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Fabricating Lead-Free $CH_3NH_3SnI_3$ Perovskite Absorber Layer from Powders to High-Quality Thin Film

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Abstract

Lead-based perovskites have dominated the field of optoelectronics due to their exceptional efficiency, while their inherent toxicity poses environmental challenges. In this study, we report an eco-friendly synthesis route for lead-free methylammonium tin iodide ($CH_3NH_3SnI_3$) perovskite thin films, using commercially available powders as precursors. The perovskite films were fabricated via a spin-coating process under a nitrogen atmosphere, followed by low-temperature annealing. Structural characterisation using X-ray diffraction (XRD) confirmed the formation of a cubic perovskite phase. Scanning electron microscopy (SEM) revealed uniform, good film formation. Optical characterisation indicated a direct bandgap of 1.31 eV and robust visible light absorption. This facile and environmentally conscious method offers a sustainable pathway for developing high-quality, lead-free perovskites for next-generation photovoltaic and optoelectronic applications.

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1. INTRODUCTION

In recent years, organic–inorganic hybrid perovskite materials have earned considerable attention towards efficiency due to their remarkable optoelectronic properties and potential applications in solar cells, light-emitting diodes, photodetectors, and other electronic devices [1]. These materials offer several advantages including strong optical absorption, long carrier diffusion lengths, tunable bandgaps, and the possibility of low-cost solution-based fabrication methods. One of them, a high-performance perovskite solar cell material is reported which is a lead-based compound such as methylammonium tin iodide perovskite ($CH_3NH_3PbI_3$) [2-4]. Although these materials demonstrate excellent photovoltaic performance and according to National Renewable Energy Laboratory (NREL) it is observed that the efficiencies of perovskite materials have increased from 3.8% in 2009 to 26.95% till now [5]. But the presence of toxic lead raises serious environmental and health concerns. Consequently, the development of lead-free perovskite alternatives has become a milestone research direction in the field of sustainable photovoltaic technology. While tin-based perovskites show less efficiency (13.24 % till now) than lead based perovskites, theoretically it has been reported about 31 % by using SCAPS- 1D simulations [6-8]. Particularly methylammonium tin iodide ($CH_3NH_3SnI_3$) perovskites are considered promising candidates to replace lead-based materials. This material possesses a suitable bandgap of approximately 1.31 eV, which is close to the optimal value for single-junction solar cells. In addition, tin-based perovskites exhibit high absorption coefficients and favorable charge transport properties. However, to achieve high-quality tin-based perovskite thin films remains challenging due to issues such as oxidation of Sn^{2+} ions and poor film morphology. Several fabrication methods like spin coating [9], thermal evaporation [10], chemical vapour deposition [11] and physical vapour deposition method [12] have been reported. Optimizing the synthesis and deposition methods is important for improving the structural and optical properties of these materials. One of the most important methods is solution-based technique. Several studies have been reported the solution-based $MASnI_3$ crystals exhibits high uniformity and improves structural properties [13-14]. In this report, $CH_3NH_3SnI_3$ thin films were fabricated using a straightforward solution processing method based on spin coating. The films were prepared from commercially available precursor powders and subjected to controlled thermal annealing to enhance crystallinity. The structural, optical, and morphological properties of the prepared films were systematically investigated using XRD, UV–Visible spectroscopy, and scanning electron microscopy.

2. Experimental details

Chemicals

Tin iodide (SnI_2), methylammonium iodide (CH_3NH_3I), dimethylformamide (DMF) and dimethyl sulfoxide (DMSO). All precursor powders and chemicals were commercially obtained and stored in a nitrogen-filled glovebox to prevent moisture and oxygen contamination.

Synthesis of $CH_3NH_3SnI_3$ film

The $CH_3NH_3SnI_3$ precursor solution was prepared by adding 0.159g MAI with 0.3725g SnI_2, stoichiometric amounts of methylammonium iodide (MAI) and tin iodide (SnI_2) in 1ml solvent of 0.80 ml dimethylformamide (DMF) and 0.20 ml of DMSO (4:1). The solution was stirred continuously at room temperature for 2 hours. Then prepared precursor solution was filtered and deposited onto the Indium tin oxide (ITO) substrates using a spin-coating technique, before the film deposition the glass substrate was cleaned sequentially using detergent solution, deionized water, acetone, and isopropyl alcohol in an ultrasonic bath, followed by drying with nitrogen gas. The spin coating process was performed in two stages, initially at a low speed 1000 rpm for 10 seconds to spread the solution uniformly and then at 4000 rpm for next 30 seconds to achieve a uniform thin film. After deposition, the films were annealed at moderate temperature (approximately 100 °C) for 10 minutes under nitrogen atmosphere to promote crystallization and remove residual solvent. The annealing process facilitated the formation of high-quality $CH_3NH_3SnI_3$ perovskite thin films as shown in Fig.1.

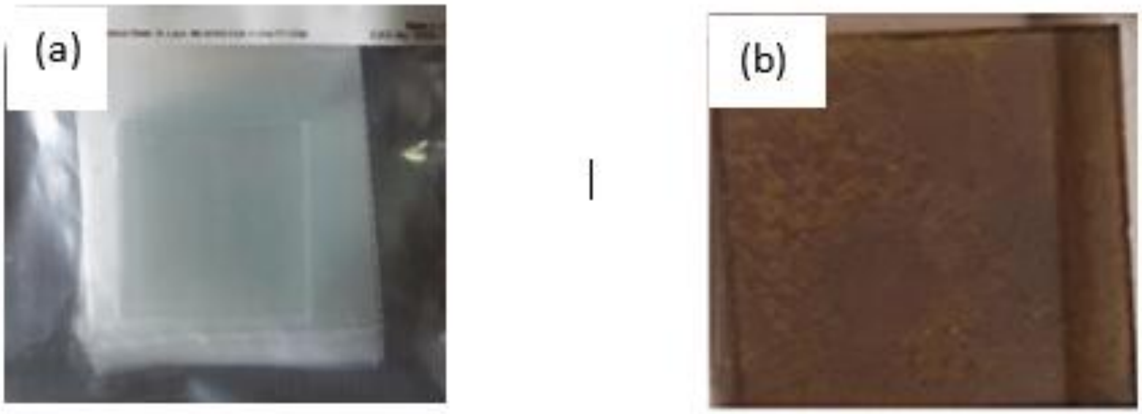

Fig.1. (a) ITO glass substrate and (b) after annealing $MASnI_3$ film

3. RESULTS AND DISCUSSIONS

3.1 XRD analysis

The synthesized perovskite thin film was analyzed by X-ray diffraction (XRD) to observe the structural properties of the $CH_3NH_3SnI_3$ film. The obtained results are shown in Fig.2. XRD pattern showed distinct diffraction peaks corresponding to the cubic perovskite structure of methylammonium tin iodide perovskite film [15]. Prominent diffraction peaks were observed at 2θ = 14.4°, 24.4°, 28.1°, 41° corresponds to the cubic structure of $CH_3NH_3SnI_3$. The clear peaks at 12°, 20°, 26° represents the presence of SnI_2. The absence of additional impurity peaks indicates the successful formation of phase-pure $CH_3NH_3SnI_3$ thin films. The sharp and intense diffraction peaks suggest good crystallinity of the films. The Scherrer equation is used to calculate the average crystallite size [16].

$$D = \frac{K\lambda}{\beta \cos\, \theta} \qquad (1)$$

where, D = crystallite size (nm), K = shape factor (≈ 0.9), λ = wavelength of X-ray (Cu Kα = 0.154 nm), β = FWHM (in radians) and θ = Bragg angle. From the structural analysis, it can be observed that the $CH_3NH_3SnI_3$ powder shows crystallinity of 96 % and that of $CH_3NH_3SnI_3$ thin film is 95.61 %, improved crystallinity is attributed to the controlled spin-coating process and optimized annealing conditions. It is confirmed that the perovskite film is highly crystalline and phase pure, which is desirable for its application in optoelectronic devices.

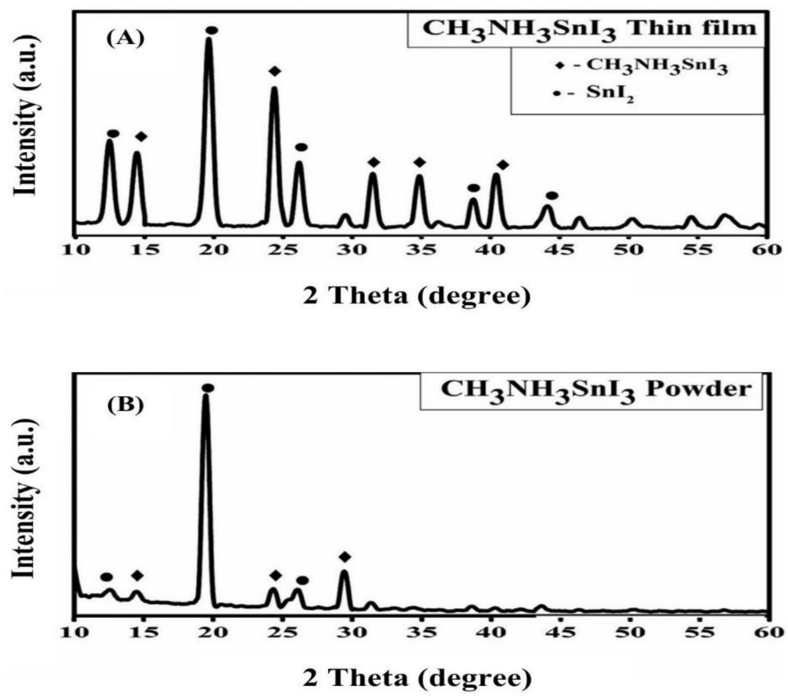

Fig.2. XRD pattern of (A) $CH_3NH_3SnI_3$ thin film, (B) $CH_3NH_3SnI_3$ powder

3.2 Optical properties

The optical properties of the $CH_3NH_3SnI_3$ thin films were investigated using UV–visible absorption spectroscopy in the wavelength range of 500–900 nm. The absorption spectrum revealed strong absorption in the visible region, which is a desirable characteristic for photovoltaic absorber materials. The absorption edge of the film was observed around 900 nm as shown in Fig.3, indicating a narrow bandgap suitable for efficient solar energy harvesting [17]. To determine the optical band gap, a Tauc plot was constructed by plotting $(\alpha h\nu)^2$ vs photon energy $(h\nu)$, assuming a direct allowed transition [18].

$$\alpha = \frac{2.303 \times A}{t} \qquad (2)$$

$$(\alpha h\nu)^{\gamma} = B\left(h\nu - E_g\right) \qquad (3)$$

where, α = absorption coefficient, A = absorbance, t = thickness, h = Plank's constant, υ = frequency, γ = 2 for direct band gap semiconductor and Eg = band gap. The linear portion of the curve was extrapolated to intersect the energy axis, and the band gap was estimated from this intercept as shown in the Fig.3(b), the optical band gap (E_g) is found to be approximately 1.31 eV. This value is consistent with reported band gaps for tin-based perovskites and confirms that $CH_3NH_3SnI_3$ is a narrow band gap semiconductor. The relatively low band gap allows efficient absorption of a broad portion of the solar spectrum, particularly in the visible and near-infrared regions. This bandgap value is highly favorable for solar cell applications, as it allows efficient absorption of a large portion of the solar spectrum.

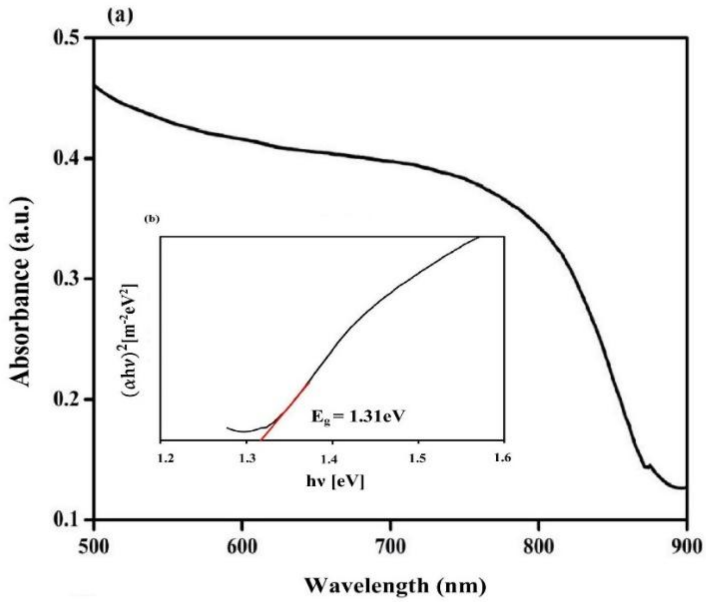

Fig. 3 (a) UV–vis spectra of $CH_3NH_3SnI_3$ thin film and (b) Tauc plot

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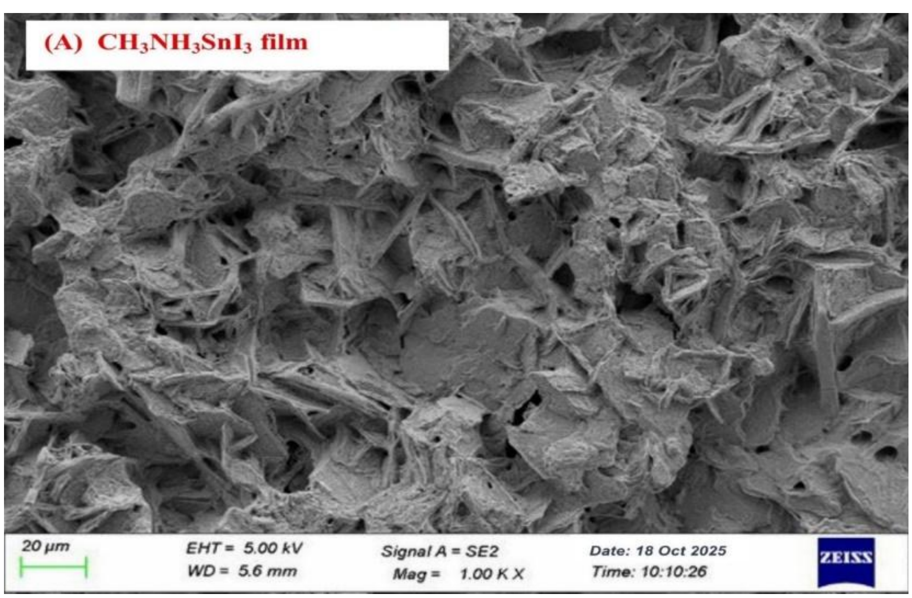
3.3 Morphological analysis

The SEM spectra of $CH_3NH_3SnI_3$ thin films and powder are shown in Fig.4 (A) and (B). The $CH_3NH_3SnI_3$ thin film demonstrates a well-covered surface morphology, indicating complete surface coverage and good film formation. Such morphology suggests controlled nucleation and growth during the film deposition process, leading to improved structural quality [19]. On the other side, the $CH_3NH_3SnI_3$ powder shows an aggregated morphology composed of irregular flake-like particles. Although the powder consists of larger clusters, the thin film exhibits better surface uniformity and improved packing density, which are essential for device applications.

The surface roughness analysis further supports these observations [20]. The perovskite powder exhibits an average roughness of 42.9 nm, whereas the thin film shows slightly lower values of 39.1 nm**,** indicating a comparatively smoother and more homogeneous surface. Overall, the SEM analysis confirms that the prepared $CH_3NH_3SnI_3$ thin film possesses good quality morphology, reduced roughness, and absence of defects**,** making it suitable for high-performance optoelectronic applications.



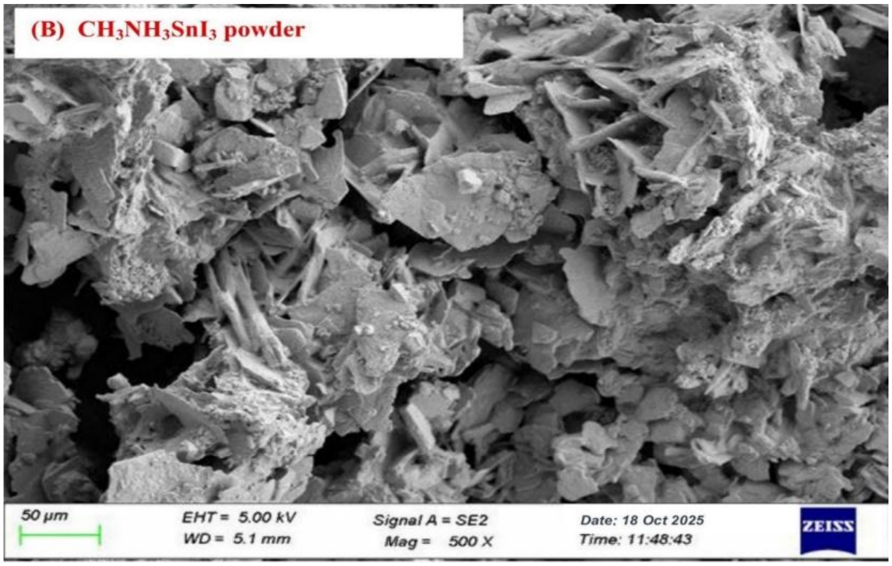


Fig.4. SEM image of (A) $CH_3NH_3SnI_3$ thin film and (B) $CH_3NH_3SnI_3$ powder.

4. CONCLUSION

A simple and effective strategy for fabricating lead-free $CH_3NH_3SnI_3$ perovskite thin films has been demonstrated using commercially available precursor materials. The thin films were deposited via spin coating followed by thermal annealing to promote crystallization and improve film uniformity. Structural analysis confirmed the formation of a perovskite phase with good crystallinity. Optical measurements indicated strong absorption in the visible region with a bandgap around 1.31 eV, which is suitable for photovoltaic applications. Morphological investigation revealed better films with minimal surface defects. The results suggest that this approach provides a practical route for preparing good-quality lead-free perovskite absorber layers. Such materials have strong potential for application in environmentally friendly photovoltaic devices and other optoelectronic technologies.

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Conflict of interest

The authors declare that there is no conflict of interests regarding the publishing of this paper.

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