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# Performance Investigation of a Perovskite Solar Cell with $TiO_2$ and One Dimensional ZnO Nanorods as Electron Transport Layers

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**Abstract:** Organic-inorganic halide perovskite thin film perovskite solar cells are gaining much attention, in recent years. Designing proper electron transport layer (ETL) and hole transport layer (HTL) with high quality to achieve devices with higher efficiency are fundamental. One dimensional (1D) nanostructures are newly introduced materials with high mobility and low recombination rate, which may improve the device performance. In this paper, 1D ZnO nanorods (ZnO-NRs) as well as planar $TiO_2$ are considered as the ETL of the device and their electrical performance are compared with different HTL materials in Sn- and Pb- perovskite. In addition, impact of critical design parameters including absorber thickness, interface defect density, back contact electrode materials on the performance of the device are comprehensively assessed. In this work, the simulations have been carries out using a 1D Solar Cell Capacitance Simulator (SCAPS-1D). The results show that in Sn- perovskite, ZnO-NRs has superior performance in comparison with $TiO_2$ with maximum photon conversion efficiency (PCE) of 16.7 % and short circuit current density of 30.21(mA/cm2). However, in terms of Pb-perovskite planar $TiO_2$ has given the best performance with PCE of 19.6%. The results in this paper pave the way for introducing inexpensive high performance solar cell.

**Keywords: Perovskite Solar Cell, Electron Transport Layer, Hole Transport Layer, Efficiency, SCAPS-1D, Nanorods.**

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## 1. INTRODUCTION

Basically, solar photovoltaic is a promising technology for large scale renewable energy harvesting [1-4]. Organic-inorganic halide perovskite solar cells are, undoubtedly, the ever-increasing branches of thin film photovoltaic devices in the recent years [5-7].

In a typical solar cell structure, perovskite layer is the middle layer, which absorbs light across almost all visible wavelength. When the cell is illuminated by the sun beams, electron-hole pairs are created in the intrinsic perovskite layer. Following that, carriers are extracted by the electron transport layer (ETL) and hole transport layer (HTL) and finally, the charges are collected at the top and back electrodes [8-12]. The quality of ETL/HTL interface with the absorber layer and energy band offset at the heterojunctions are critical parameters that may affect feasibility of the photovoltaic device. Research in designing and fabricating ETL/HTL active layers with high quality to achieve devices with higher efficiency is still under study. Spiro- MeOTAD (Spiro- OMeTAD) is one of the most studied and appropriate organic HTL material [13]. However, its high cost as well as instability in light, heat and humidity has limited the industrial development of perovskite solar cell. It is evident that alternative materials should be introduced for HTL, which are inexpensive and have high stability under different ambient conditions [14].

Moreover, choosing appropriate ETL materials are significant for efficient charge collection. The proper ETL material should have high enough band gap in the UV and visible light spectrum so that photon can travel through it easily and can be absorbed by the absorber layer. Up to now, Titanium dioxide ($TiO_2$) and Tin(IV) oxide ($SnO_2$) have been employed as n-type ETL materials in high efficiency photovoltaic devices [15,16,17]. Basically, as Tin has +4 oxidation state (IV), and oxidation state of oxygen is −2, so there must be two oxygen atom so that neutral compound can be balanced.

Recently, one-dimensional nanorods (NRs) ETL materials are introduced, which provide rapid carrier transport in solar cell. The application of nanorods in perovskite solar cells may enhance the device efficiency by reducing the carrier recombination losses and by diminishing decoupling of light propagation in orthogonal directions [18,19]. Basically, ZnO nanorods with vertical alignment are fabricated via a multi-annealing process in reducing ambient [20]. ZnO thin film is deposited on the FTO substrate. The deposition process is performed in an Argon ambient. Following the deposition procedure, ZnO thin film is placed on a quartz boat set in the middle part of the furnace. The gas $H_2$ (Hydrogen) and $N_2$ (Nitrogen) are inserted to the furnace. The oxygen vacancies are produced due to the effect of reducing ambient and they increase the conductivity of ZnO nanorods. However, the oxygen vacancies on the surface of ZnO nanorods may increase the recombination rate of carriers. The solution is to coat $TiO_2$ thin layer on the fabricated ZnO nanorods by the chemical vapour deposition method to prevent the recombination rate of carriers and improve the stability of the structure [21]. It is worth to notify that ZnO-NRs is one of the most important nanomaterials that due to its relatively high conductivity, high electron mobility, and low cost [17], provides excellent optical properties. In addition, besides Spiro-MeOTAD, new inexpensive abundant non-toxic inorganic materials including Cuprous oxide ($Cu_2O$) [22-24], Cupric oxide (CuO) [24], Copper(I) Thiocyanate (CuSCN) are introduced as novel HTL materials to design a photovoltaic device with superior efficiency.

In this paper, a comprehensive investigation has been carried out via two different ETL structures, planar Titanium dioxide ($TiO_2$) and 1-Dimensional (1D) Zinc oxide nanorods (ZnO-NRs) to assess their impact on the fundamental solar cell characteristics including short circuit current density ($J_{SC}$), open circuit voltage ($V_{OC}$), fill factor (FF) and photon conversion efficiency (PCE). Numerical simulations are carried out for two different common absorber materials including Sn- and Pb- based perovskite, $CH_3NH_3Pb(Sn)I_3$. In addition, different materials are considered for the back contact electrode to investigate how the work function of electrode materials affects energy band bending at the interface of back contact and HTL.

The paper is organized as follows: simulation design parameters and material properties are precisely presented in section two. Next, the effect of different physical and structural design parameters on the critical performance metrics of the cell are comprehensively discussed in section three. Finally, the paper is summarized in section four.

## 2. Device structure and Simulation Set up

The schematic of the proposed structures with $TiO_2$ and ZnO-NRs as ETL materials are illustrated in Fig. 1. Numerical simulations are carried out with SCAPS-1D and initial parameters for the materials are summarized in Table.1. The energy band diagram of the materials that are considered in this study are presented in Fig.2.

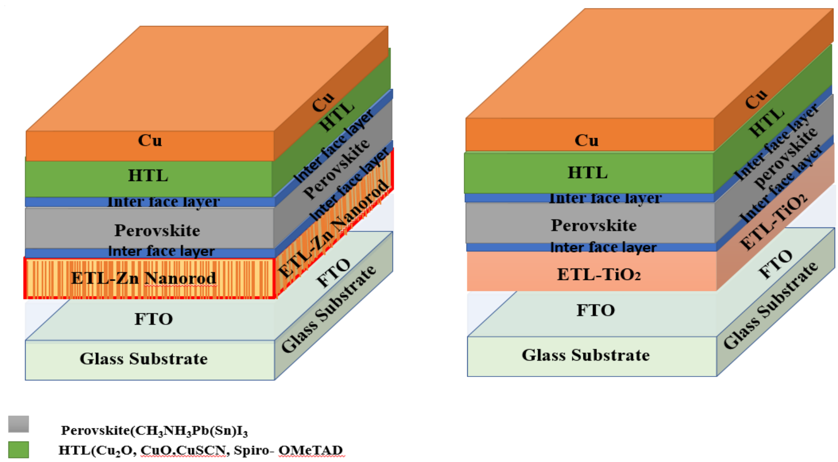


**Fig.1**: Schematic of the perovskite solar cell with $TiO_2$ and ZnO-NRs as the ETL materials.

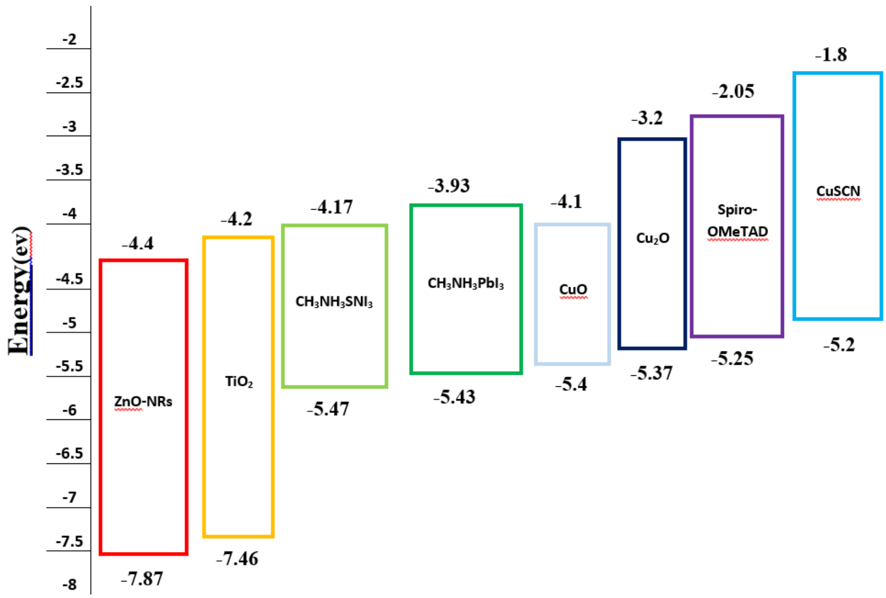


**Fig.2**: Energy band diagram of the materials that are considered in this study.

The ETL material plays an important role in the solar cell to extract electrons from the perovskite layer and to block recombination of carriers. Basically, $TiO_2$ is one of the most widely used ETL materials with excellent stability and unique optoelectronic properties. This material has a wide band gap (3.2eV) and provides high efficiency. $TiO_2$ can crystallize with different phases of rutile, anatase, and brookite which each of them exhibits different band gaps. In this study, anatase form of $TiO_2$ has been employed. Due to the proper optoelectronical properties such as wider band gap and compatible energy level versus perovskite [25], we consider the anatase phase for the simulation. Moreover, ZnO is a low cost direct wide band gap material (3.37eV) and has specific electrical and optical properties. It is important to mention that ZnO-NRs are 1D nanostructures that have exciting chemical and physical properties. This material provides higher mobility along the rods, higher minority carrier lifetime as well as less recombination rate, which makes it attractive for optoelectronic applications.

**Table.1**: Material properties that are used for SCAPS-1D simulation [17]

| Parameters | ZnO NRs | $TiO_2$ | $CH_3NH_3SnI_3$ | $CH_3NH_3PbI_3$ | CuO | $Cu_2O$ | Spiro- OMe TAD | CuSCN |
|---|---|---|---|---|---|---|---|---|
| Thickness(nm) | 500 | 300 | 450(Var) | 450(Var) | 300 | 350 | 350 | 350 |
| Bandgap (ev) | 3.47 | 3.2 | 1.3 | 1.50 | 1.51 | 2.17 | 3.2 | 3.4 |
| Electron effinity (ev) | 4.3 | 4.2 | 4.20 | 3.9 | 4.07 | 3.2 | 2.1 | 2.19 |
| Dielectric Permittivity (relative) | 9 | 10 | 10 | 10 | 18.1 | 7.1 | 3 | 10 |
| CB effective density of states (1/cm3) | 2E+18 | 2E+17 | 1E+18 | 2.25E+18 | 2.2E+19 | 2.5E+18 | 2.5E+18 | 2.5E+18 |
| VB effective density of states (1/cm3) | 1.8E+20 | 6E+17 | 1E+18 | 1E+18 | 5.5E+20 | 1.8E+19 | 1.8E+19 | 1.8E+19 |
| Electron thermal velocity (cm/s) | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 |
| Hole thermal velocity (cm/s) | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 | 1E+7 |
| Electron Mobility (cm2/Vs) | 1E+2 | 1E+2 | 1.6E+0 | 2.2E+0 | 1E+2 | 2E+2 | 2E-4 | 2E-4 |
| Hole Mobility (cm2/Vs) | 2.5E+1 | 2.5E+2 | 1.6E+0 | 2.2E+0 | 1E-1 | 8E+2 | 2E-4 | 1E-2 |
| Shallow uniform acceptor density,NA(1/CM 3) | 0 | 0 | 3.2E+15 | 1E+18 | 1E+16 | 9E+21 | 1E+20 | 1E+18 |
| Shallow uniform doner density ,ND(1/cm3) | 1E+17 | 1E+17 | 0 | 0 | 0 | 0 | 0 | 0 |
| Defect type | Neutral | Neutral | Neutral | Neutral | Neutral | Neutral | Neutral | Neutral |

Hole transport materials are also fundamental for efficient photo generated charge extraction in perovskite solar cells as well as for achieving high power conversion efficiency. Accordingly, for assessing the main features of the proposed photovoltaic devices, impact of different materials for the HTL are thoroughly investigated. Their properties are summarized as follows: Cuprous oxide ($Cu_2O$) is a direct bandgap semiconductor with a bandgap energy of nearly 2.1eV and can be considered as a promising material for absorber layer of thin-film solar cells. This material is intrinsically p-type without the need for premeditated doping and have low cost fabrication methods. Copper(II) oxide or cupric oxide (CuO) is a natural p-type semiconductor with band gap energy of 1.5 eV. It is an abundant material and relatively non-toxic with high optical absorption. Two atoms of copper (Cu) react with an oxygen molecule to form 2 units of copper oxide (CuO). Since the net charge of the ionic compound must be zero, the Cu ion has a +2 charge. The sign II indicates +2 copper ion. Copper(I) thiocyanate (CuSCN) is a p-type wide band gap semiconductor material (3.6 eV) that behaves as an efficient HTL material for a variety of active layer materials, including from polymers or small molecules to hybrid perovskite solar cells. The sign I indicates +1 copper ion. In addition, CuSCN is a commercially available chip material. Finally, Spiro-MeOTAD has been demonstrated as an efficient HTL material and has widespread application in perovskite solar cells.

## 3. RESULTS AND DISCUSSIONS

The current density (J)-voltage (V) characteristics of the Sn- and Pb-perovskites are illustrated in Fig. 3 (a)-(d) for different HTL materials as well as for $TiO_2$ and ZnO-NRs as the ETL of the device. The critical cell parameters (Jsc, Voc, FF and PCE) are obtained from the J-V characteristics, summarized in Table.2. The results demonstrate that in Pb- perovskite, ZnO-NRs provides higher Jsc in comparison with $TiO_2$, due to the higher mobility of carriers along the rods. However, for Sn- perovskite, the short circuit current density has been considerably improved owing to the reduction of energy difference between the lowest unoccupied molecular orbital (LUMO) and conduction band of the ETL materials.

**Table.2**: Solar cell critical electrical parameters for Sn- and Pb- perovskites.

| **Pb-Perovskite** | | | | | |
|---|---|---|---|---|---|
| **ETL** | **HTL** | **Jsc (mA/cm$^2$)** | **Voc (V)** | **FF (%)** | **PCE%** |
| **ZnO-NRs** | **CuO** | **18.90** | **0.71** | **75.60** | **10.21** |
| | **CuSCN** | **21.76** | **0.86** | **65.56** | **12.53** |
| | **$Cu_2O$** | **21.93** | **0.86** | **83.30** | **15.83** |
| | **Spiro-MeOTAD** | **21.66** | **0.86** | **80.82** | **15.17** |
| **$TiO_2$** | **CuO** | **17.80** | **0.79** | **75.30** | **14.24** |
| | **CuSCN** | **20.79** | **0.84** | **95.65** | **16.7** |
| | **$Cu_2O$** | **20.90** | **0.97** | **85.55** | **17.46** |
| | **Spiro-MeOTAD** | **20.62** | **0.99** | **95.57** | **19.6** |
| **Sn-Perovskite** | | | | | |
| **ETL** | **HTL** | **Jsc (mA/cm$^2$)** | **Voc (V)** | **FF (%)** | **PCE%** |
| **ZnO-NRs** | **CuO** | **28.69** | **0.69** | **53.26** | **10.58** |
| | **CuSCN** | **29.35** | **0.83** | **49.07** | **12.02** |
| | **$Cu_2O$** | **30.19** | **0.84** | **63.62** | **16.05** |
| | **Spiro-MeOTAD** | **29.79** | **0.83** | **61.58** | **15.29** |
| **$TiO_2$** | **CuO** | **28.65** | **0.68** | **53.33** | **10.47** |
| | **CuSCN** | **29.40** | **0.83** | **48.95** | **12.01** |
| | **$Cu_2O$** | **30.25** | **0.83** | **63.46** | **16.04** |
| | **Spiro-MeOTAD** | **29.86** | **0.83** | **61.44** | **15.28** |

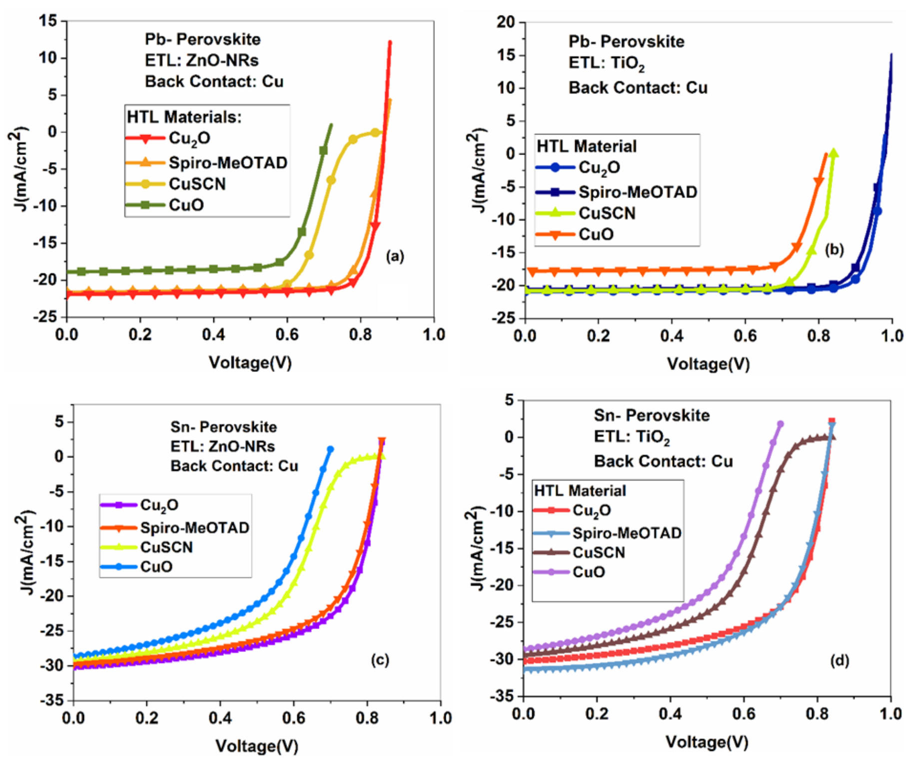


**Fig.3**: Current-Voltage characteristics of (a), (b) Pb-Perovskite and (c), (d) Sn-Perovskite absorber layer for different ETL and HTL materials.

The photon conversion efficiency (PCE) of solar cell (expressed as a percentage) with different HTL and $TiO_2$ and ZnO-NRs as ETL are thoroughly calculated. The simulations are carried out for two different categories of Sn- and Pb – based perovskites and the results are illustrated in Fig.4. It is evident for Sn-based perovskite that ZnO-NRs has a slight higher efficiency in comparison with $TiO_2$. It is observed that $CU_2O$ provides the highest efficiency among the other HTL materials with PCE of 16.05%. Basically, the energy band difference between perovskite and the ETL/HTL layers effectively modify the carrier transport. However, it seems that a distinct difference exists between efficiency of $TiO_2$ and ZnO-NRs for Pb-based perovskite and high PCE of 19.6% has been achieved for $TiO_2$ with Spiro-MeOTAD as the HTL material.

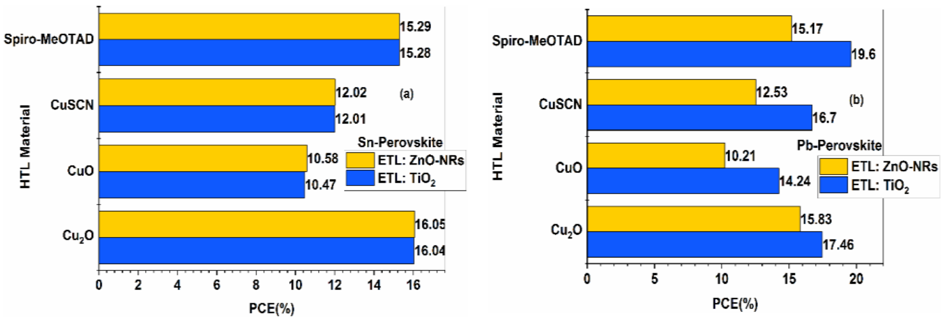


**Fig.4**: PCE of (a) Sn-Perovskite and (b) Pb-Perovskite for different ETL and HTL materials.

Basically, the back contact material is very critical for the final efficiency of perovskite solar cell. The effect of different metals with different workfunctions on the PCE of the solar cell are thoroughly investigated for different HTL materials and for $TiO_2$ and ZnO-NRs as ETL materials. Following metals including Ag, Fe, Cu, Au, Ni and Pt with work functions of 4.74 eV, 4.81 eV, 5 eV, 5.1 eV, 5.5 eV and 5.7eV are considered as the back contact electrodes, respectively. The results are presented in Fig.5. It is observed that the difference in the PCE of the cell with different metals is mainly attributed to the band bending formed at the interface of metal and the HTL. Generally, an ohmic contact is required for efficient charge collection. For low workfunction metals, a barrier exists at the HTM and back contact electrode which degrades hole movement. However, as the metal workfunction increases, an ohmic contact is formed, which ultimately enhances the PCE. Moreover, in Sn-Perovskite, PCE of the cell with CuO and CuSCN HTLs are highly sensitive to the metal workfunction and the device characteristics can be improved with high workfunction metals.

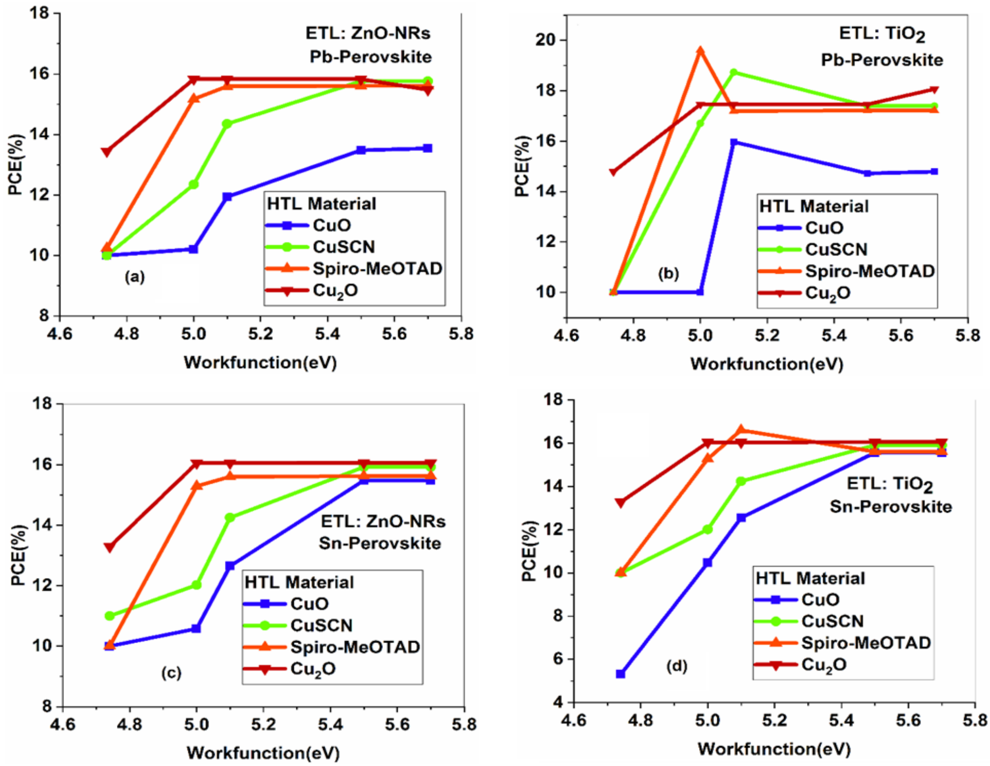


**Fig.5**: PCE of (a), (b) Pb- perovskite and (c), (d) Sn- perovskite as a function of different materials for the back contact electrode.

The effect of perovskite thickness on the efficiency of the proposed photovoltaic device is thoroughly investigated and illustrated in Fig.6 for $TiO_2$ and ZnO-NRs as ETL materials. The HTL and electrodes are chosen based on the materials that provide the highest efficiency, assumed from the results demonstrated in Fig.4. Basically, the absorber thickness is an important and critical parameter for cell performance and optimum thickness should be determined. In planar structure solar cell, a reciprocal relation exists between photo generated carrier collection and photon absorption. In thinner perovskite thicknesses, low efficiency is achieved due to the partial photo absorption and as a consequence, less photo generation. As the absorber material thickness increase, the light absorption increases and results in the efficiency enhancement. However, in the case of thicker perovskite devices, generated carriers that are produced close to the center of perovskite layer will recombine when the perovskite thickness exceeds the diffusion length. The results demonstrate that employing $TiO_2$ as the ETL material makes the efficiency of the solar cell less sensitive to the thickness variation. However, in case of ZnO-NRs in Pb-perovskite, considerable reduction of efficiency occurs due to the increment of recombination rate and lower rate of carrier collection. Moreover, impact of perovskite thickness on the solar cell fundamental electrical characteristics including $V_{oc}$ and $J_{sc}$ are illustrated in Fig.7. The results indicate that $TiO_2$ in Pb-perovskite solar cell is highly insensitive to the thickness variation, in comparison with other structures. The short circuit current increses as the absorber thickness increases which manifests enhancement of the light absorption.

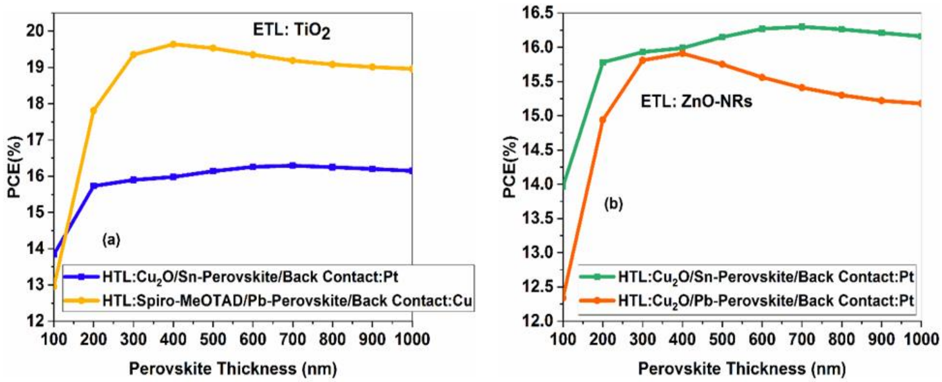


**Fig.6:** Impact of perovskite thickness on the PCE of cell for different ETL materials: (a) $TiO_2$ and (b) ZnO-NRs.

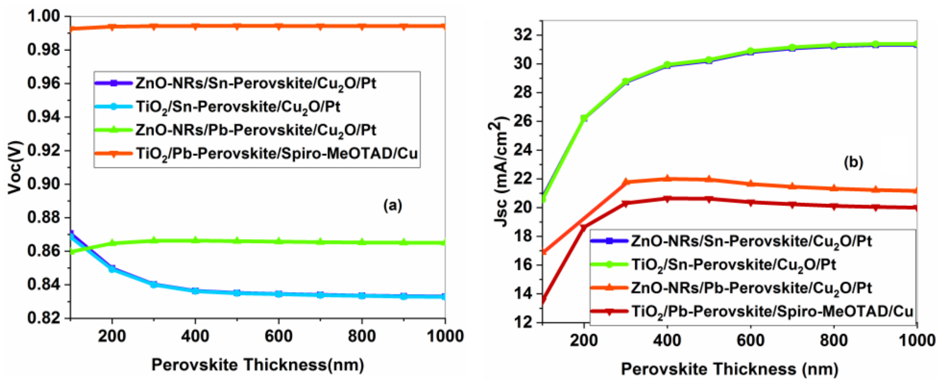


**Fig.7**: Impact of perovskite thickness on the (a) Voc and (b) Jsc for devices with different ETL and HTL materials.

The effect of defect density at the ETL/HTL and the absorber layer on the efficiency of the proposed devices is comprehensively investigated and the results are presented in Fig.8. The charge transfer process at the ETL/ perovskite interface is in strong competition with the recombination process. The defect density degrades the device performance in terms of charge collection and expedites the recombination rate. It is observed that $TiO_2$ employs a proper interface with the absorber layer and the efficiency of the device is insensitive with the defect density. However, as the defect density exceeds $10^{16}$ $cm^{-3}$, degradation occurs in the efficiency of the device with ZnO-NRs as the ETL layer. This effect mainly attributed to the enhancement of recombination rate at the ZnO-NRs/ absorber interface.

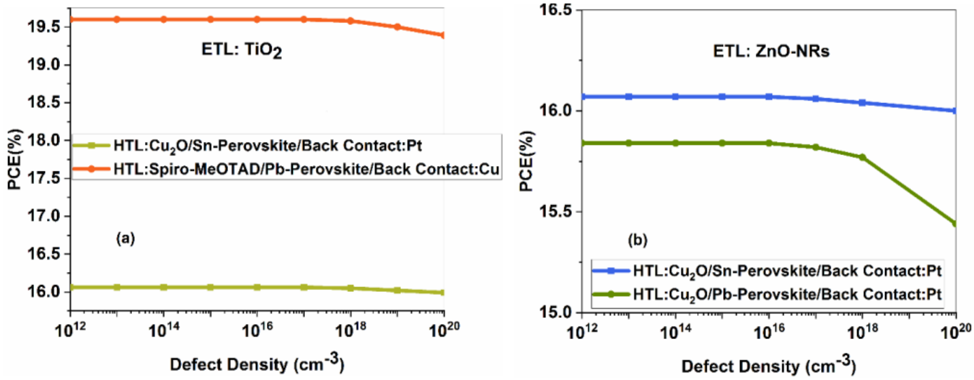


**Fig. 8**: Impact of defect density on the PCE of cell for different ETL materials: (a) $TiO_2$ and (b) ZnO-NRs.

The simulation results based on the optimum structures employing perfect electrical performance with highest efficiency are summarized in Table.3. The energy band diagram of these structures are also illustrated in Fig.9. The results demonstrate that in terms of Sn-Perovskite, the improved performance of ZnO-NRs results from the high carrier mobility. It is observed that in Sn- perovskite, ZnO-NRs has superior performance in comparison with $TiO_2$ while an inexpensive abundant non-toxic inorganic HTL material is employed. However, in terms of Pb-perovskite planar $TiO_2$ has given the best performance with maximum PCE of 19.6%.

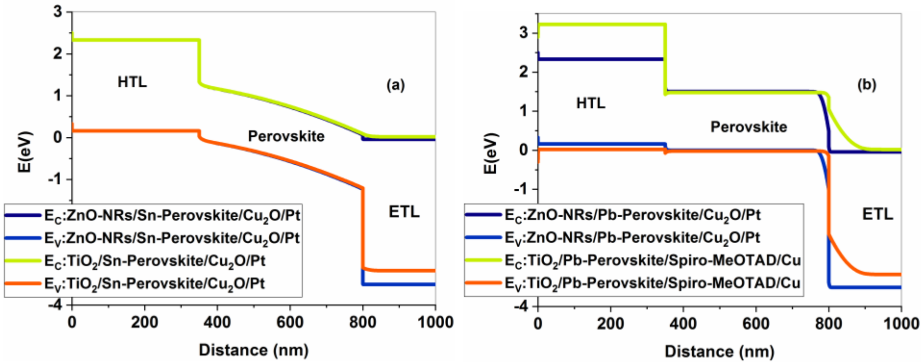


**Fig.9**: Energy band diagram of (a) Sn- perovskite and (b) Pb- perovskite with improved electrical performance.

**Table.3**: Main electrical characteristics of the photovoltaic devices with optimum performance

| Perovskite solar cell | Jsc(mA/cm$^2$) | Voc(V) | FF(%) | PCE(%) |
|---|---|---|---|---|
| ZnO-NRs /Sn- /$Cu_2O$/ Pt | 30.21 | 0.83 | 63.64 | 16.7 |
| $TiO_2$ /Sn- /$Cu_2O$/ Pt | 30.27 | 0.83 | 63.48 | 16.06 |
| ZnO-NRs /Pb- /$Cu_2O$/ Pt | 21.95 | 0.86 | 83.31 | 15.84 |
| $TiO_2$/Pb-/Spiro-MeOTAD / Cu | 20.62 | 0.99 | 95.57 | 19.6 |

## 4. CONCLUSION

In this paper, the electrical characteristics of Sn- and Pb- perovskite solar cell with $TiO_2$ and 1D ZnO-NRs as the ETL of the device are comprehensively investigated. Impact of critical design parameters on the performance of the proposed devices are assessed. The results indicate that in Sn- perovskite, ZnO-NRs with $Cu_2O$ as the HTL material has outstanding performance in comparison with $TiO_2$ yielding PCE of 16.7% and $V_{OC}$=0.83V. However, in terms of Pb-perovskite planar $TiO_2$ has given the best performance with maximum PCE of 19.6% and $V_{OC}$=0.99V. The perovskite thickness is a fundamental design parameter that may affect the efficiency and recombination rate of the device. Accordingly, optimum value should be determined for the absorber layer. The ZnO-NRs performance is highly sensitive to the absorber layer thickness and as a consequence, optimum thickness should be determined. The role of different metals as the back contact electrode on the performance of photovoltaic device is thoroughly investigated. Results show that increasing the workfunction of metals provides a barrier free junction for the majority carriers in the HTL. Effect of interface defect density on Performance of Cell has been comprehensively investigated. It is observed that defect free ZnO-NRs/absorber interface are required for efficient device performance.

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