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Improved Perovskite Solar Cell Performance Using Semi-transparent CNT Layer

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Abstract:
In this paper, the effect of using semi-transparent Carbon nanotube layer (CNT) on the efficiency of perovskite solar cell (PSC) is investigated. One of the most important process in PCS is charge collecting. In this regard, Carbon nanotubes have the ability to act as charge collector layer in solar cell. Carbon nanotubes, due to suitable optical and electrical properties such as transparency, high mobility and stability have been widely used in solar cell structures. In the proposed structure, we use semi-transparent CNT layer as charge collector on top of PSC. This layer with low resistance path for transport charge carriers has increased short circuit current and other performance parameters of solar cell. The proposed device structure ITO/CNT/TiO2/CH3NH3PbI3/Spiro-OMeTAD is simulated with Silvaco TCAD. The simulation results show that the efficiency of the perovskite solar cell with semi-transparent CNT layer is reached 23.59% which is 3.15% higher than simple perovskite solar cell structure under AM1.5G.

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1. INTRODUCTION

Solar energy as renewable energy is one of the most abundant types of energy in the world and it is a promising alternative for fossil fuels. Therefore, Solar cells play an effective role in converting solar energy into electricity to deal environment challenge and global energy consumption. Many studies have been performed on the structures and materials of solar cells to achieve three objectives: high efficiency, low cost and long-lifetime. Due to special properties of perovskite, such as appropriate band gap [1], high absorption coefficient [2], low exciton binding energy [3], and long diffusion length of carriers [4, 5], Researchers have focused on perovskite solar cells (PSCs). Recently PSCs have shown remarkable results and low cost solutions for solar cell deployment.

In planar architecture PCSs, perovskite as active layer is located between two layers, hole transport layer (HTL) and electron transport layer (ETL). After perovskite absorbed sun-light, three steps occur in solar cell: (1) in perovskite layer excitons are generated and dissociated in interface of perovskite and transport layers, (2) electrons and holes caused by the dissociation of excitons move towards the ETL and HTL layers, (3) Electrodes lead the collected electrons and holes to the external circuit to produce current. In this process, for better efficiency, a low resistance path for charge transport should be provided. To achieve this aim, the researchers have changed layers or added materials to the conventional structure of the solar cell [6, 7]. One of these materials is Carbon nanotube (CNT).

CNTs have attracted attention researchers because of their application properties such as excellent conductivity, high optical transparency and thermal stability [8]. In this regard, most of the research is focused on the use of carbon nanotubes in solar cells as charge collector due to their high mobility and absorption of more photons by preventing their scattering on the surface of the solar cell [9-10].

CNTs have also been used to improve performance of perovskite solar cells. In most of these applications, CNTs act as additives to complete charge extraction, improve solar cell stability, and in some cases, act as charge collectors.

CNTs have also been widely used in different applications in PSCs. As one of the first studies, Li et al. used CNT network as HTL to replace the metal electrode [11], in another study double-walled CNT as transparent electrode contributed to high device performance PSC [12], Habisreutinger et al. added CNT as doping in Spiro-OMeTAD layer and enhanced hole extraction of device [13], Zhang et al. added sulfonate-carbon nanotube and improved performance of PSCs by enhanced the grain size of perovskites [14], Aitola et al. increased the stability of PSC by combining single-walled nanotubes (SWCNT) and Spiro-OMeTAD as HTL and electrode [15]. In other ways, the use of CNT in perovskite solar cells has been mentioned [16]. Table 1 summarize some of these studies.

TABLE 1
SUMMERY OF THE REPORTED FUNCTION OF CNTs IN PSCs STRUCTURES

CNT Function	Device Structure	Reference
Metal Electrode	TiO2/Perovskite/Spiro-OMeTAD/CNTs	11
Transparent Electrode	ITO/DWCNTs/PTAA/Perovskite/C60/BCP/Cu	12
HTL	FTO/TiO2/Perovskite/P3HT/SWCNTs/Spiro-OMeTAD/Ag	13
Added in Perovskite layer	FTO / c-TiO2 / mp-TiO2 /Perovskite+CNTs / Spiro-OMeTAD/ Ag	14
HTL and Electrode	FTO / c-TiO2 / mp-TiO2 /Perovskite / SWCNTs+ Spiro-OMeTAD/ Ag	15

In this study, we propose different structure perovskite solar cell by using semi-transparent CNT layer. In order to absorb surface current effectively and increase the performance of perovskite solar cell, we add a thin semi-transparent CNT layer on top of solar cell. This layer makes low resistance path for surface current due to high mobility and improves performance of solar cell. We simulate proposed structure by using Silvaco TCAD and evaluate solar cell performance parameters such as: short circuit current, open circuit voltage, fill factor and efficiency to show the effect of CNT layer. In section 2, the structure of perovskite solar cell with CNT layer is shown. In section 3, theoretical model for simulation is described. Section 4 is dedicated to show simulation results and finally conclusion is given in section 5.

2. STRUCTURE OF PEROVSKITE SOLAR CELL WITH CNT LAYER

Fig 1 presents proposed schematic structure of perovskite solar cell with CNT layer. The simulated solar cell planar structure is CNT/TiO2/Perovskite/Spiro-OMeTAD, where Methylammonium lead iodide (CH3NH3PbI3) is organic material as perovskite with a thickness of 500nm, TiO2 is inorganic material with a thickness of 100nm as ETL and Spiro-OMeTAD is organic material with a thickness of 200nm as HTL. For improving the electrical characteristics of device, thickness of perovskite, ETL and HTL layers are optimized [17]. The details of the parameters related to the used layers are shown in Table 2.

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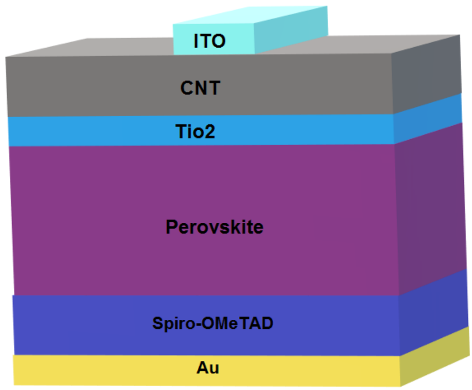

Fig. 1. Proposed schematic structure for perovskite solar cell.

TABLE 2
LAYER PARAMETERS

Layer	Material	Thickness(μm)	Doping(cm3)
Electrode	ITO	0.1	_
Semi transparent	CNT	0.2	_
Electron transport	Tio2	0.1	N type-3e19
Absorber	CH3NH3PbI3	0.5	P type-1e13
Hole transport	Spiro-OMeTAD	0.2	P type-1e18
Anode	Au	0.1	_

Carbon materials are used in four categories in the structure of perovskite solar cells, which include graphene (GR) and its derivatives, CNTs, fullerene and its derivatives, and the fourth category is carbon black and graphite as some conductive carbon materials [18].Among them , Carbon nanotubes, were discovered in 1991 , have attracted many researchers attention because of their unique properties and potential applications. CNTs are cylindrical structures that consist of rolled-up graphene sheets in different number of walls. Chirality and Diameter of nanotubes determine metallic or semiconductor CNTs properties. It can be said that the most interesting property of SWCNTs is huge carrier mobility, which is more than crystalline silicon. They also have a maximum current density that makes them comparable to similar metals used as conductors. The chirality of CNTs cannot be guaranteed during production. Random arrays of nanotubes on the substrate affect the transparency of CNT network. Recently, several types of semi-transparent CNT thin film were prepared and studied that High electron mobility and optical transparency make them ideal options for use in solar cell devices [19].

3. THEORETICAL MODELING

In both form of organic semiconductors (molecular and polymeric semiconductors) the overlap of electron wave functions of neighboring atoms leads to two orbitals: The Highest Occupied Molecular Orbital (HOMO) and the Lowest Unoccupied Molecular Orbital (LUMO). The gap between the HOMO and LUMO defines the excitation boundary of the molecule and determine the amount of solar spectrum absorption. The absorbing light and converting into electricity process in organic semiconductor include four steps: exciton generation, exciton diffusion, exciton dissociation into free charge-carriers and finally charge transport to the electrodes. Each of these steps affects the efficiency of organic solar cells.

Drift diffusion model can be used to describe carrier dynamics in organic solar cells. This model involve following equations: the current continuity equations for electron, hole and exciton and the Poisson's equation for the electrostatic potential (φ). The equation shown below is the Poisson equation:

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

Where ε_0 and ε_r are vacuum permittivity and relative dielectric constant. p is hole and n is electron concentrations. q is the elementary charge.

The following equations are the electron, hole and exciton continuity equations, respectively [20].

$$\frac{\partial n}{\partial t} = D(E,x) - R(n,p) - \frac{1}{q}\nabla\left[qn\mu_n \nabla\varphi - K_B T \mu_n \nabla n\right] \tag{2}$$

$$\frac{\partial p}{\partial t} = D(E,x) - R(n,p) - \frac{1}{q}\nabla\left[-qp\mu_p\nabla\varphi - K_B T\mu_p\nabla p\right] \tag{3}$$

$$\frac{\partial x}{\partial t} = G(r) + \frac{1}{4}R(n,p) - R(x) - D(E,x) - \frac{1}{q}\nabla\left[-K_B T\mu_x\nabla x\right] \tag{4}$$

where x is exciton concentration. μ_n, μ_p, and μ_x are mobility of electron, hole and exciton. T and K_B are the temperature the Boltzmann constant respectively. In equations (2) and (3), the first gradient term indicates the drift of charge due to local electric field and the second gradient term indicates the diffusion of carries. The excitons photogeneration is determined by $G(r)$ [21]. Dissociation of excitons and generation charge carriers are described by Onsager's theory and following equation [22]:

$$D(E,x) = xN_f\int_0^\infty K_D(E,a)F(a)da \tag{5}$$

Where $K_D(E,a)$ is given by Braun as follows [23]:

$$K_D(E,a) = K_R\frac{3}{4\pi a^3}e^{-\frac{\nabla E}{K_B T}}(1 + b + \frac{b^2}{3} + \frac{b^3}{18} + ...) \tag{6}$$

Where K_R and b are defined by $K_R = q\langle\mu\rangle\frac{\varepsilon_0}{\langle\varepsilon_r\rangle}$ and $b = \frac{q^3|E|}{8\pi\langle\varepsilon\rangle K_B^2 T^2}$ and ∇E is exciton binding energy. $F(a)$ is the distribution function of acceptor-donor separations, and N_f is a normalization factor for the function [22].

One of the characteristics of organic materials is low mobility of free carriers. In this case, Langevin gave the theory for free charge carriers recombination which is denoted by $R(n,p)$ in above equations. $R(n,p)$ is given by [24]:

$$R_l = \frac{q(np - ni^2)}{\varepsilon}(\mu_n + \mu_p) \tag{7}$$

Refer to (4), $R(x)$ is exciton recombination and define by $R(x) = (\frac{x}{\tau_x})$.

Where τ_x is the an exciton average lifetime . According to the structure of the electronic band CNT, the statistics of the carriers are written as the following equation [25]:

$$p - n = sign(E_D - E_f)\frac{1}{\pi\hbar^2 vf^2}(E_f - E_D)^2 \tag{8}$$

In the above relation , E_D is the Dirac point , n is the electron density and p is the hole density in the CNT layer. E_f is the Fermi level of the CNT sheet, Besides vf and $\hbar$ are the Fermi velocity in the CNT and the reduced Planck's constant respectively.

By combining current continuity equation and drift-diffusion equations in heterostructure CNT/Tio2 and solving the above equations, various parameters can be determined to evaluate the solar cell performance. For solar cell, the I–V characteristics can be described as [26]:

$$I = I_l + I_0(\exp(\frac{qv}{N_{IF}KT}) - 1) \tag{9}$$

Where I_0 is the saturation current, I_l is short circuit photocurrent (Under short-circuit condition $I_l = I_{sc}$) and the junction ideality factor is defined by N_{IF} .

Therefore, the pn-junction voltage is given by:

$$V = V_T \ln(1 + \frac{I_l - I}{I_0}) \tag{10}$$

the open-circuit voltage(V_{oc}) is defined by:

$$V_{oc} = V_T \ln(1 + \frac{I_l}{I_0}) \tag{11}$$

The efficiency (η) of solar cell is given by:

$$\eta = \frac{P_m}{P_{in}} * 100\% \tag{12}$$

where P_m is the maximum power output and P_{in} is input power from incident light. At the maximum power point per unit area, the voltage and current are determined by I_{mp} and V_{mp} respectively and the maximum power output is $P_m = I_{mp}V_{mp}$. Another parameter of solar cell performance is the Fill Factor (FF) which defined as:

$$FF = \frac{P_m}{V_{oc}I_{sc}} = \frac{V_m I_m}{V_{oc}I_{sc}} \tag{13}$$

Finally, the efficiency is therefore defined as:

$$\eta = \frac{P_m}{P_{in}} = \frac{FF V_{oc} I_{sc}}{P_{in}} \tag{14}$$

4. SIMULATION AND RESULTS

The aim of this study was to design and simulate perovskite solar cell with CNT layer and investigate the effect of CNT layer on collecting the surface current of perovskite solar cell. Then the performance of a simple perovskite cell was compared with a designed cell via using Silvaco TCAD.
Among the ways to increase solar cell efficiency is to increase the absorption of the solar cell surface current. According to this aim, a low-resistance pathway is required to collect charge. Therefore, the structure of a solar cell is modified by adding a layer with low resistance on top surface of the cell. But remarkable point is the transparency of this layer, without this ability, light absorption is impaired and efficiency is decreased. CNT layer, with high mobility and transparency, is one of the candidates for this purpose. Institute for Micro Structural Science of Canada has fabricated CNT layer with these specifications and this semi-transparent layer with sheet resistance of 128Ω/□ is used in proposed structure. The mentioned CNT structure is heterogeneous network of nanotubes which are placed randomly, 1/3 of them are metallic, while the remaining 2/3 are semiconductors. For modeling this layer, 4H-SiC from library of Silvaco was selected as basic material and Some material parameters, such as mobility and band gap, were set to achieve the experimental layer [27]. The input parameters that we used for simulation are given in Table 3.

TABLE 3
MATERIAL PARAMETERS USED IN NUMERICAL SIMULATION[17, 25]

Material properties	CNT	Tio2	CH3NH3PbI3	Spiro-OMeTAD
E_g (eV)	0.026	3.2	1.55	3
ε_r (Fcm^{-1})	5.4	19	100	3
μ_n ($cm^2\ V^{-1}\ s^{-1}$)	8138.2	0.2	1	2e-4
μ_h ($cm^2\ V^{-1}\ s^{-1}$)	8138.2	0.1	1	2e-4
N_c (cm^{-3})	3e17	2.2e18	2e18	2e18
N_v (cm^{-3})	3e17	2e19	2e19	1.9e19

In order to simulate, silence index and refractive index (n, k) for CH3NH3PbI3 layer are extracted from [28] and for Spiro-OMeTAD layer are extracted from [29].

One of the ATLAS software capabilities is access to different recombination and mobility models for organic and inorganic materials, so simulation results are closer to the experimental results. In this work, for organic layers (CH3NH3PbI3 and Spiro-OMeTAD) Langevin recombination and Poole-Frenkel mobility models are considered. For inorganic layers, Shockley-Read-Hall (SRH), Optical recombination (OPTR), Auger and concentration mobility model are used.

The AM1.5 spectrum represents the standard beam which is useful for evaluate the performance of solar cells relative to each other. Any layer placed on the surface of solar cell can absorb the amount of light energy and reduce the power of emitted light on solar cell, so transparency of the upper layers is important parameter. To model CNT semi-transparent layer, it is assumed that this layer is completely transparent and the effect of reducing emitted light power is modeled by changing AM1.5 spectrum[25]. Fig. 3 shows standard beam Am1.5 and alternative beam used in simulation after passing through the CNT layer.

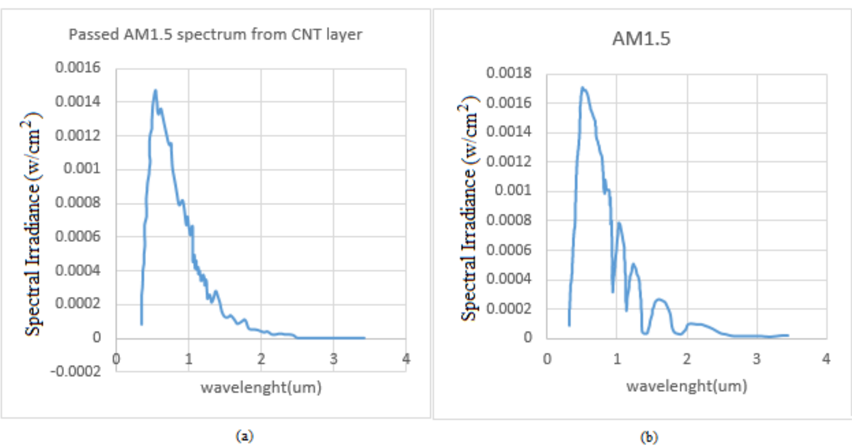

Fig. 2. (a) The alternative beam used in simulation, (b) The standard AM1.5 beam

The I-V curves of simple perovskite solar cell and CNT-based perovskite solar sell are shown in Fig.3. By comparing characteristics, the effect of CNT layer as low resistance for carriers on increasing short circuit current is determined.

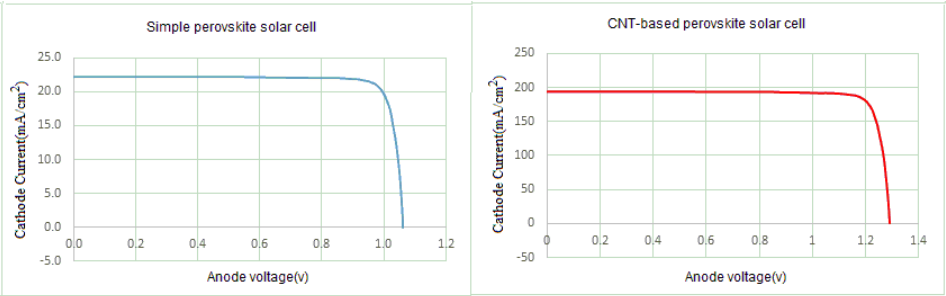

Fig. 3. I-V characteristics for simple and CNT based perovskite solar cell.

Fig.4 shows the effect of CNT layer as charge collector on increasing surface current solar cell. CNT-based solar cell has higher current values than simple perovskite solar cell. Other extracted performance parameters are presented in Table 4.

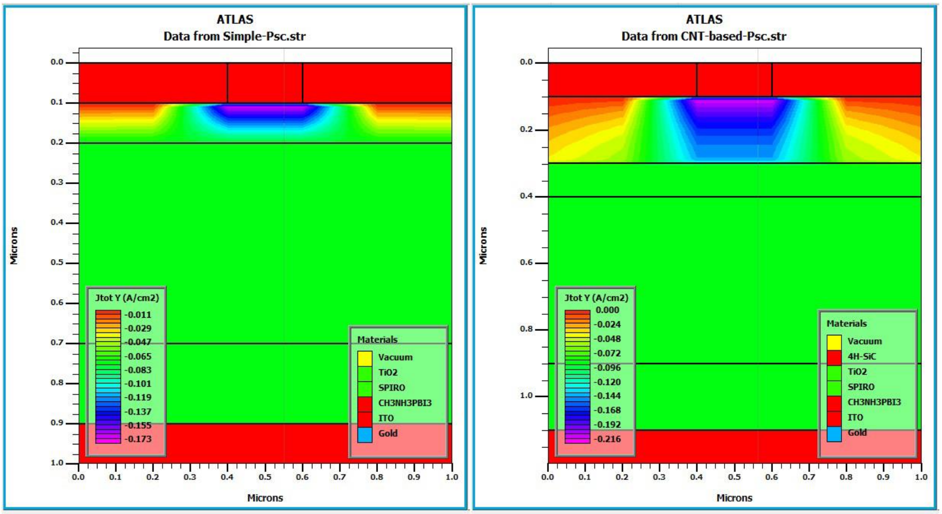

Fig. 4. Total current density in simple perovskite solar cell and CNT based perovskite solar cell.

TABLE 4
COMPARING THE PERFORMANCE PARAMETERS OF THE PROPOSED STRUCTURE WITH SIMPLE PEROVSKITE SOLAR CELL

Solar Cell	V_{oc} (v)	I_{sc} (mA/cm^2)	FF	E_{ff}
Simple perovskite solar cell	1.06	22.17	86.83	20.44%
CNT-based perovskite solar cell	1.29	194.88	87.05	23.59%

As shown in the results of table 3, the efficiency has increased from 20.44% to 23.59% and also other parameters have improved. Another advantage of CNT layer, which is a path with less resistance for charge carriers to reach the upper contact, is the reduction of shading losses due to metal grid on the solar cell surface. Actually, the cell width can be increased compared to constant contact width and the percentage of shading can be gradually reduced[25]. When the more area of the solar cell absorbs light, the more charge carriers are generated, therefore the solar cell efficiency can be increased.

5. CONCLUSION

In this paper, the structure of perovskite solar cell has been proposed by adding CNT layer as charge collector. This proposed structure simulated with Silvaco TCAD. Then to evaluate the effect of CNT layer on solar cell performance parameters, we compared the proposed solar cell simulation results with the simple perovskite cell simulation results. Due to the high mobility of CNT layer, charge carriers pass through less resistance path to reach the solar cell surface, thus the short circuit current in the cell is increased. Simulation shows that the efficiency of perovskite solar cell has improved from 20.44% to 23.59%. Other performance parameters, including open circuit voltage and fill factor have also increased.

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