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Development of Halide Perovskites for Solar Cell Applications

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ABSTRACT

Halide perovskites have shown great potential for high efficiency solar cells, owing to their excellent optical and electrical properties. In this work, the preliminary development of methylammonium lead iodide ($MAPbI_3$) perovskite thin film and planar negative-intrinsic-positive (*n-i-p*) thin film-based perovskite solar cell were demonstrated. The $MAPbI_3$ thin film was successfully prepared using the anti-solvent assisted "one-step" solution deposition method. Ultraviolet-Visible (UV-Vis) absorbance analysis on the $MAPbI_3$ thin film indicated the presence of broad absorption over the visible region from 450 to 800 nm. The Tauc plot bandgap energy was determined to be 1.535 eV. Atomic force microscope (AFM) imaging evidenced a uniform and smooth perovskite film with an average grain size of 90 nm and a root-mean-square roughness of 3.3746 nm. The current-voltage (*I-V*) curve of the perovskite solar cell demonstrated a linear relationship and subpar photovoltaic performance, revealing the need for further investigations and improvements in the design and fabrication process of the solar cell.

1. Introduction

Perovskite solar cells (PSCs) are widely utilised and known as the emerging class of 3rd generation solar cell technology [1]. In recent years, researchers have gained tremendous interest in PSCs due to their high-power conversion efficiency (PCE), reaching as high as 25.7% [2,3] comparable to commercialised crystalline silicon-based solar cells. The PSC device structures are mainly classified into two major categories, which are planar and mesoporous structures that correspond to the structures without and with mesoscopic metal oxide scaffolds, respectively. Furthermore, these two structures are further divided into negative-intrinsic-positive (*n-i-p*) "regular" and positive-intrinsic-negative (*p-i-n*) "inverted" types, depending on which direction the light radiates on the solar cell. Thus, in the *n-i-p* structure, the light is incident through the electron transport layer (ETL) side. In contrast, in the *p-i-n* structure, the light is incident through the hole transport layer (HTL) side.

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In PSCs, they typically utilise perovskite material, which is an organic-inorganic metal halide compound, while acting as the active layer of the device to absorb solar radiation. Subsequently, electron-hole pairs are generated following photoexcitation of the perovskite layer. The created free charge carriers, electrons, and holes will then be transported to the respective electrodes via ETL and HTL, respectively. Hence, a photocurrent in the outer circuit of the device is produced when the two electrodes are connected.

The perovskite materials can be represented in a general formula of ABX_3, where A is an organic cation (methylammonium, MA ($CH_3NH_3^+$) or formamidinium, FA ($HC(NH_2)_2^+$)), B is a metal cation (Pb^{2+}, Sn^{2+}, Cs^{2+}, Cd^{2+} or Bi^{3+}), and X is a halide anion (I^-, Br^- or Cl^-), yielding various types of perovskite materials, such as $MAPbI_3$, $FASnI_3$, $MASnBr_3$, and $FA_3Bi_2I_9$. The lead (Pb) -free perovskite materials (Sn and Bi -based halide perovskite) and antimony-based perovskite-like materials ($MASb_2I_9$ and $Cs_3Sb_2I_9$) are potential candidates for low-toxicity PSC applications [4-7]. The perovskite material has a characteristic cubic lattice-nested octahedral layered crystal structure, which results in remarkable impressive optical, thermal, and electromagnetic properties [8-10].

The perovskite absorber layer can be fabricated via several different approaches. Typically, the "one-step" solution deposition technique [11,12] consists of a mixture of BX_2 and AX from common organic solvents (GBL, DMF, DMSO) and spin-coating the precursor over the ETL-coated transparent conductive oxide (TCO) glass substrate. Alternatively, in the "two-step (sequential)" solution deposition technique [13,14], the BX_2 precursor is initially spin-coated. Then, the substrate was sequentially exposed to a solution of AX either by dip-coating or spin-coating to induce the solid-liquid reactions. Aside from the solution synthesis methods, the one-step and two-step deposition routes can also be produced using vapour deposition techniques (thermal evaporation, pulse laser deposition, vapour-assisted solution process) [15-18].

In addition to the variousness of the perovskite materials, numerous materials have been utilised for the other constituent layers of the PSC, such as TiO_2, ZnO, and Al_2O_3 for ETL [19], and both organic materials (spiro-OMeTAD, PEDOT: PSS, PTAA) and inorganic materials (CuO, CuSCN, CuI, NiO) for HTL [20-23]. For back electrode, metals (Au, Ag, Al) or carbon [24] were commonly utilised. Noticeably, a wide variety of PSC architectures, material sets, and fabrication techniques could be employed in conjunction with perovskite. Nevertheless, to enable the commercial production of the most cost-effective PSC devices, a simple device architecture and easy fabrication procedures are required. In this work, the preliminary development of methylammonium lead iodide ($MAPbI_3$) perovskite thin film and planar *n-i-p* thin film-based PSC with the device structure composed of FTO/TiO_2/$MAPbI_3$ perovskite/CuI/C (as illustrated in Figure 1) are demonstrated.

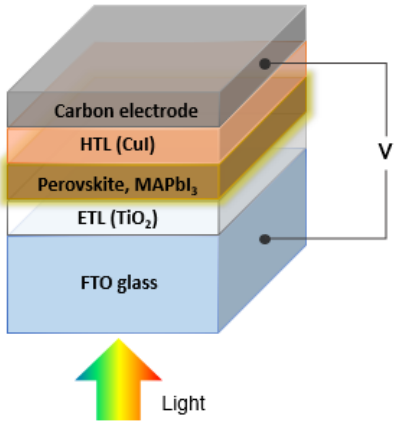

Fig. 1. Device structure of perovskite solar cell in this work

The remainder of this paper is structured as follows: Section 2.1 details the methodology employed to synthesise the $MAPBI_3$ perovskite thin film. Section 2.2 describes the fabrication procedures used to develop the PSC device, and Section 2.3 shows the types of sample characterisations that were conducted. Section 3 presents the experimental results and discussions, specifically analysing the optical absorption and bandgap of the film in Section 3.1, the surface morphology and average roughness of the film in Section 3.2, and the current-voltage (*I-V*) characteristics and photovoltaic (PV) performance of the device in Section 3.3. Section 4 closes with a conclusion.

2. Methodology

2.1 Synthesis of Perovskite Thin Film

The $MAPbI_3$ perovskite thin film was synthesised on top of a pre-cleaned FTO glass substrate via the anti-solvent assisted "one-step" solution deposition technique. The $MAPbI_3$ precursor solution was prepared by mixing an equimolar ratio of methylammonium iodide (MAI) and lead iodide (PbI_2) in dimethylformamide (DMF) solvent. The mixture was heated at 70°C until the materials were fully dissolved. Subsequently, the solution was filtered using a 0.45 μm PTFE filter. Then, the filtered precursor solution was spin-coated onto the FTO substrate. The chlorobenzene (CB) anti-solvent treatment was performed by drenching the CB onto the perovskite film and spinning the film. Finally, the perovskite film was annealed at 100°C for 20 mins.

2.2 Fabrication of Perovskite Solar Cell

The complete procedures to fabricate the FTO/TiO_2/$MAPbI_3$/CuI/C perovskite solar cell are illustrated in Figure 2.

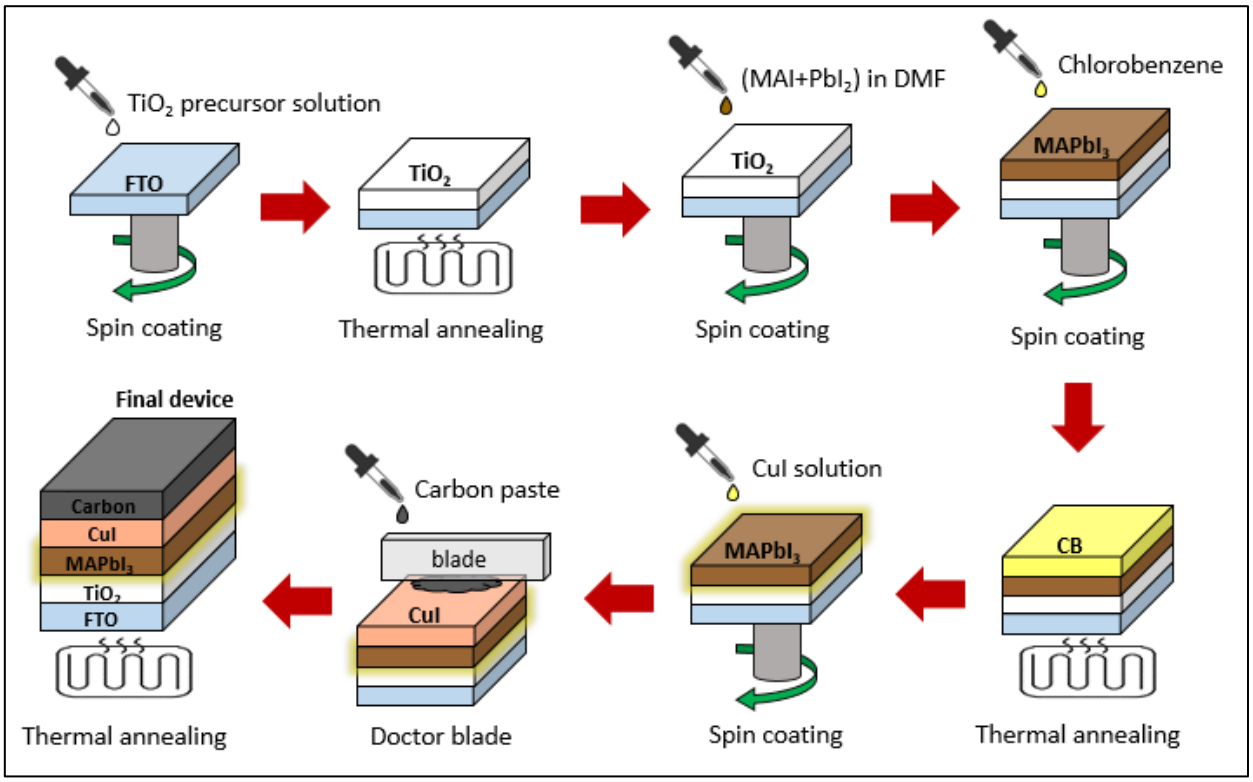

Fig. 2. Fabrication procedures of perovskite solar cell

Firstly, the ETL titanium dioxide (TiO_2) precursor solution prepared via the sol-gel method was spin-coated onto a pre-cleaned FTO substrate, then the TiO_2 film was sintered at 450°C for 30 mins. Subsequently, perovskite deposition and CB treatment were carried out onto the TiO_2-coated FTO, following similar processes described in Section 2.1. Afterwards, the HTL copper iodide (CuI) solution, prepared by dissolving the CuI powder in a mixture consisting of CB, acetonitrile, and 4-tert-butylpyridine (tBP), was spin-coated onto the CB-treated perovskite film. Finally, the carbon back electrode layer was deposited on top of the CuI film via doctor blade by supplying the carbon paste that contained a mixture of graphite, carbon black, ethyl cellulose, and terpineol. The final device was obtained upon annealing the carbon layer at 90°C for 30 mins. Note that the whole fabrication process was conducted under ambient conditions, and all spin-coating processes were operated at 3000 rpm for 30 secs.

2.3 Sample Characterisations

The deposited perovskite thin film was characterised in thickness, optical absorbance, and surface morphology using a surface profilometer, ultraviolet-visible (UV-Vis) spectroscope, and atomic force microscope (AFM), respectively. The photovoltaic (PV) performance of the perovskite solar cell was measured using the solar simulator to obtain both the current-voltage (*I*-*V*) characteristics and the PV parameters of the device, which include the open-circuit voltage (V_{oc}), short-circuit current (I_{sc}), fill factor (FF), and efficiency (η).

3. Results and Discussions

3.1 Optical Properties of the Perovskite Thin Film

The $MAPbI_3$ perovskite thin film that has been synthesised was 100 nm in thickness, measured by a profilometer. Figure 3 reveals the optical absorption spectrum of the $MAPbI_3$ film and the corresponding Tauc plot, $(\alpha h\nu)^{1/n}$ as a function of the photon energy, where α is the absorption coefficient in cm^{-1} (2.303*Absorbance/Film thickness), $h\nu$ is the photon energy in eV, and n is ½ as for a direct bandgap semiconductor. It can be observed from the absorbance spectrum that the film has a broad absorption over the visible range from 450 to 800 nm (the absorption edge), which indicates the typical characteristics of the $MAPbI_3$ perovskite film [25-27]. Based on the point where the linear extrapolation of the Tauc plot intercepts the x-axis, the optical bandgap, E_g, of the film was calculated to be 1.535 eV. Therefore, the bandgap measured was comparable to the reported literature values, ranging between 1.5 to 1.6 eV [26,28,29].

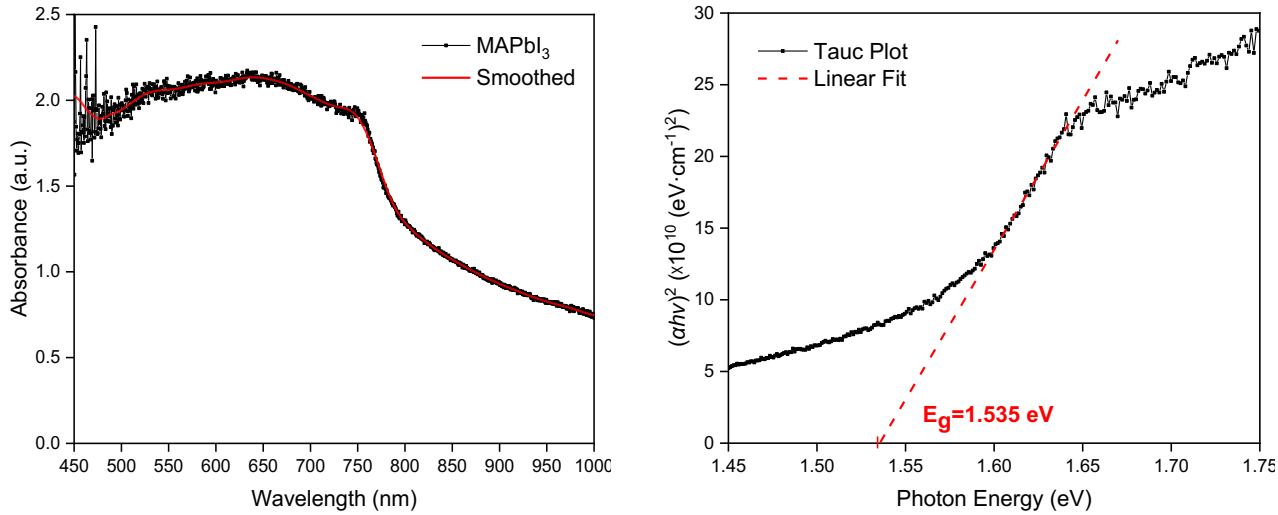

Fig. 3. Absorption spectrum and Tauc plot of $MAPbI_3$ perovskite film

3.2 Surface Morphology of Perovskite Film

Figure 4 displays the 2D and 3D topographical images of the $MAPbI_3$ perovskite film. It can be observed that the film surface exhibits the common $MAPbI_3$ morphology [26,30]. The film is well covered with considerably uniform spherical $MAPbI_3$ grains with an average grain size of 90 nm, and the root-mean-square (RMS) roughness of the film was determined to be 3.3746 nm. The results indicated that the chlorobenzene anti-solvent effect could have taken place during the synthesis process to induce a rapid crystallisation of the perovskite, producing a smooth and homogeneous film [31,32].

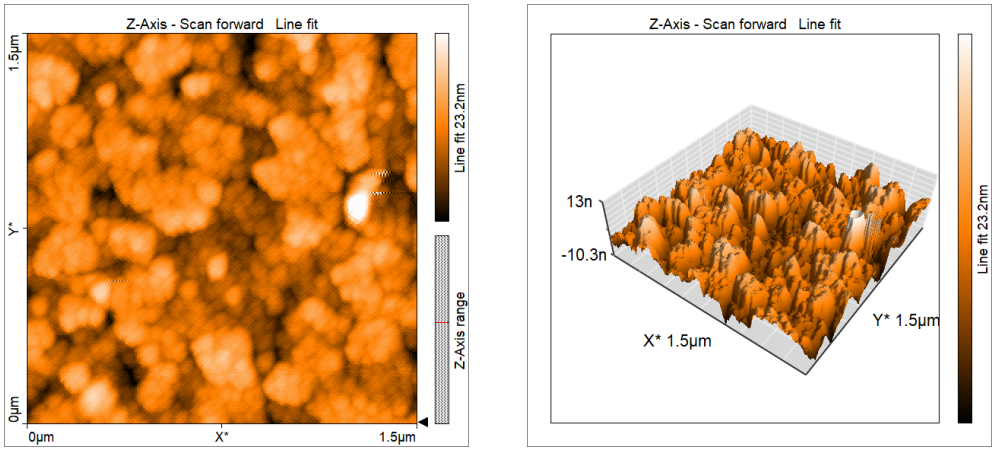

Fig. 4. 2D and 3D 1.5µm×1.5µm AFM images of $MAPbI_3$ perovskite film

3.3 Photovoltaic Performance of Perovskite Solar Cell

The *I-V* characteristic curve of the fabricated perovskite solar cell ($FTO/TiO_2/MAPbI_3/CuI/C$) is demonstrated in Figure 5. Meanwhile, the photovoltaic (PV) parameters are listed in Table 1. Instead of getting a standard concave solar cell *I-V* curve (nearly no change of current at a small voltage and a sharp decrease in current at a certain point of voltage) [33], the curve obtained indicated a straight-line (linear) *I-V* characteristic with exceptionally low PV performance of V_{oc} = 0.27 V, I_{sc} = 1.82 mA, FF = 0.25, and η = 0.12%. The results implied that the device has very high series resistance (R_s) due to

inferior charge transport behaviours in the absorber, charge-collecting layers and/or contact electrodes, and very low shunt resistance (R_{sh}) due to the presence of leakage paths (pinholes) in the device [34, 35]. The high R_s reduced both the I_{sc} and FF; on the other hand, the low R_{sh} reduced the V_{oc} and FF [36]. Consequently, low efficiency was obtained.

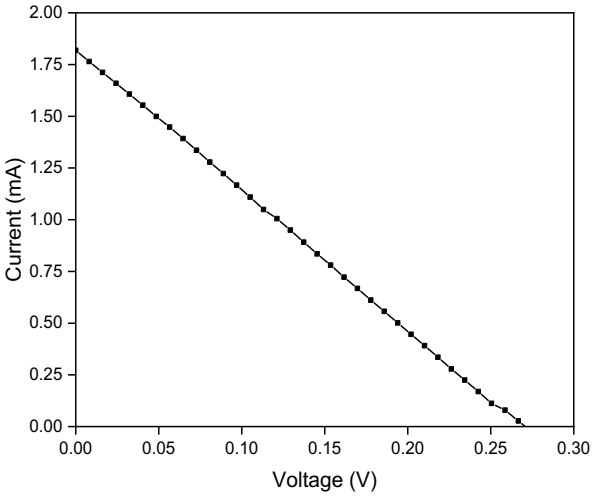

Fig. 5. *I-V* characteristic curve of perovskite solar cell

Table 1
The photovoltaic performance of perovskite solar cell

Device structure	Open-circuit voltage, V_{oc} (V)	Short-circuit current, I_{sc} (mA)	Fill factor, FF	Efficiency, η (%)
FTO/TiO_2/$MAPbI_3$/CuI/C	0.27	1.82	0.25	0.12

The results clearly showed the necessity to re-examine the existing design and the fabrication processes of the perovskite solar cells. More importantly, each layer prior to fabricating the stack layers of the device would need to be optimised to achieve improved PV performance. Besides, it would be better to conduct the fabrication process in a controlled environment, such as an inert gas-filled glove box, since the perovskite materials are extremely sensitive to moisture and oxygen [37,38]. Furthermore, the final device should be encapsulated to protect it against the intrusion of external agents for longer operational stability [39].

4. Conclusion

In conclusion, the preliminary development of the $MAPbI_3$ perovskite thin film and the planar *n-i-p* FTO/TiO_2/$MAPbI_3$ perovskite/CuI/C perovskite solar cell was successfully demonstrated. The as-deposited $MAPbI_3$ perovskite film exhibited optical properties and surface morphology that are comparable to those reported in the literature. On the contrary, the photovoltaic performance of the fabricated perovskite solar cell was inferior. It also demonstrated the characteristics of a shunted solar cell device. The results obtained reveal the need for many further investigations and improvements in the design and the fabrication processes for realizing the planar *n-i-p* thin film-based perovskite solar cell.

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