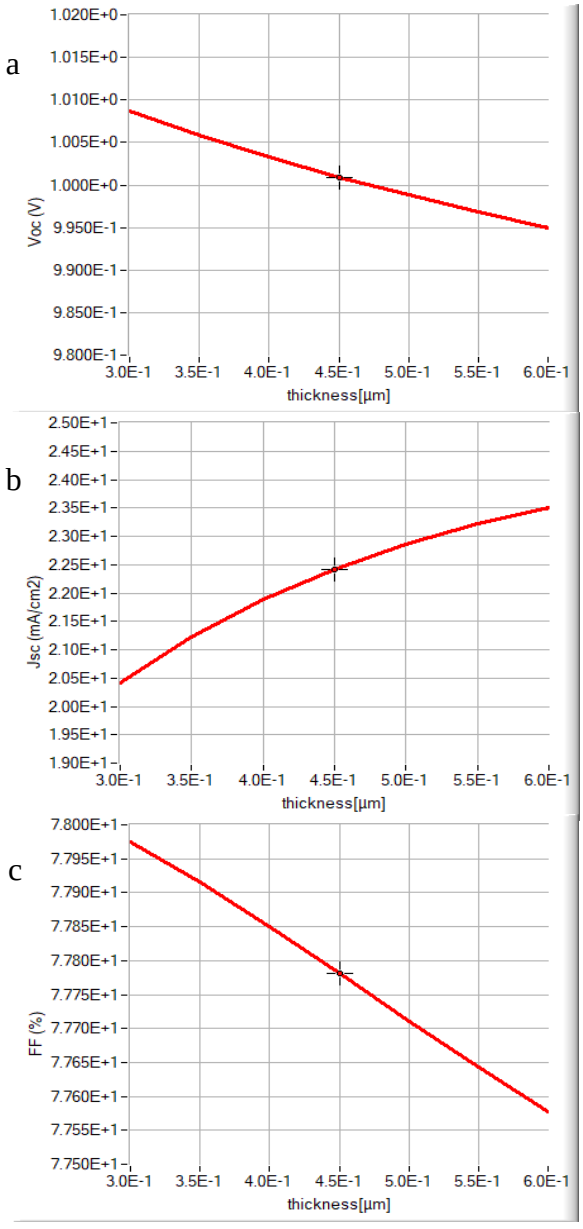
Figure 2 Perovskite solar cell (a) simple (b) modified structure used in this work

3.1. Effect of thickness

3.1.1. Effect of absorber layer thickness

The absorber layer thickness has an important impact on the cell's efficiency. Results indicated that by the increment of perovskite thickness, the cell's overall performance will be improved. Here the thickness was changed from 300-600 nm and results showed that the efficiency increased from 16.05% to 18.15%. This increase is probably due to the greater absorption of photons and the

consequent production of more charge carriers with increasing absorber thickness. In addition, from the plot that was represented in figure 3, it can be seen that the values of V_{OC} and FF have not changed significantly. J_{SC} of the cell increased from 20.40 mA/cm^2 to 23.51 mA/cm^2 and the cell generated more current as the thickness changed. Figure 3 illustrates the influence of changing the thickness of the perovskite layer (absorber) on the photovoltaic parameters of the simple perovskite solar cell that was used in this work.



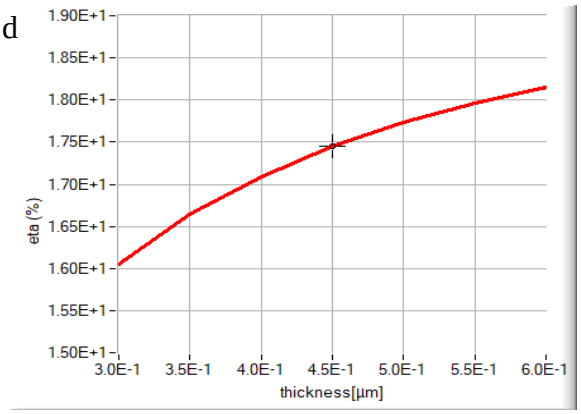


Figure 3 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency (eta) by the increment of absorber thickness

3.1.2. Effect of ETM thickness

Generally, ETM's higher thickness harms the cell's performance. This can be observed from figure 4 that represents the effect of changing ETL's thickness from 50-150 nm vs. photovoltaic parameters of cell with the optimized thickness for absorber layer (600 nm) that was obtained in the previous section. However, as the results indicate, the change in thickness in this layer does not have a considerable impact on cell performance. The best performance was for the thickness of 50 nm with the photovoltaic parameters of V_{OC}= 0.996 V, J_{SC}= 23.52 mA/cm^2, FF= 78.35%, and PCE= 18.36%.

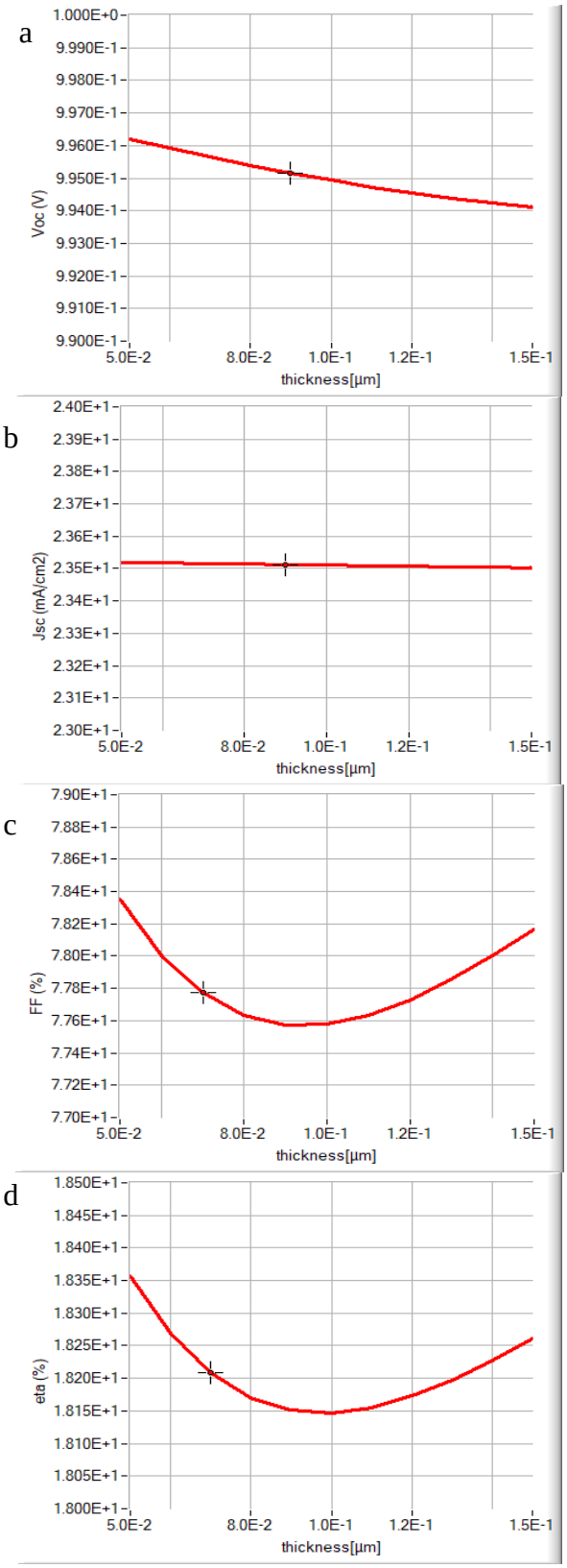


Figure 4 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency by the increment of ETM thickness

3.1.3. Effect of HTM thickness

The HTM layer thickness change has some impacts on the cell's performance too. The plots of figure 5 confirm this subject. From the results, we can understand that thickness increment doesn't have any positive impact on a cell's efficiency. However, this amount shouldn't be less than a custom value. In this investigation, the thickness of the HTM layer was varied from 100-400 nm. Results indicated that the minimum thickness in this range showed the best performance with the optimum values of the layers in the previous sections. Cell parameters at this thickness were obtained at 0.996 V for V_{OC}, 23.52 mA/cm^2 for J_{SC}, 79.01% for FF, and 18.51% for efficiency.

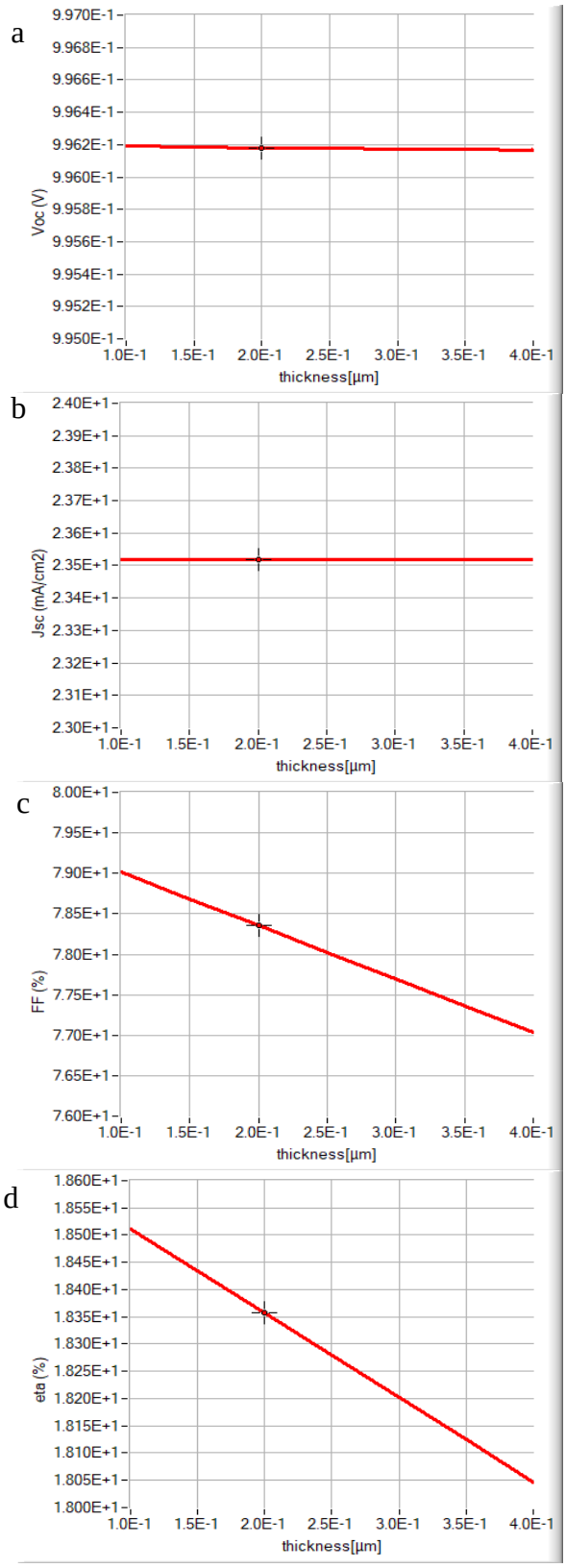


Figure 5 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency (eta) by the increment of HTM thickness

3.2. Effect of dopant concentrations

The dopant concentration of layers indicates the density of charge carriers in a layer in solar cells. It is obvious that as this value was changed to a higher amount, the cell's efficiency will be improved. In the next three sections, this parameter was changed from 10^{15}-10^{19} 1/cm^3 and the results were obtained with the optimal values for the layers' thicknesses. All of the optimal values were the maximum amount of dopant concentrations ranges (10^{19} 1/cm^3). Each section was investigated with the optimum values of its previous sections.

3.2.1. Effect of dopant concentrations of absorber

Figure 6 illustrates the photovoltaic parameters of the studied cell vs. changing dopant concentration of the perovskite layer. Plots indicated the efficiency of 16.18% and 21.50% for the minimum and maximum dopant concentration value of the absorber, respectively. This considerable change is probably because of perovskite's important role in the cell's performance and electricity generation. Results also dedicated that short-circuit current didn't change significantly in the mentioned range but the values of V_{OC} and FF underwent considerable changes.

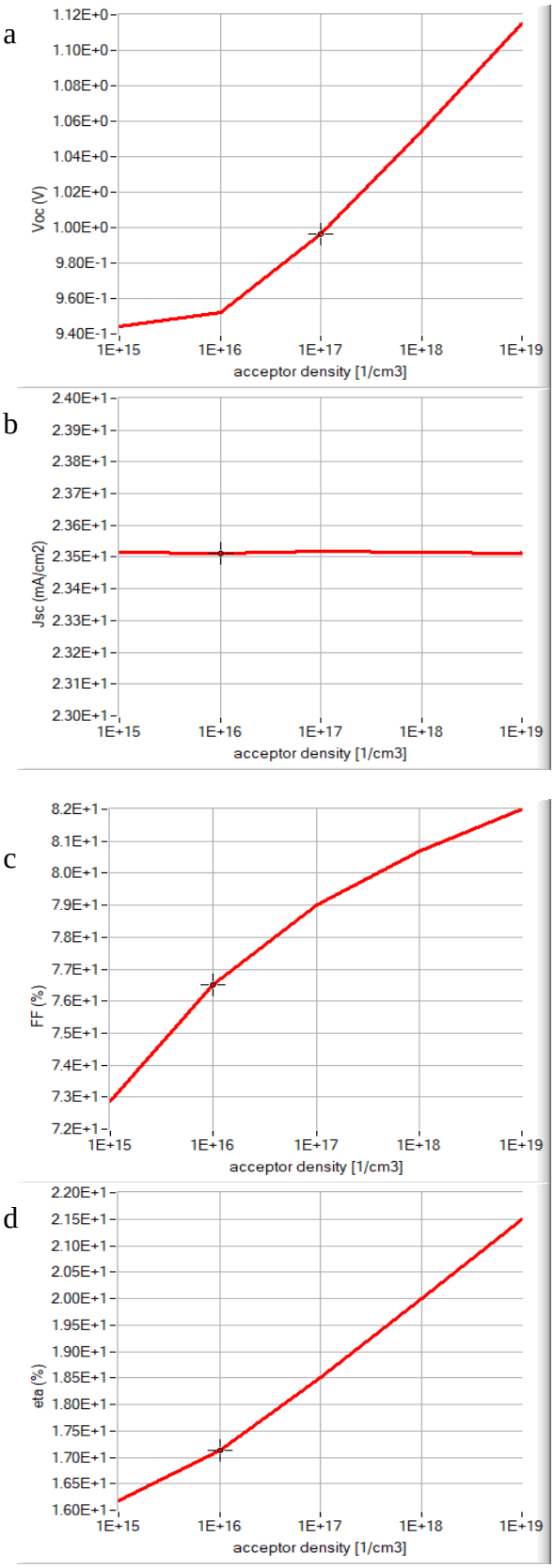
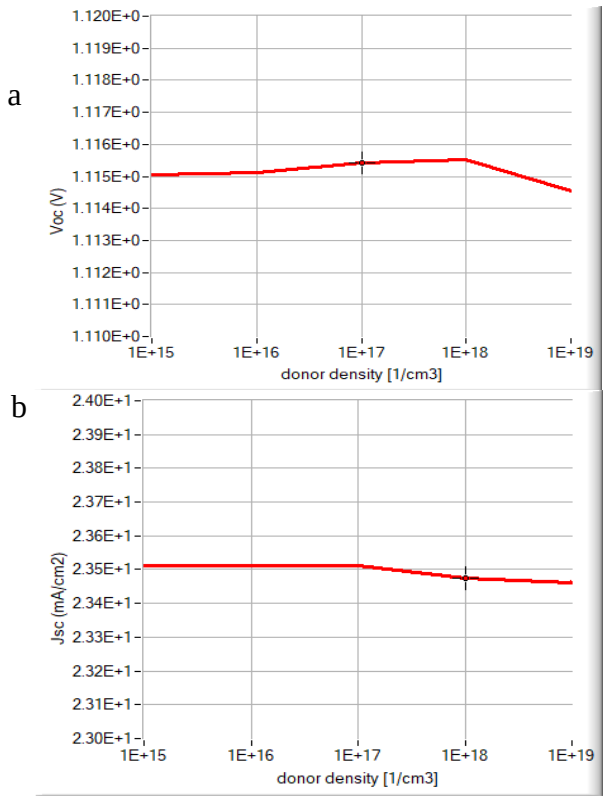


Figure 6 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency (eta) by the increment of dopant concentrations of absorber

3.2.2. Effect of dopant concentrations of ETM

Changes in the dopant concentration of the ETM layer, such as changes in thickness, do not have a significant effect on cell performance. This can be seen in figure 7. However, the main parameters of the cell with the optimal values of the previous sections were obtained in the values of V_{OC}=1.114V, J_{SC}= 23.46 mA/cm^2, FF= 87.48%, and PCE= 22.87%.



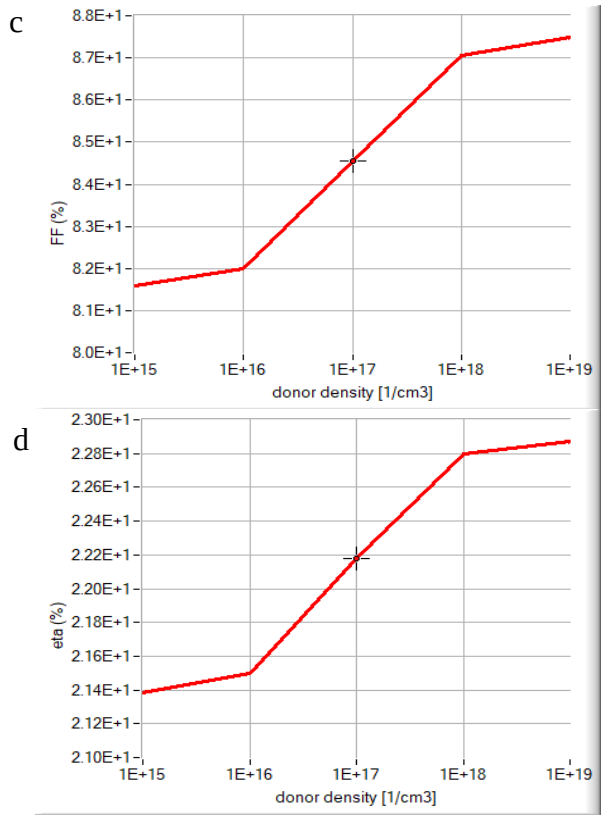


Figure 7 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency (eta) by the increment of dopant concentrations of ETM

3.2.3. Effect of dopant concentrations of HTM

Figure 8 demonstrates the effect of variation of HTM's dopant concentration on the cell's performance with the previous optimal values. Results showed that V_{OC} and J_{SC} didn't undergo considerable changes in their values, but the amounts of FF and PCE indicated almost 30% and 8% in the variation range of dopant concentration, respectively. The optimum value for the acceptor density of HTM was obtained 10^{19} 1/cm^3 that represented the 23.02% value for efficiency.

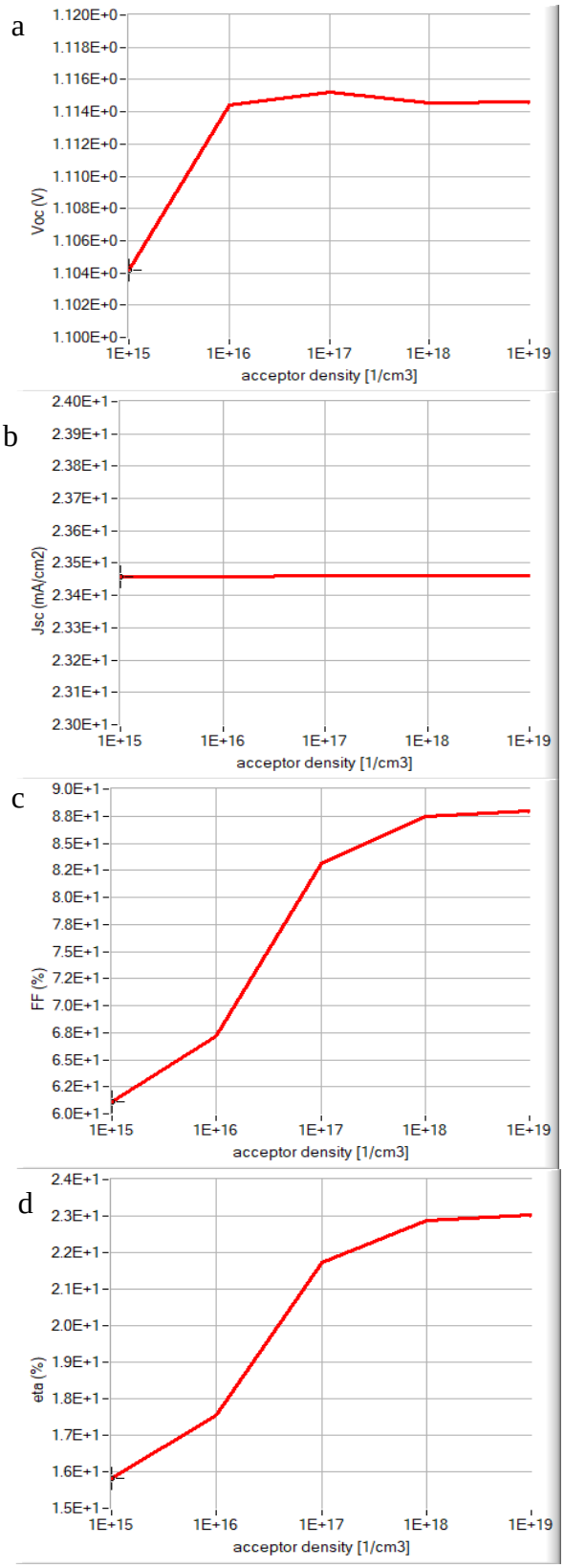
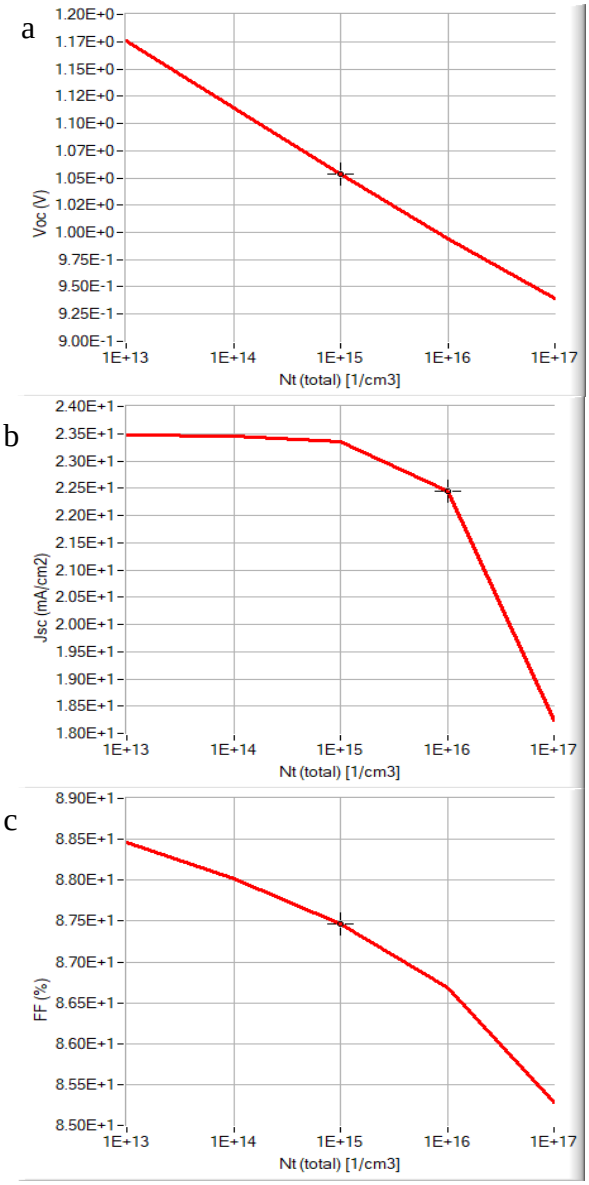


Figure 8 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency (eta) by the increment of dopant concentrations of HTM

3.3. Effect of absorber defect density

Different defects of solar cells' layers always have negative impacts on the cell's performance. This is due to the trapping of charge carriers in the defect sites of the layers. Results of variation of photovoltaic parameters vs. the defect density increment in figure 9 confirm this subject too. Plots represent that there is a 10% difference between the efficiency of the highest and lowest value in the defect density range. The best result was for the density of 10^{13} 1/cm^3 with the previous optimized values and represents the efficiency of about 24.5%. Other photovoltaic parameters for this value were VOC= 1.18 V, JSC= 23.47 mA/cm^2, FF= 88.46%.



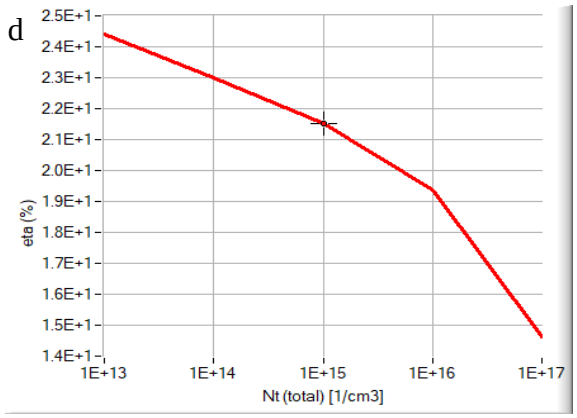


Figure 9 Variation of (a) open-circuit voltage (b) short-circuit current (c) fill-factor (d) power conversion efficiency by the increment of defect density of the absorber

3.4. Effect of interface modification

As it was mentioned in the previous sections, the addition of an ultrathin polymeric layer to the interface between the absorber layer and hole transporting material avoids corrosion among them and lessens the defects because of its regular structure. Besides the stability issue, it helps to improve the cell's efficiency because of less recombination in the interface layer. Our results in this study confirm this effect too. By using an ultrathin 20 nm polymeric P3HT layer in the perovskite and Spiro-OMeTAD layers' interface, the cell's efficiency increased from about 24.5% to 26.5%.

3.5. Final I-V curve

After optimizing all of the factors mentioned above, and the addition of the polymeric modifier to the cell's structure, the final current-voltage (I-V) curve of the modified perovskite solar cell was obtained. The curve was given in figure 10 with its corresponding photovoltaic parameters. Results were in good agreement with the experimental works [11-14]. The final results were represented in table 2.

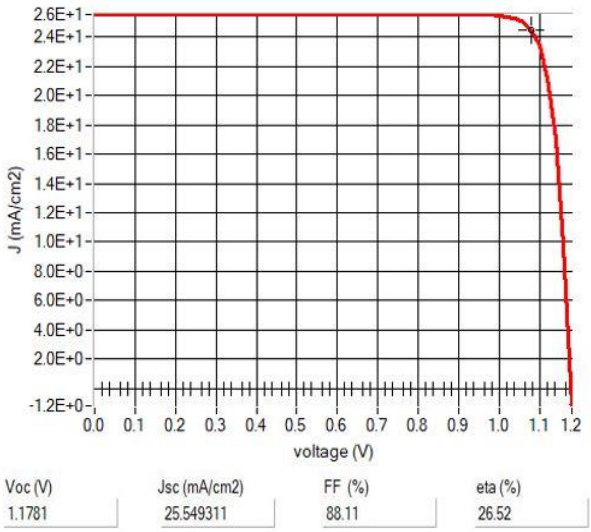


Figure 10 The final current-voltage curve of the modified cell by the optimized parameters

Table 2 Photovoltaic parameters of selected modified and single cells obtained by SCAPS software and their comparison with experimental works [11-14]

Cell's structure	V_{OC} (V)	I_{SC} (mA/cm2)	FF (%)	PCE (%)
Simple (SCAPS-1D)	1.18	23.47	88.46	24.41
Simple (experimental)	0.98	21.2	77.6	18.7
Modified (SCAPS-1D)	1.18	25.54	88.11	26.52
Modified (experimental)	1.13	-	-	20.8

4. CONCLUSIONS

In this study, two simple and modified structures of a custom perovskite solar cell based on $CH_3NH_3PbI_3$ were simulated by SCAPS-1D in two simple and modified cases and their performance was investigated separately with the optimized parameters for each layer. Results represented that the modified cell with P3HT as a polymeric interface between the perovskite and HTM layer indicated the best efficiency. The optimized thicknesses for the layers were 100 nm, 600 nm, 50 nm for HTM, absorber, ETM layers, respectively. The optimal value of dopant concentration of the mentioned layers was 10^{19} $1/cm^3$ for each layer. With the optimal values that were obtained, the final and best efficiency was about 26.5% for the modified cell.

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The Declaration of Conflict of Interest/ Common Interest

This study was produced from the Seyyed Reza Hosseini, Master thesis which was accepted on April 14, 2021, in the Department of Chemical Engineering, the University of Tabriz with supervisor Prof. Aligholi Niaei and advisor Dr. Nagihan Delibaş.

Authors' Contribution

Within this study, all authors were involved and cooperated in idea formation, simulation, evaluation of the data, and results.

The Declaration of Ethics Committee Approval

This study does not require ethics committee permission or any special permission.

The Declaration of Research and Publication Ethics

The authors of the paper declare that they comply with the scientific, ethical, and quotation rules of SAUJS in all processes of the paper and that they do not make any falsification on the data collected. In addition, they declare that Sakarya University Journal of Science and its editorial board have no responsibility for any ethical violations that may be encountered and that this study has not been evaluated in any academic publication environment other than Sakarya University Journal of Science.

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