

Republic of Iraq Ministry of Higher Education and Scientific Research University of Diyala College of Science Department of Physics

Fabrication and Characterization of Perovskite Nanostructured Multijunction Solar Cells

A Thesis

Submitted to the Council of College of Science University of Diyala in Partial Fulfillment of the Requirements for the Degree of Doctor of Philosophy in Physics

By

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هُوَ **الَّذِي جَعَلَ الشَّمْسَ ضِيبَاءً** وَالقَمَرَ نُورًا وَقَدَّرَهُ مَنَازِلَ مْلَمُوا عَدَدَ السِّنِينَ لِتَعْلَمُوا عَدَدَ السِّنِينَ
وَالحِسَابَ مَا خَلَقَ اللهُ ذَلِكَ إِلَّا **بِـالـحَقِّ يُفَصِّلُ الآَيَاتِ لِقَـوْمٍ** يَعْلَمُونَ

 (يونس – اية 5)

DEDICATION

To:
The teacher and the source of human knowledge of *our Prophet Muhammad (peace be upon him)*

To:

The lights of my life My father and my mother

To:

The soul of my beloved brother (Muhammad)

To:

The lights of my eyes Brothers and sisters

To:

My beloved country Iraq The martyrs of Iraq with all the love and appreciation

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 Finally, I thank all my classmates and I wish them success and I would also express my deep sense of gratitude to Shahlaa Munther and Widean Kadim.

Supervisor Certification

We certify that this thesis entitled "Fabrication and **Characterization of Perovskite Nanostructured Multijunction** Solar cells" submitted by "Shaimaa Mufeed Jassim", was prepared under our supervision at the Department of Physics, College of Science, University of Divala in partial fulfillment of requirements needed to award the Degree of Doctor of Philosophy (Ph.D.) in Physics.

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Abstract

Perovskite and Multijunction solar cells are a third generation type of solar cells. Their working principle is based on the conversion of sunlight into electrical energy.

In this study, firstly three types of methyl ammonium halide nanopowders MAX $(X = \text{halide} = I, Br, and Cl)$ have been synthesized by using chemical bath method. After that, eight different types of organicinorganic perovskite thin films have been prepared by using spin coating technique which is $(MAPbI₃, MAPbBr₃, MAPbCl₃, MAPbICI₂$ MAPbIBr₂, FAPbI₃, CsPbI₃ and MASnCl₃). In addition, eight single inverted planar perovskite solar cells (SIPPSCs) and multijunction inverted solar cells (MJISCs (c-Si and SIPPSCs)) have been fabricated successfully by using the prepared different perovskite films as a sensitized absorption layer (active layer).

The structure of SIPPSC consisted of (fluorine tin oxide (FTO) substrate/ Poly (3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS)/ Perovskite/ [6,6]-Phenyl C_{71} butyric acid methyl ester $(PC₇₁BM)$ Aluminum (Al)). In beginning, PEDOT: PSS layer was deposited on FTO coated glass substrate by spin coating technique which acts as hole transport layer (HTL). The different types of perovskite material that act as an absorbing layer were deposited on PEDOT: PSS layer also by spin coating technique. Then, $PC₇₁BM$ layer which represents the electron transport layer (ETL) was deposited on perovskite layer by spin coating technique. Finally, Al as a metal layer was deposited on PC_{71} BM film by thermal evaporation technique, to get cathode electrode.

MJISC structure consisted of (Al/ crystal-silicon (c-Si)/ tin oxide $(SnO₂)/ PC₇₁BM/Perovskite/ PEDOT: PSS/ FTO)$. For the fabrication of MJISCs, initially the $SnO₂$ as a transparent layer was deposited on the c-Si solar cell by using thermal evaporation technique. Then (Al) was deposited as a conductive electrode on the back face of the silicon cell also by using thermal evaporation technology. Finally, the single inverted perovskite solar cell without Al layer was stacked on c-Si solar cell with $SnO₂$ film by using simple mechanical method.

 The structural and morphological properties of prepared powders, different perovskite thin films and $SnO₂$ film have been investigated by X-ray diffraction (XRD) and field emission scanning electron microscope (FESEM) techniques respectively. The optical properties (absorbance and energy gap) of different perovskite films, $SnO₂$ and PEDOT: PSS films have been investigated by (UV-Vis) spectrophotometer. While, photovoltaic characterizations of the fabricated IPPSCs and MJISCs based on different perovskite films have been measured by solar cell simulator.

The XRD results showed that all powders and $SnO₂$ film were polycrystalline with tetragonal structure. All the perovskite films were polycrystalline with different structures which are (tetragonal MAPbI3, cubic (MAPbBr₃, MAPbCl₃, MAPbICl₂ and MAPbIBr₂), rhombohedral $FAPbI₃$, orthorhombic CsPbI₃ and triclinic MASnCl₃).

The FESEM images showed that the powders possess nanonails at $(X= I)$ and nanospheres at $(X= Br$ and Cl) like shapes. While, the perovskite films possess different shapes and $SnO₂$ film has cauliflower like shapes.

The results of UV-Vis spectra showed that all perovskite films have a good absorbance in visible and near IR region. Consequently, it was chosen as an active absorption layer in applications of solar cell.

The optical energy gap for allowed direct electronic transition was calculated using Tauc's model and it was found that the values of energy gap of all perovskite films vary with different variables.

The photo current density-voltage (J-V) curve characteristics of fabricated solar cells have been measured under simulated solar light (100 $mW/cm²$). On the other hand, the parameters (including Voc, J_{SC}, FF and efficiency) of solar cells were calculated depending on J-V curve.

The results showed that the fabricated SIPPSCs and MJISCs based on perovskite (methylammonium lead iodide (MAPbI3) layer as an active layer recorded higher efficiency of (10% and 12.8%) compared to the other fabricated solar cells.

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Chapter One

Introduction & Literature Review

1. Introduction

The increase in industrialization and rapid growth in human population is envisaged to intensify the demand for energy in the near future by a significant proportion [1]. The primary source of energy fossil fuel, which accounted for 66.7% of the world energy consumption has been predicted to exhaust in supply sooner or later. Also, the primary source of energy does produce carbon dioxide $(CO₂)$, which has been identified as the main cause of global warming [2]. Therefore, substantial scientific researchers have been focusing on developing a greener and more sustainable energy to alleviate the environmental impacts [3]. Photovoltaic (PV) is a technology that uses semiconducting materials that exhibit photovoltaic effects and convert sunlight into electricity [4]. Due to abundance, low cost, and environmental friendliness, solar energy has gained significant attention as an outstanding candidate for future power generation [5]. The solar energy is the most promising because of its enormous theoretical and technical potential. The amount of energy from sunlight striking the earth in 1 hour is about $(4.3 \times 10^{20} \text{ J})$, which is higher than all of the energy currently consumed on the planet in one year $(4.1 \times 10^{20} \text{ J})$ [1]. Solar cells can be used as an alternative and renewable energy source that are able to convert light into electric current, both the direct sun light and also artificial light, together with no noise, pollution or moving parts, making them robust, reliable, and long lasting [6]. A new generation of organic-inorganic halide perovskite emerged to prominence in the past few years. Comparing to conventional silicon solar cell, the perovskite-based solar cell has a higher energy return on investment due to low material utilization and ability to the solution process [7]. The efficiency of perovskite solar cells (PSCs) has increased up from 3.8% in 2009 to 23.9% in 2019 [8].

Crystalline silicon (c-Si) solar cells have achieved enormous dominance in the photovoltaic market due to their high efficiencies, excellent material quality, near-optimal band gap energy, and reduction in manufacturing costs [9]. c-Si solar cells are approaching their theoretical Auger limit of 29.4% with a current record at 26.7%. One promising approach to overcome this limit relies on reducing thermalization losses by stacking several absorbers of different band gaps together in a multijunction or tandem device [10]. Tandem solar cells (TSCs) have been proposed to achieve higher conversion efficiency than single-junction solar cells [11]. TSCs are multijunction photovoltaic devices, a unique combination of two sub-cells that arranging in form top solar cell (high band gap) and bottom solar cell (low band gap) that stacked on top of each other [12]. The band gap of the top sub-cell is required to be higher than the band gap of silicon (1.1 eV) in order to absorb the photons of higher energy, thus yielding two complementary absorbing subcells [13]. The incident sunlight first hits the top cell which has a higher band gap and harvests the high-energy photons at a higher voltage, while the low-energy photons are transferred to the bottom cell which has a lower band gap and corresponding lower voltage [14]. In this way, high-energy photons are able to contribute more voltage to the device instead of losing their excess energy by thermalization [15]. Organic-inorganic perovskite materials are excellent photovoltaic materials for tandem applications due to strong absorption coefficients, long electron-hole diffusion lengths, tunable band gap, high charge carrier mobilities, solution-processable, and low cost [16, 17]. As a result, tandem cells with a perovskite top cell and a silicon bottom cell have the potential to reach efficiencies beyond 30%. Consequently, the tandem structure has been proved to be an efficient method to enhance the efficiency of organic or other types of solar cells.

 Organic-inorganic perovskite and c-Si solar cells are fabricated in mainly two configurations: mechanically stacked 4-terminal (4-T) tandem and monolithic 2-terminal (2-T) tandem [18]. TSCs are promising results that have been achieved for 2-T and 4-T perovskite/silicon tandem solar cells reaching energy conversion efficiencies of up to 23.6% and 26.4%, respectively [19].

1.2Literature Review

In 2009, Kojima et al. prepared two types of perovskite methyl ammonium lead triiodide (MAPbI3) or CH3NH3PbI3) and methyl ammonium lead tribromide (MAPbBr₃ or $CH₃NH₃PbBr₃$) by chemical bath method to fabricate solar cells. Perovskite solar cells were fabricated in the structure of order (TiO₂/MAPbI₃ or MAPbBr₃/Spiro-MeOTAD). The solar cell based on MAPbI3 obtained results of short circuit current density (Jsc), open-circuit voltage (V_{OC}) , fill factor $(F.F)$, and power conversion efficiency (PCE) were equal to 11 mA.cm⁻², 0.61 V, 0.57, and 3.81%, respectively. While, the solar cell based on MAPbBr₃ recorded $J_{SC} = 5.75$ mA.cm⁻², Voc=0.96 V, F.F=0.59 and PCE=3.13% respectively [20].

In 2011, Im et al. fabricated quantum dot sensitized solar cell based on MAPbI₃ material as an active layer with the following structure (TiO_2) $MAPbI₃/Pt$). Spin coating method was used to deposit MAPbI₃ thin film onto the surface of titanium oxide. From the results, it was found that efficiency of the fabricated solar cell recorded 6.54% at AM1.5 (1 sun) illumination [21].

In 2013, Kim et al. synthesized nanoparticles perovskite from the reaction of methylamonium iodide (MAI or CH₃NH₃I) with lead iodide (PbI2) solutions. Also, perovskite solar cells were fabricated in the structure order of (TiO2/MAPbI3/Spiro-MeOTAD) by spin coating technique. From

the results, it was found that Jsc was 17 mA/cm2 , Voc was 0.888 V, F.F was 0.62, and PCE was 9.7% [22].

In 2013, Bi et al. synthesized CH3NH3PbI3 by two-step spin coating technique for methylammonium lead iodide perovskite films for the preparation of highly reproducible solar cells, with the following structure $(FTO/C-TiO₂/m-TiO₂/CH₃NH₃PbI₃/Spiro-OMeTAD/Ag)$. It was found that Jsc was 18.3 mA/cm2 , Voc was 0.89 V, F.F was 0.58 and efficiency reached up to 9.5% which was achieved under AM 1.5 illumination of 1000 W m-2 [23].

In 2014, Eperon et al. fabricated planar hetero-junction perovskite solar cells (PHJPSCs) with the following structure $(FTO/C-TiO₂/$ Perovskite (CH3NH3PbI3-xClx)/Spiro-OMeTAD/Au). The perovskite layer was deposited on the surface of the $C-TiO₂$ film by using spin coated method. From current density-voltage (J-V) curve, it was noted that J_{sc} =15.3 mA/cm², V_{OC}=0.8 V, F.F=0.55, and PCE= 6.7% [24].

In 2014, Eperon et al. synthesized formamidinium lead trihalide $(FAPbX_3$ or $NH_2CH=NH_2PbX_3$, X=halide= I and Br) perovskite films and showed the effect of replacing the methyl ammonium cation in this perovskite ($FAPbI_zBr_{3-z}$), with the slightly larger formamidinium cation, with z increasing from $(0 \text{ to } 1)$. FAPbI_zBr_{3-z} films have a band gap tunable between 1.48 and 2.23 eV, respectively. They took the 1.48 eV band gap perovskite as most suited for single junction solar cells, and demonstrated long-range electron and hole diffusion lengths in this material, making it suitable for planar hetero-junction solar cells (PHJSCs). They fabricated such devices, and due to the reduced band-gap they achieved high shortcircuit currents of >23 mA/cm2 , resulting in power conversion efficiencies of up to 14.2%, the highest efficiency yet for solution processed PHPSCs. Formamidinum lead triiodide (FAPbI3) was hence promising as another candidate for this class of solar cells [25].

In 2015, Chen et al. synthesized perovskite/polymer in tandem solar cell structure. The wide band gap perovskite absorber MAPbI₃ was processed via a one step deposition. It was observed that PCE of 9.1% was obtained for a single-junction, planar structured MAPbI₃ solar cell. While the hybrid tandem solar cell based on perovskite/polymer subcells could achieve an optimal efficiency of 10.2% which is greater than both of the perovskite and polymer single-junction cells [26].

In 2015, Yuan et al. fabricated inverted planar perovskite solar cells (IPPSCs) by using FAPbI3 as a layer for light harvesting. X-ray diffraction (XRD), field emission scanning electron microscope (FESEM) and J-V curve properties of FAPbI₃ film were studied. IPPSCs were fabricated with the following structure (ITO/PEDOT:PSS/ FAPbI3/PCBM/BCP/Ag) by spin coating technique. From the curves, it was found that PCE of 13.56% was obtained with high short circuit current density of 21.48 mA/ cm² [27].

In 2015, Eperon et al. prepared inorganic perovskite material cesium lead triiodide (CsPbI3). They also fabricated planar inverted perovskite solar cells with structure (ITO/PEDOT:PSS/Perovskite(CsPbI₃)/PCBM/Al). The XRD and Ultraviolet-Visible (UV-Vis) characterizations were carried out for synthesized CsPbI₃ thin films. From J-V characteristics, it was found that the planar inverted device generates PCE of 1.7% [28].

In 2015, Loper et al. fabricated 4-T monolithic tandem solar cell consisting of a methyl ammonium lead iodide as top cell and a c-Si as bottom cell. The tandem soar cell was fabricated in the structure order of (Ag/c-Si/c-TiO2/m-TiO2/Perovskite (MAPbI3)/Spiro-MeOTAD/Ag). The MAPbI₃ top cell displayed broad band transparency owing to its design free of metallic components and yields a transmittance of >55% in the near infrared spectral region. This allowed generating a short-circuit current density of 13.7 mA/cm² in the bottom cell. From the results, it was shown that the 4-T tandem solar cell yielded an efficiency of 13.4 % (perovskite cell: 6.2 % and c-Si cell: 7.2 %) [29].

In 2015, Werner et al. fabricated monolithic perovskite/silicon heterojunction tandem solar cells yielding efficiencies of up to 21.2 and 19.2% for cell areas of 0.17 and 1.22 cm², respectively [5].

In 2016, Ren et al. fabricated 4-T perovskite/c-Si tandem solar cells. The perovskite solar cell as top cell was consisting of (FTO/TiO2/Perovskite(MAPbI3)/Spiro-MeOTAD/MoO3/Au/MoO3). While, c-Si solar cell as bottom cell was consisting of (Al/ p-n Si/ SiNx/ Ag). The tandem cells were produced by combining the perovskite with c-Si solar cells by mechanical method. From the results, it was found that PCE of 23.6% for a multijunction device is achieved compared to 18.1% and 19.1% of individual perovskite and c-Si solar cells respectively [30].

In 2016, Albrecht et al. fabricated a monolithic tandem device formed by a silicon hetero-junction as bottom cell and a perovskite (CH3NH3PbI3) as top cell. The monolithic integration was achieved via low temperature processing of the perovskite subcell where an energetically aligned electron selective contact was fabricated by atomic layer deposition of tin oxide. The hole selective transparent top contact was formed by a stack of the organic hole transport material spiro-OMeTAD, molybdenum oxide and sputtered indium tin oxide. It was recorded that the PCE of silicon, perovskite, and tandem are 15.7%, 10.4%, and 19.1% respectively [31].

In 2017, Fan et al. prepared 4-terminal tandem (Si/Ps) solar cell. The fabricated perovskite solar cell acting as the top cell was composed of $(FTO/c-TiO₂/Perovskite (CH₃NH₃PbI_{3-x}Cl_x)/Spino-MeOTAD/Au).$ Then the tandem cell was synthesized by combining top cell (perovskite cell) with bottom cell (crystal silicon) by simple mechanical method. From the results, it was found that the tandem solar cell yielded an efficiency of

14.8%, with contributions of the top 8.98% and the bottom cell 5.82% respectively [32].

 In 2017, Suhail et al. prepared organolead halide perovskite solar cells based on $(CH_3NH_3PbI_3$, $CH_3NH_3PbBr_3$, $CH_3NH_3BrPbI_2$, $CH_3NH_3IPbCl_2$ $CH_3NH_3IPbBr_2$ and $CH_3NH_3BrPbCl_2$) as absorption layer. These cells recorded efficiency of (2.15 %, 0.74 %, 0.56 %, 0.27 %, 0.12 $\%$, and 0.07 $\%$) respectively [3].

In 2018, Singh et al. synthesized perovskite material methyl ammonium tin trichloride (MASnCl3) and studied its application in fabrication of PSCs. MASnCl₃ was prepared by direct deposition of equimolar concentration of methylamonium chloride (MACl) and tin chloride $(SnCl₂)$ in Anhydrous N,N dimethyl formamide (DMF) solution. The XRD, UV-Vis, and FESEM characterization of the prepared thin films were performed. The perovskite solar cell was fabricated in the structure order of (FTO/ b-TiO₂/ n-TiO₂/ Perovskite (CH₃NH₃SnCl₃)/ Electrolyte/ Pt/ FTO) by spin coating method. It was noted the fabricated perovskite solar cells showed PCE of 0.55% at 100 mW/cm2 [33].

In 2018, Kanda et al. prepared perovskite single-junction solar cells and 2-T tandem solar cells. The perovskite solar cells were fabricated based on MAPbI3 material as absorbent layer with the following structure $(FTO/bI-TiO₂/m-TiO₂/MAPbI₃/Spiro-MeOTAD/ MO3)$. While, tandem cell was fabricated by combining silicon solar cell with perovskite solar cell in mechanically stacked form by direct contact of transparent conductive oxide (TCO) layers of each sub-cell. The photovoltaic parameters of tandem device solar cell were $J_{\text{SC}}=16.2 \text{ mA/cm}^2$, $V_{\text{OC}}=1.42$ V, FF= 0.602, PCE=13.9% [34].

In 2018, Pisoni et al. prepared tandem solar cell based on perovskite as top cell and $Cu(In,Ga)Se₂ (CIGS)$ as bottom cell. The perovskite solar cell based on MAPbI3 material as an active layer was fabricated in the structure following $(IZO/C_{60}/perovskite (MAPbI₃)/Spino-OMeTAD/Au)$. Then, the tandem cell was fabricated by combining the perovskite with CIGS solar cell by mechanical stacked method. It was found that perovskite cell, CIGS cell and perovskite/CIGS tandem devices were demonstrated with an efficiency of 14%, 5.6%, 19.6%, respectively [35].

 In 2018, Zhuk et al. fabricated monolithic perovskite/silicon heterojunction 2-T tandem solar cells. The perovskite solar cell with structure (FTO/SnO2/MAPbBr3/Spiro-OMeTAD/Au) was synthesized. From the results, it was found that perovskite solar cell and perovskite/silicon multijunction solar cell were obvious with a PCE of 14.39% and 18.42%, respectively [36].

In 2018, Bush et al. fabricated 2-T monolithic tandem solar cells. They chose the silicon as bottom cell and perovskite as top cell. In addition, the cesium formamidinium lead halide was used as an active layer in perovskite solar cells. The single-junction perovskite solar cells were fabricated in the structure order of $(ITO/NiO/Cs_{0.17}FA_{0.83}Pb$ (Br $_{0.17}$) $I_{0.83}$)₃/PC₆₀BM/SnO₂/ITO/LiF/Ag). The results showed that the total power conversion efficiency of tandem cell reached 23.6% [17].

In 2018, Qiu et al., synthesized four mixed perovskite thin films $(FA_{0.48}MA_{0.37}Cs_{0.15}PbI_{2.23}Br_{0.77}$, $FA_{0.57}MA_{0.43}PbI_{2.4}Br_{0.96}$, $FA_{0.5}MA_{0.38}$ $Cs_{0.12}PbI_{2.04}Br_{0.96}$ and $FA_{0.51}MA_{0.38}Cs_{0.11}PbI_{1.85}Br_{1.15}$ with different band gaps 1.65 eV, 1.69 eV, 1.69 eV and 1.72 eV respectively by spin coating technique. Also, monolithic tandem solar cells were fabricated by using the prepared perovskite films in the following structure (Si cell/ITO/SnO2/Perovskite/Spiro-OMeTAD/MoOx/ITO). From the results, it was found that the perovskite film with a 1.69 eV band gap showed the best performance when incorporated in the top subcell and consequently the resulting device achieved an efficiency of 22.2% [37].

In 2018, Baron et al. prepared two types of perovskite thin films $(MAPbBr₃$ and $MAPbBr₂Cl$) by one step coating method. The structural, morphological and optical properties of perovskite films were studied. They also fabricated single perovskite solar cells based on prepared perovskite films with the following structure $(FTO/TiO₂/Ncs/$ $MAPbX₃/Ag$). From results, it was found that PCE achieved was 1.2 % and 0.9 % for MAPbBr₃ and MAPbBr₂Cl perovskite solar cells respectively [38].

In 2020, Lamanna et al. fabricated two-terminal perovskite/silicon tandem solar cell by mechanical stacking of the sub-cells. Mixed-halide perovskite $(Cs_{0.06}FA_{0.78}MA_{0.16})$ Pb $(Br_{0.17}I_{0.83})$ was used as an active layer for perovskite solar cell. From the results, it was observed that tandem device displayed 26.3% efficiency over an active area of 1.43 cm² [39].

1.3 Objectives of the Study

- **1.** Preparation of single inverted planar perovskite solar cells (SIPPSCs) based different types of perovskite films by spin coating technique.
- **2.** Fabrication of multijunction inverted solar cells (MJISCs) based perovskite solar cell as a top cell and c-Si solar cell as a bottom cell to improve the efficiency.