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Research Paper

A Simulation Study around Investigating the Effect of Polymers on the Structure and Performance of a Perovskite Solar Cell

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Abstract

Polymers are a very vast classification of materials that possess a lot of applications in various industries. For instance, they have application in structure modification of the perovskite solar cells (PSCs). Polymers' application in perovskite solar cells can be divided into their usage as hole-transporting materials (HTMs) and the ultrathin interfaces between hole transporting materials and the perovskite layer. In the present research, we tried to highlight this application from the simulation perspective using SCAPS-1D software. For this purpose, this study investigates the effect of using different polymeric HTMs and interfaces from the photovoltaic parameters view. The total PSC structure was in the form of Au (Back contact)/ HTM/ polymeric Interface (if there are)/ CH₃NH₃PbI₃ (absorber)/ TiO₂ (Electron Transporting Material: ETM)/FTO (counter electrode). Results represented the best hole transporting material and interface as PEDOT:PSS and P3HT layers. The final efficiency was obtained at 18.77% with the optimal mentioned layers' materials.

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1. INTRODUCTION

The vast classification of polymers and their excellent chemical, physical and mechanical features make them be solid choices for many industrial applications [1]. Solar cells are no exception. Polymers can act as effective layers in various types of solar cells. Some studies reviewed the polymers' impacts on solar cells performance [2, 3].

One of the essential roles of polymers is their application in perovskite solar cells (PSC) [2]. These types of materials can be in the form of hole-transporting materials (HTMs), electron transporting materials (ETMs), and ultrathin interface layers in PSCs [2]. Typically, perovskite solar cells represent acceptable device performances, but they still suffer from stability problems [4-6]. The stability problem in PSCs contains thermal stability caused by their high-temperature processing [7] and interface instability arising from interlayers' corrosion [8-10]. For these reasons, in recent years, a science named perovskite solar cells' interface engineering was developed [4, 9]. Regarding polymers' high crystallinity and ability to cross-link their adjacent layers, they are the most outstanding selection for interface modification [11].

Moreover, utilizing polymers such as HTM can improve device stability and increase efficiency due to their relatively better hole mobilities [12]. Generally, ultrathin polymeric interfaces between the perovskite layer and HTM in perovskite solar cells are a kind of HTMs and can perform hole transportation processes [4]. Therefore, using polymeric HTMs and interfaces simultaneously can help the hole transportation process and improve the device performance.

Despite different research around other types of solar cells (e.g., CIGS, CZTS, GaAs, etc.) [13-15], regarding their relatively higher performances, perovskite solar cells study has become an interesting issue for researchers in both simulation and empirical phase in last decade. For instance, in 2020, a simulation study was performed by Jalalian et al. [16] by using various organic and inorganic HTMs including Spiro-OMeTAD, CuO, and Cu₂O. Results revealed Cu₂O as the optimum selection with an efficiency of 22.12%. In 2021, Rafiee Rafat et al. [17] compared using ZnO and TiO₂ in Pb-based and Sn-based PSCs from the simulation view. Results represented the better performance for ZnO in Sn-based and TiO₂ for Pb-based PSCs.

However, regarding mentioned polymeric layers' acceptable results, researchers performed some experimental and simulation studies about using polymers as interfaces and HTMs in recent years. In 2018, Huang et al. [4] investigated using P3HT, PTAA, MEH-PPV, poly-TPD, and PBDTTT-CT polymers as interfaces between the absorber and HTM of a PSC. Results

revealed that the performance of P3HT-modified PSC possessed a higher value relative to other interfaces and non-modified cells. Another study was performed by Cai et al. [18] in 2018 around employing PMMA, PEG, and MEH-PPV interfaces. Results indicated that using PMMA as an interface revealed the best result which approximately represented 2% higher efficiency relative to the reference structure. In 2021, Hosseini et al. [19] performed a SCAPS-1D simulation study around utilizing P3HT as an interface layer. Results represented about a 2% increment due to the usage of the interface. In the case of polymeric HTMs investigation, a simulation was performed by Tan et al. [20] in 2016 around using Spiro-OMeTAD, PTAA, CuI, MEH-PPV, P3HT, PTV, PCPDTBT as hole transporters. Among polymeric HTMs, PTAA resulted in better power conversion efficiency (PCE) of about 17.44%.

In the present study, intending to select proper polymer layers, we tried to study three different polymeric interfaces, including P3HT (thinner), PTAA, and Poly-TPD, and four different HTMs containing PANI, PEDOT:PSS, P3HT (thicker), and Spiro-OMeTAD from the performance and efficiency perspective. For this purpose, we employed the SCAPS-1D simulation tool. Fig. 1 demonstrates the schematic of mentioned solar cell's overall structure.

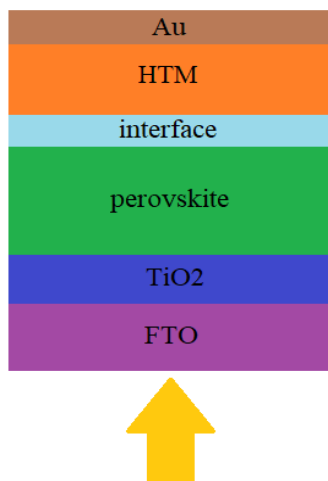


Fig. 1. The perovskite solar cell structure used in the present work

2. MATERIALS AND METHODS

A. Simulation Environment

The main goal of the present work is the performance comparison of some common polymers employed in the perovskite solar cells' structure. For this purpose, we selected the simulation study utilizing a one-dimensional optical-electrical software package named SCAPS-1D (Solar Cell Capacitance Simulator). This program was developed by Marc Burgelman et al. at the Department of Electronics and Information Systems (ELIS) of the University of Gent, Belgium. The calculation in this software is based on basic Poisson and electron and hole continuity equations [21].

For device simulation in this program, some crucial parameters of layers are required. Moreover, other work points like temperature and spectrum should be considered. Therefore, mentioned parameters for polymeric and non-polymeric layers of the simulated PSC are collected in Table I and Table II. The simulation temperature was set at 300 K. The spectrum file is calibrated on A.M.1.5.

In this work, the overall perovskite solar cell structure that was introduced in the simulation is Au (Back contact)/ HTM/ polymeric Interface (if there are)/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ (absorber)/ TiO_2 (ETM)/FTO (Front contact). The work functions of electrodes are Au= 5.1 eV, FTO= 4.4 eV. There are some reasons to employ the mentioned materials as the studied PSCs constructing layers. For Back Contact, we employed gold (Au). Despite its lower conductivity compared to other metals such as silver (Ag) and Aluminum (Al) and higher work function, its less chemical activity and lower oxidation makes it a more efficient electrode. Therefore, in most studies about PSCs, Au is utilized as Back Contact [22]. $\text{CH}_3\text{NH}_3\text{PbI}_3$ was utilized as an absorber by many researchers because of its inherent suitable properties that result in higher efficiencies [23]. TiO_2 layer was chosen as a useful semiconductor in various applications such as solar cells. It can be because of its high refractive index and chemical stability. Hence, such as many other studies about PSCs, we utilized TiO_2 as ETM [24]. Finally, about Front contact, in the PSC study case, most researchers utilize FTO because of its suitable work function, low-temperature fabrication, and low operational costs [25]. In the case of selection of the hole transporting material, we discussed in detail using polymeric materials in the next section. Furthermore, Spiro-OMeTAD was used as HTM for more compared with the polymers. It is because of its less recombination, less series resistance, and high efficiency [23].

TABLE I
LAYER PARAMETERS OF NON-POLYMERIC LAYERS EMPLOYED IN THE PRESENT WORK
[26]

Properties	TiO ₂	CH ₃ NH ₃ PbI ₃	Spiro-OMeTAD
Thickness (nm)	50 [4]	210 [4]	150 [4]
bandgap (eV)	3.2	1.5	3.06
electron affinity (eV)	3.9	3.9	2.05
dielectric permittivity (relative)	9	30	3
CB effective density of states (1.cm ⁻³)	1.00E+19	2.50E+20	2.80E+19
VB effective density of states (1.cm ⁻³)	1.00E+19	2.50E+20	1.00E+19
the thermal velocity of electron (cm.s ⁻¹)	1.00E+7	1.00E+7	1.00E+7
the thermal velocity of hole (cm.s ⁻¹)	1.00E+7	1.00E+7	1.00E+7
mobility of electron (cm ² . (Vs) ⁻¹)	2.00E+1	5.00E+1	1.00E-4
mobility of hole (cm ² . (Vs) ⁻¹)	1.00E+1	5.00E+1	2.00E-4
dopant concentration of donor N _D (1.cm ⁻³)	1.00E+16	0	0
dopant concentration of acceptor N _A (1.cm ⁻³)	0	1.00E+17	1.00E+18
Defect density N _t (1.cm ⁻³)	0	1.00E+13	0

TABLE II
LAYER PARAMETERS OF POLYMERIC LAYERS EMPLOYED IN THE PRESENT WORK [26]

Properties	P3HT	PEDOT: PSS	PANI	PTAA	Poly- TPD [4]
Thickness (nm)	150,15 [4]	150 [4]	150 [4]	15 [4]	15
bandgap (eV)	1.05	1.5	2.46 [27]	3 [20]	2.9
electron affinity (eV)	3.9	3.6	2.05	2.05	2.05
dielectric permittivity (relative)	3	10	3	3	3
CB effective density of states ($1.\text{cm}^{-3}$)	1.00E+20	1.00E+2 1	2.80E+1 9	2.80E+1 9	2.80E+1 9
VB effective density of states ($1.\text{cm}^{-3}$)	1.00E+20	1.00E+2 1	1.00E+1 9	1.00E+1 9	1.00E+1 9
the thermal velocity of electron ($\text{cm}.\text{s}^{-1}$)	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7
the thermal velocity of hole ($\text{cm}.\text{s}^{-1}$)	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7
mobility of electron ($\text{cm}^2. (\text{Vs})^{-1}$)	1.00E-4	1	1.00E-4	1.00E-4	1.00E-4
mobility of hole ($\text{cm}^2. (\text{Vs})^{-1}$)	1.00E-4	40	2.7E-5 [27]	5.00E-4 [20]	1.00E-4
dopant concentration of donor N_D ($1.\text{cm}^{-3}$)	0	0	0	0	0
dopant concentration of acceptor N_A ($1.\text{cm}^{-3}$)	1.00E+16	1.00E+1 9	1.00E+1 8	1.00E+1 8	1.00E+1 8
Defect density N_t ($1.\text{cm}^{-3}$)	0	0	0	0	0

B. Polymers Used

Understanding the structures and properties of mentioned polymers can help to recognize their effects on their corresponding application. Therefore, here, we tried to introduce some of the features of the polymers utilized in this study.

Despite other polymer candidates that could be used in this study, we tried to select the most common ones employed in recent works [4, 18]. All of them in both available layers, including interface and HTM, have some promotive effects on the cell's performance. For instance, utilizing them leads to less

charge recombination, higher device stability, and generally higher efficiency [4]. The chemical structure polymers employed in this work and their molecular arrangement are demonstrated in Fig. 2.

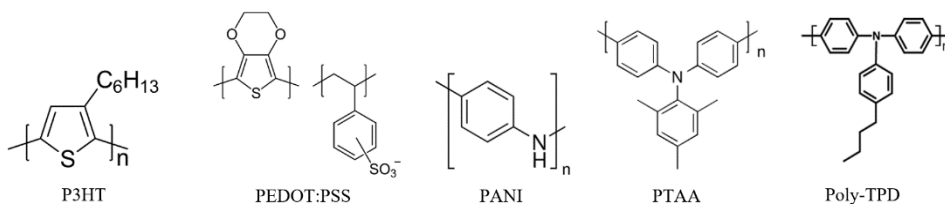


Fig. 2. chemical structure of polymers employed in this study

3. RESULTS AND DISCUSSION

A. Comparison of the polymeric hole-transporting materials

In the first step, four different polymeric HTM, including P3HT, PANI, PEDOT:PSS, and Spiro-OmeTAD, have been studied and simulated. We compared the results in the form of photovoltaic parameters. Moreover, their Current Density-Voltage behavior was obtained and demonstrated in Fig. 3. According to the parameters' values reported in Table III, the short-circuit current (J_{sc}) of P3HT and Open-circuit voltage (V_{oc}) of Spiro-OMeTAD possess a maximum amount compared to other polymeric HTMs. Besides, PEDOT:PSS has 17.41% power conversion efficiency (PCE) and maintains maximum efficiency among other HTM. Generally, it can be understood that PEDOT:PSS and Spiro-OMeTAD represent better performance parameters. In the present work, PEDOT:PSS was selected as the optimum HTM. In a similar position, Karimi et al. [26] 2016 investigated the effect of hole-transporting materials (Spiro-OMETAD, PEDOT:PSS, NPB, MEH-PPV, P3HT) on the performance of perovskite solar cells. The results obtained from the simulation with SCAPS-1D were 23.18% for Spiro-OMeTAD and 21.60% for PEDOT:PSS.

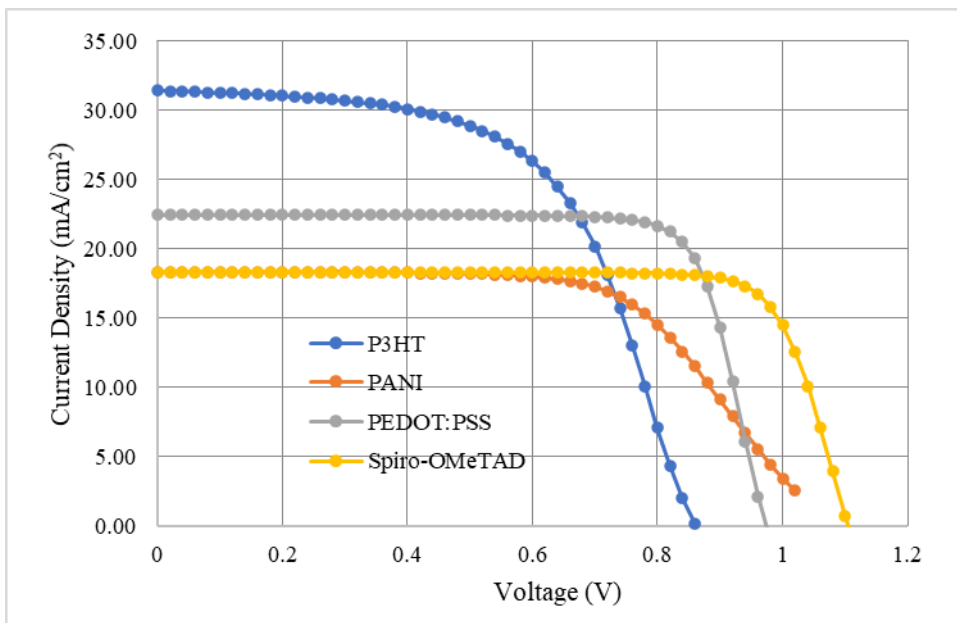


Fig. 3. The current density-voltage curve of perovskite solar cell with a different kind of Polymeric HTM

TABLE III
POLYMERIC HTM PHOTOVOLTAIC PARAMETERS

HTM	V_{oc} (V)	J_{sc} (mA.cm ⁻²)	FF (%)	PCE (%)
P3HT	0.86	31.41	58.41	15.81
PANI	1.08	18.32	61.97	12.22
PEDOT:PSS	0.97	22.45	79.66	17.41
Spiro-OMeTAD	1.10	18.30	80.63	16.30

B. Effect of various Polymeric Interfaces

In the second step, with the optimum HTM (PEDOT:PSS), the effect of modification of the PSC structure using the polymeric interface layer with a thickness of 15 nm was studied. For this purpose, three polymers containing P3HT, Poly-TPD, and PTAA were introduced. The overlapped results for the PSCs (PEDOT: PSS-based) with and without interfaces were illustrated in Fig. 4 (It should be noted that PTAA-based and Poly-TPD-based PSC curves behave

similarly. Therefore, yellow and red curves are overlapped). Furthermore, the photovoltaic parameters of the mentioned cases were collected in Table IV. According to the values, J_{SC} of P3HT-based and V_{OC} of Poly-TPD-based and PTAA-based structures revealed higher amounts. Besides, P3HT-based PSC announced 18.77% efficiency and possessed better efficiency than other polymeric interface layers. We can conclude that by inserting the interface layer into HTM, the total thickness of HTM (HTM plus Interface) enhances. This leads to rapid hole transportation, less recombination, and higher efficiency. In general, the obtained results agreed with Huang et al. [4] who worked on interface engineering of perovskite solar cells in 2018. They experimentally proved that the device with a P3HT interlayer shows more brilliant long-term stability than that without an interlayer when exposed to moisture. They declaimed that the enhanced device performance based on P3HT interlayer compared with the other polymers can be ascribed to the long hydrophobic alkyl chains and the small molecule monomers of P3HT, which contribute to the self-assembly of the polymers into insulating layers and formation of the efficient π - π stacking in polymer/spiro-OMeTAD interface simultaneously [4].

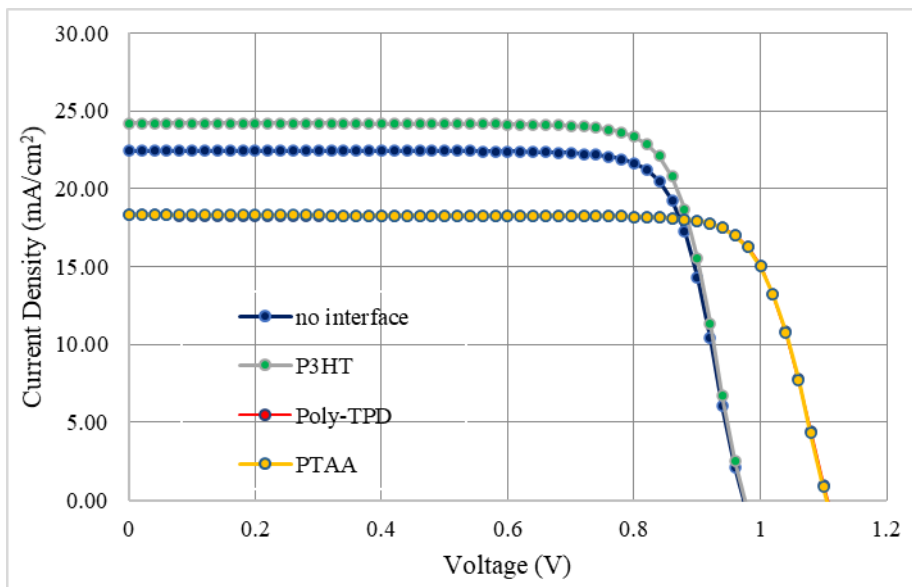


Fig. 4. Current density –voltage curve of perovskite solar cell with a different type of polymeric interface layers

TABLE IV
POLYMERIC HTM PHOTOVOLTAIC PARAMETERS

Interface	V _{oc} (V)	J _{sc} (mA.cm ⁻²)	FF (%)	PCE (%)
No interface	0.97	22.45	79.66	17.41
P3HT	0.97	24.19	79.55	18.77
Poly-TPD	1.11	18.30	81.23	16.43
PTAA	1.11	18.30	81.25	16.43

C. Overall Results

Generally, in the present study, photovoltaic parameters, including Open-Circuit Voltage (V_{oc}), Short-Circuit Current (J_{sc}), Fill Factor (FF), and Power Conversion Efficiency (PCE), are calculated with four different kinds of HTM and three various types of polymeric interface layers. All of the values of the mentioned structures are reported in Table V and Table VI. The optimum result was obtained for PEDOT:PSS as HTM and P3HT as polymeric interface layer with a PCE of 18.77%.

TABLE V
EFFECT OF THE POLYMERIC HTMS AND INTERFACES ON PHOTOVOLTAIC PARAMETERS (INCLUDING VOC, JSC, FF)

interface \ HTM	P3HT			PANI			PEDOT:PSS			Spiro-OMeTAD		
	VOC	JSC	FF	VOC	JSC	FF	VOC	JSC	FF	VOC	JSC	FF
P3HT	0.8	32.2	58.	1.11	20.5	75.6	0.97	24.1	79.	1.11	20.5	79.8
	6	4	76		6	1		9	55		3	6
Poly-TPD	1.1	18.3	80.	1.11	18.3	57.0	1.11	18.3	81.	1.11	18.3	80.2
	0	0	55		2	2		0	23		0	0
PTAA	1.1	18.3	80.	1.10	18.3	54.6	1.11	18.3	81.	1.11	18.3	80.5
	0	0	53		2	2		0	25		0	6

TABLE VI
EFFECT OF THE POLYMERIC HTMS AND INTERFACES ON DEVICE EFFICIENCY (PCE)

interface \ HTM	P3HT	PANI	PEDOT:PSS	Spiro-OMeTAD
P3HT	16.36	17.27	18.77	18.22
Poly-TPD	16.24	11.55	16.43	16.23
PTAA	16.23	11.05	16.43	16.29

4. CONCLUSION

In this paper, we tried to study three different polymeric interfaces, including P3HT (thinner), PTAA, and Poly-TPD, and four different HTMs containing PANI, PEDOT:PSS, P3HT (thicker), and Spiro-OMeTAD from the performance and efficiency perspective. For this purpose, we used the SCAPS-1D simulation tool. As a result, first, the $\text{TiO}_2/\text{CH}_3\text{NH}_3\text{PbI}_3/\text{HTM}$ structure was simulated. The maximum efficiency obtained was 17.41% for PEDOT:PSS polymeric HTM. In the next step, with the optimum, we compared the outcome of a simulation on interface-modified perovskite solar cells with different polymeric interfaces. For the optimum PEDOT:PSS-based PSC, the best device efficiency result was obtained 18.77% for the P3HT interface.

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