Performance Comparison of Different Hole Transport Layer Configurations in a Perovskite-based Solar Cell using SCAPS-1D Simulation

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Abstract

Due to the solar cell industry, environmentally friendly and low-cost electricity generation processes, the use of non-renewable energy sources, especially fossil fuels, is developing day by day. Among the different solar cells under use, perovskite solar cells have recently experienced rapid growth in research due to their high performance and low production costs at the same time. Perovskite solar cells typically consist of some main layers such as absorbent, carrier layers and electrodes. The hole transport layer (HTL) is very important in the perovskite solar cell structure due to its important role in cell performance. The light absorbed by the perovskite layer leads to the formation of electrons and holes. These load carriers are then transported to the electrodes by the electron and hole transport layers. There are several types of HTL, such as small molecules in the cell structure, polymeric and inorganic HTLs. In addition, these different options can be in various configurations such as tandem, composite and single structures. In this study, three common HTL types, Spiro-OMeTAD, P3HT and Cu_2O, were studied and their effects on cell performance in different composite, tandem and single forms were investigated and their results were compared. These comparisons were made in the simulation environment in SCAPS-1D software. The final results showed approximately the best 27% efficiency of the use of tandem structure in the HTL configuration with Spiro-OMeTAD and P3HT in the special perovskite solar cell created in this study.

Keywords: Tandem, composite, perovskite solar cell, HTL, SCAPS-1D.

SCAPS-1D Simülasyonu Kullanılarak Perovskit Tabanlı Güneş Pilinde Farklı Delik Aktarım Katmanı Konfigürasyonlarının Performans Karşılaştırması

Öz

Güneş pili endüstrisi, çevre dostu ve düşük maliyetli elektrik üretim süreçleri nedeniyle, yenilenemeyen enerji kaynaklarının özellikle fosil yakıtların yerine, kullanımı her geçen gün artmaktadır. Aynı zamanda bilinen farklı güneş pilleri arasındaki perovskit güneş pilleri ile yapılan araştırmalarda yüksek performansları ve düşük üretim maliyetleri sebebiyle hızlı bir büyüme yaşanmıştır. Perovskit güneş pilleri tipik olarak; emici, taşıyıcı katmanlar ve elektrotlar gibi bazı ana katmanlardan oluşur. Hücre performansındaki önemli rolü nedeniyle delik taşıma tabakası (HTL), perovskit güneş pili yapısında çok önemlidir. Perovskit tabakası tarafından emilen ışık, elektron ve deliklerin oluşumuna yol açar. Bu yük taşıyıcıları daha sonra elektron ve delik taşıma katmanları ile elektrotlara taşınır. Hücre yapısındaki küçük moleküller, polimerik ve inorganik HTL'ler gibi çeşitli HTL türleri vardır. Ayrıca bu farklı seçenekler, tek yapılı, tandem ve kompozit gibi çeşitli konfigürasyonlarda olabilir. Bu çalışmada, üç yaygın HTL tipi, Spiro-OMeTAD, P3HT ve Cu_2O oluşturulmuş, bunların farklı kompozit, tandem ve tek formlardaki hücre performansı üzerindeki etkileri araştırılmış olup sonuçları karşılaştırılmıştır. Bu karşılaştırmalar SCAPS-1D yazılımındaki simülasyon ortamında yapılmıştır. Nihai sonuçlar, bu çalışmada oluşturulan özel perovskit güneş pilinde HTL tipi, Spiro-OMeTAD ve P3HT konfigürasyonundaki tandem yapı kullanımın yaklaşık % 27 ile en iyi verimlilik olduğunu göstermiştir.

Anahtar Kelimeler: Tandem, kompozit, perovskit güneş pili, HTL, SCAPS-1D.

1. Introduction

Energy sources and energy generation are always global issues around the world research industry. From the perspective of finite and infinite sources, energies generally divide into two common nonrenewable and renewable energies, respectively. In recent years, because of nonrenewable energies like fossil fuel finishing sources, trends have moved to researches about renewable sources like wind and solar energies. Due to the advantages like the infinite source of sunlight and its generality and environmental friendliness, solar cells have become suitable electricity generation devices. Besides, their fabrication costs are lower than other nonrenewable sources. Silicon solar cells were the first developed generation of solar cells that represented good efficiencies. Silicon solar cells' high fabrication costs led to developing new solar cell generations with other materials representing high efficiencies and low costs. Perovskite solar cells are a new type of solar cells that were developed recently because of their good properties like their tunable bandgaps, low fabrication costs, and high efficiencies.

Perovskite solar cells generally consist of several layers. Like all of the solar cells, the mechanism of electricity generation in these cells is the same but with a little different process. The main layer of a perovskite solar cell is the absorber layer that is a perovskite organic-inorganic material. Bandgaps of this layer are almost in the range of 1.5-2.2 eV. This wide range helps perovskite solar cells to work in different wavelengths by changing the absorbers. The perovskite layer absorbs the photon illuminated from sunlight and then generates electron and hole pair due to the solid-state band theory and charge carrier's generation process. These charge carriers in the perovskite layer are separated by the employing of electron transport layer (ETL) and hole transport layer (HTL) and moved to the electrodes to generate the electric potential and current. These layers are demonstrated in Figure 1.
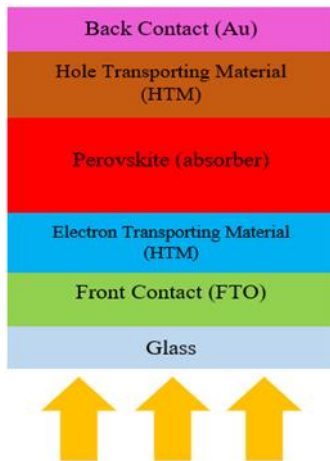

Figure 1. Perovskite solar cell's general structure

Perovskite solar cells are unique since they can be fabricated through different types of materials in each layer. For example, the general formula of the absorber layer is MAX_3 that M is a monovalent cation that can be organic or inorganic cations like MA (methylammonium), FA (formamidinium), Cs, Rb, etc., A is a divalent cation like lead (II), tin (II), germanium (II), etc., and

X is a halide ion like iodine, chlorine, bromine. Utilizing each of these ions leads to different materials in this layer. Moreover, each of the hole transport layers and electron transport layers can be in organic, polymeric, and inorganic forms. Electron transport layers are usually in two organic (like fullerenes) and common inorganic (TiO_2, ZnO, In_2O_3, SnO_2, etc.) forms. Hole transport layers can find in three small molecules, polymer, and inorganic types. Spiro-OMeTAD is the most common HTL used in the perovskite solar cell structure that is a small molecule. It represented good efficiencies, but its high costs led to utilizing other forms like low-cost and high stability polymeric HTLs and inorganic HTLs [1].

We can find that each of these HTLs has some advantages and drawbacks. Therefore, using hybrid structures help the cell to have each type of HTLs' advantages, simultaneously. Hybrid structures in the perovskite solar cells' all layers refer to tandem, composite, etc. configurations. In the tandem one, two different types of the custom layer are deposited in continuous form. For example, two Spiro-OMeTAD and P3HT hole transport layers can be given in the structure instead of single Spiro-OMeTAD or P3HT, and this helps to efficiency increment and higher stability in some cases. The other important hybrid form for the layers is the composite form. In this configuration, in the desired layer two different materials with various compositions can be synthesized.

In 2017 ramli et al. simulated a tandem perovskite/silicon-based solar cell through the SCAPS-1D simulation program. The simulation was done for single silicon solar cells and tandem solar cells. Results represented that using tandem structure had a positive impact on the cell's efficiency and it was changed from almost 24% to 27% from single to tandem structure, respectively [2]. Another simulation was done in 2020 by Hossain et al. that utilized two perovskite absorbers including lead-free $Cs_2AgBi_{0.75}Sb_{0.25}Br_6$ and $CH_3NH_3SnI_3$ absorber as upper and lower layers, respectively. The simulation was done in SCAPS-1D software and they achieved to improved efficiency of about 25% [3].

In this study, using different types and also different configurations of hole transport layers in a perovskite-based solar cell will be investigated via simulation tool for the first time. Three different HTLs including Spiro-OMeTAD as a small molecule, P3HT as a polymer, and Cu_2O as an inorganic one was chosen to compare them in three single, composite, and tandem configurations.

2. Material and Method

The structures mentioned above including various types of HTLs with different configurations were simulated in the SCAPS-1D (Solar Cell Capacitance Simulator) simulation tool. It is a program specially developed for solar cells by Mark Burgelman at the Department of Electronics and Information Systems (ELIS) of the University of Gent, Belgium. It can give us information about charge recombination and their curve, I-V curve, and photovoltaic parameters containing V_{OC}, J_{SC}, FF, PCE [4]. In the mentioned perovskite-based structure, Au were employed as cathode (work function =5.1 eV), FTO as anode (work function =4.4 eV), TiO_2 as the electron transport layer (thickness= 100 nm), and $CH_3NH_3PbI_3$ as the absorber layer (thickness= 450 nm), and three Spiro-OMeTAD, P3HT, Cu_2O (thickness= 200 nm) in the hybrid and single forms as the hole transport layer.

Table 1. Material parameters used in the cells [5-7]

properties	TiO$_2$	CH$_3$NH$_3$PbI$_3$	P3HT	Spiro-OMeTAD	Cu$_2$O
Thickness (nm)	100	450	200	200	200
bandgap (eV)	3.2	1.5	1.05	3.06	2.17
electron affinity (eV)	3.9	3.9	3.9	2.05	3.2
dielectric permittivity (relative)	9	30	3	3	7.11
CB effective density of states (1/cm^3)	1.00E+19	2.50E+20	1.00E+20	2.80E+19	1.00E17
VB effective density of states (1/cm^3)	1.00E+19	2.50E+20	1.00E+20	1.00E+19	1.00E19
thermal velocity of electron (cm/s)	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7
thermal velocity of hole (cm/s)	1.00E+7	1.00E+7	1.00E+7	1.00E+7	1.00E+7
mobility of electron (cm²/Vs)	2.00E+1	5.00E+1	1.00E-4	1.00E-4	5.00E+1
mobility of hole (cm²/Vs)	1.00E+1	5.00E+1	1.00E-4	2.00E-4	5.00E+1
dopant concentration of donor N_D (1/cm^3)	1.00E+16	0	0	0	0
dopant concentration of acceptor N_A (1/cm^3)	0	1.00E+17	1.00E+18	1.00E+18	1.00E+18
Defect density Nt (1/cm^3)	0	1.00E+13	0	0	0

Each of the utilized layers has its properties like their thicknesses and their charge amount.

SCAPS-1D simulates the custom solar cell by solving the essential Poisson and electron and hole continuity equations including these properties, simultaneously. The equations are given below:

$$\vec{\nabla}.\varepsilon\vec{\nabla}\varphi = -q(p - n + N_D + -N_A-) \tag{1}$$

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

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

In these equations, $\varphi, \varepsilon, q, p, n, N_D, N_A-, \vec{J_p}, \vec{J_n}, G, R$ are the 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.

All of the needed parameters of the mentioned layers are given in Table 1.

In this study, three single, composite, and tandem configurations for the HTL were considered to compare the results. In the composite form, the thickness and dopant concentration of all samples were considered the same. In the tandem configuration, the thickness for each layer was considered as 100 nm. Moreover, in the tandem state, each case was repeated in the inverted structure, too.

The simulation was performed at the temperature of 300K and the sunlight angle of 48° with the vertical line of the earth (A.M.1.5) and light power of 1000 W/m2.

3. Results and Discussion

The simulation was done in the mentioned conditions and the results are represented in this section. The results were given in the following sections as the single, composite, tandem forms, and final I-V curve.

3.1. Comparison of HTLs in a single form

In the first step, the best candidates of the three mentioned categories for HTLs including Spiro-OMeTAD, P3HT, and Cu$_2$O were chosen and their results were compared. Results indicated that the best performance was obtained for Cu$_2$O with 19.77% efficiency compared to 19.26% and 15.92% for Spiro-OMeTAD and P3HT, respectively. Figure 2 illustrates the comparison of these three single HTLs in the perspective of photovoltaic parameters.
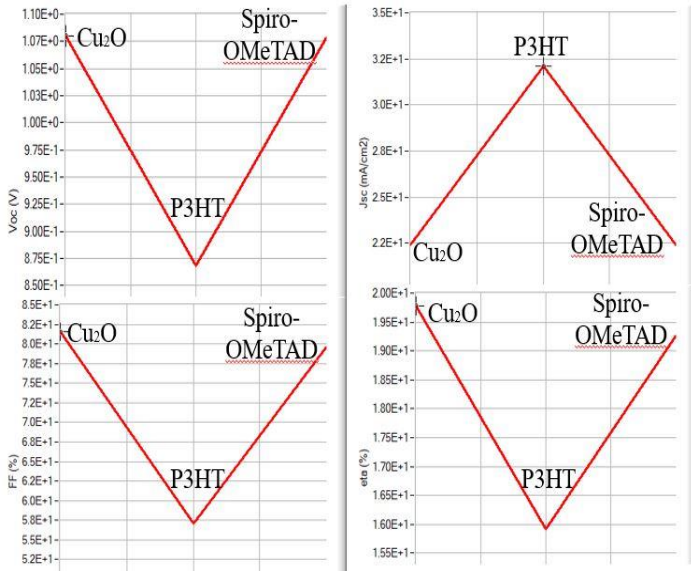
Figure 2- Photovoltaic parameters for the single HTLs

3.2. Comparison of HTLs in composite form

In this step, three composite configurations were simulated with the use of Spiro-OMeTAD, P3HT, Cu_2O as HTLs two by two with each other. In each composite including Spiro-OMeTAD/P3HT, Spiro-OMeTAD/Cu_2O, and P3HT/Cu_2O, the composition of each single HTM can be varied from 0 to 1. Here, each composite was optimized from the view of the composition of HTLs. Results represented that the optimum composition for the material in Spiro-OMeTAD/P3HT, Spiro-OMeTAD/Cu_2O, and P3HT/Cu_2O composites were 0.1/0.9, 0/1, 0.6/0.4, with the efficiencies of 24.57%, 19.77%, and 20.66%, respectively. Figure 3 (a-c) displays the variation of photovoltaic parameters with the change of composition of HTLs in each composite configuration.
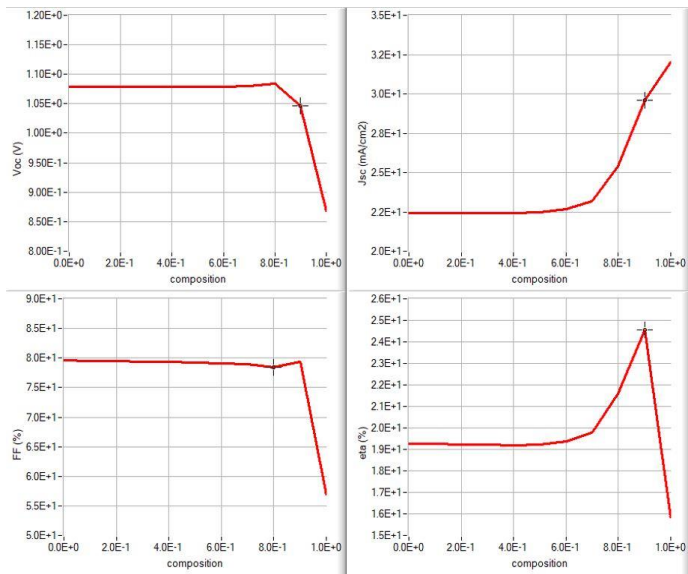

Figure 3-a. Photovoltaic parameters vs. composition of P3HT in the Spiro-OMeTAD/P3HT composite
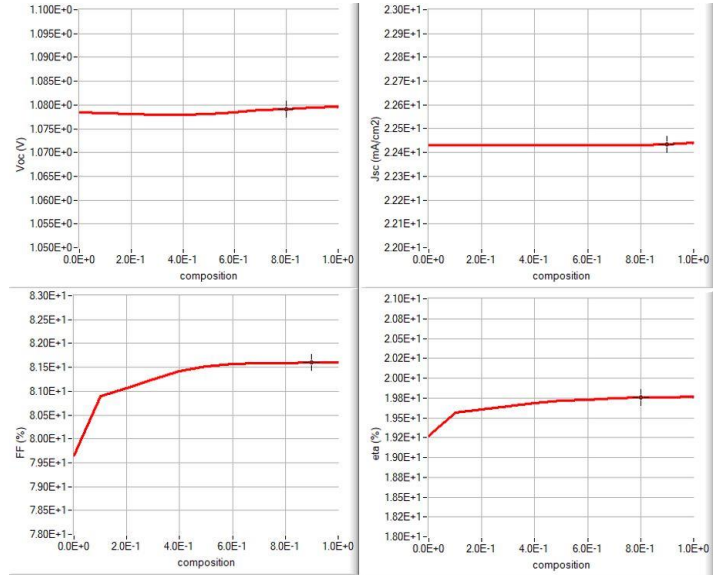

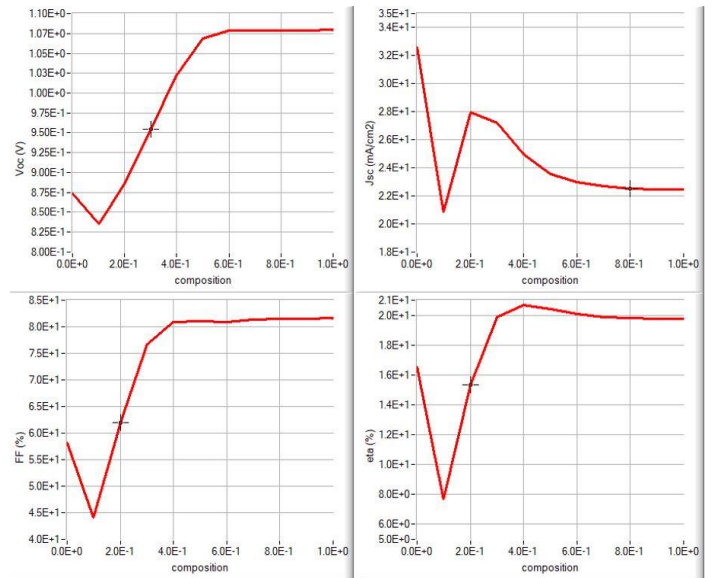*Figure 3-b. Photovoltaic parameters vs. composition of Cu_2O in the Spiro-OMeTAD/Cu_2O composite*

Figure 3-c. Photovoltaic parameters vs. composition of Cu_2O in the P3HT/Cu_2O composite

3.3. Comparison of HTLs in tandem form

In this form, the mentioned HTLs were placed in continuous and series configuration named as tandem structure. In this section, all of the possible cases of HTLs' tandem configurations were simulated and their results were investigated. The cases include Spiro-OMeTAD/Spiro-OMeTAD (or single Spiro-OMeTAD), Spiro-OMeTAD/P3HT, Spiro-OMeTAD/Cu_2O, P3HT/ Spiro-OMeTAD, P3HT/P3HT (or single P3HT), P3HT/Cu_2O, Cu_2O/ Spiro-OMeTAD, Cu_2O/P3HT, and Cu_2O/Cu_2O (or single Cu_2O) with 100 nm thickness for all HTLs (the first HTLs mentioned here, refer to the layer near the electrode). Results showed that Cu_2O/P3HT and Spiro-OMeTAD/P3HT tandem structures represent better performance with efficiencies of about 27% compared to other structures.

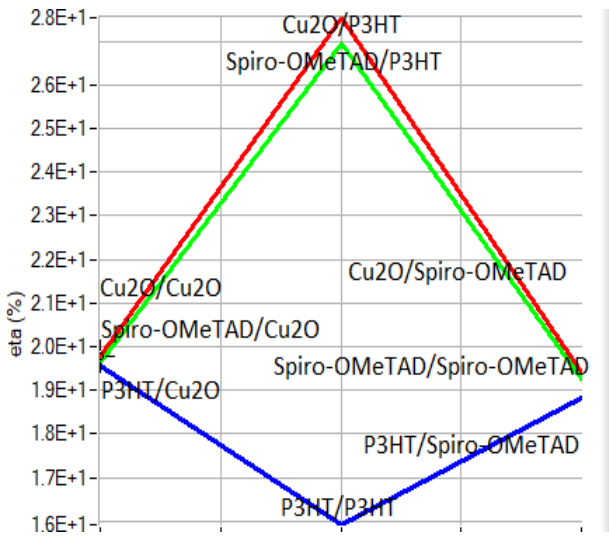Figure 4 confirms these results in the form of power conversion efficiency.

Figure 4- Efficiency of all tandem HTL structures

3.3. Final I-V curve

With the comparison of all configurations mentioned above, it can be understood that the best efficiency among all structures belongs to the type of tandem HTL cells that the P3HT layer was placed between the upper HTL near the electrode and the absorber layer. This could be because of the structural modification role of P3HT layer. However, these implementations were demonstrated in figure 5 (a,b) and table 2. The results were in good agreement with experimental works, too [8-11].
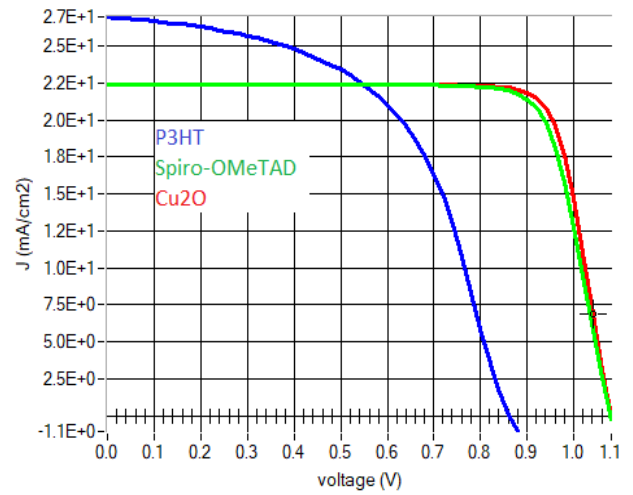

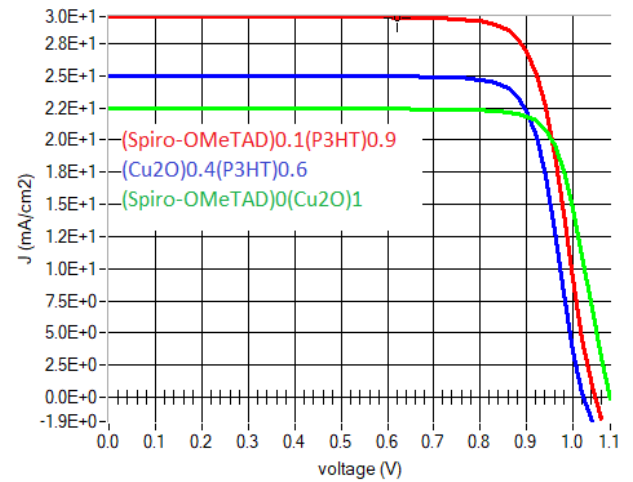*Figure 5-a. Current-density (I-V) curve of different HTLs in the single configuration*

Figure 1-b. Current-density (I-V) curve of different composite HTLs

Table 2- Photovoltaic parameters of configurations used for HTLs

configuration	HTL	V_{OC}		J_{SC}		FF		PCE	
simple	Spiro-OMeTAD	1.08 (SCAPS)	0.98 (exp)	22.43 (SCAPS)	21.20 (exp)	79.63 (SCAPS)	77.60 (exp)	19.26 (SCAPS)	18.7 (exp)
	P3HT	0.87 (SCAPS)	0.80 (exp)	32.09 (SCAPS)	19.20 (exp)	57.09 (SCAPS)	73.30 (exp)	15.92 (SCAPS)	15.4 (exp)
	Cu_2O	1.08 (SCAPS)	0.99 (exp)	22.44 (SCAPS)	19.70 (exp)	81.60 (SCAPS)	75.00 (exp)	19.77 (SCAPS)	18.4 (exp)
composite	(Spiro-OMeTAD)$_{0.1}$(P3HT)$_{0.9}$	1.05		29.59		79.37		24.57	
	(Spiro-OMeTAD)$_0$(Cu_2O)$_1$	1.08		22.44		81.60		19.77	
	(Cu_2O)$_{0.4}$(P3HT)$_{0.6}$	1.02		25.00		80.84		20.66	
tandem	Spiro-OMeTAD/P3HT	1.09		31.47		78.37		26.97	
	P3HT/Spiro-OMeTAD	1.08		22.43		77.86		18.81	
	Spiro-OMeTAD/Cu_2O	1.08		22.44		81.03		19.63	

4. Conclusions and Recommendations

Recently, the usage of newly developed perovskite solar cells has experienced rapid improvements because of their tremendous features. Perovskite solar cells consist of several layers that each one has its specific role in the cell structure. In this study, the hole transport layer with different configurations in a perovskite-based solar cell was investigated. Results represented that developing new structures like composite forms and tandem configurations, significantly lead to efficiency increment in the cell. The best structure for the custom cell used in this study obtained about 27% for the Cu_2O/P3HT tandem structure.

5. Acknowledge

6. Funding

7. 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 Ass. Prof. Dr. Nagihan Delibaş.

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