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Effect of Annealing Temperature on the Morphology, Structure and Optical Properties of Spin-Coated SnO_2 Films for Solar Cell Application †

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Abstract: Perovskite solar cells (PSCs) have rapidly become a hot area of research in the photovoltaic field due to their (e.g., 26.2%), ease of fabrication, and low cost. Over the last decade the electron transport layer (ETL) has been one of the most critical elements in achieving high-performing solar cells, necessitating a higher electron mobility and superior charge extraction ability. Consequently, there is a significant demand for an improved ETL that is not only cost-effective but also exhibits high charge extraction and mobility, particularly in the context of planar solar cell architecture. Tin Oxide (SnO_2) has emerged as one of the most promising high-performance inorganic ETLs suitable for PSCs. In this work, we synthesize solution-processed SnO_2. The spin-coated SnO_2 thin films undergo annealing at relatively low temperatures ranging from 130 °C to 180 °C and various characterization tools are employed for the evaluation of thin films.

Keywords: transparent conductive oxide (TCO); electron transport layer (ETL); post annealing; perovskite solar cell; charge carriers

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1. Introduction

PSCs are emerging third-generation photovoltaic (PV) devices, and serve as the frontiers of science in PV due to superior optoelectronic properties and high charge diffusion length (e.g., 175 µm) [1]. Although PSCs have shown a significant increase in PV efficiency in the last decade, they are still fairly behind the theoretical limit (31%) of single-junction solar cells due to possible structural and architectural defects and shortcomings. One of the best-practiced architectures of PSCs is planer structure, which mainly consists of an electron transport layer (ETL), perovskite absorber and hole transport layers (HTL). There is a need to optimize the whole structural elements of PSCs [1,2].

ETLs should possess some characteristic properties, e.g., transparency within the visible region, high conductivity, continuity in the film, low resistivity, and low temperature processibility [1]. There is numerous metal oxide-based ETL materials that are under the limelight in the PSC research domain, e.g., TiO_2, ZnO_2, and SnO_2, etc. Among these SnO_2 thin films is an attractive material to enhance the efficiency of planner PSCs. Tin oxide films are emerging as one of the most successful ETLs due to favorable electrical, optical, and electrochemical characteristics in addition to chemical inertness against acids and bases [2,3].

Typically, SnO_2 thin film deposition is performed by physical vapor deposition (PVD), as well as solution-processable techniques such as spray pyrolysis, spin coating dip coating, and so on. Besides the PVD method, solution-processable techniques are cost-effective and easy to perform, but this is a challenge to get pin-hole-free, highly adhered, and defect-free film formation via solution-processable fabrication under ambient conditions. High-temperature post-annealing is mostly needed after fabricating thin films [2].

The literature indicates that SnO_2 ETL films exhibit low resistivity (higher than 10^{-3}), as well as high optical transparency (more than 80%) within the visible region. It is also evident that the optical transparency rises with an increasing annealing temperature. The XRD crystallinity peaks become more intense and sharper as we increase the annealing temperature [4,5].

This study is motivated by the prevalent use of elevated temperatures (e.g., 500 °C and higher) for applying SnO2 as an electron transport layer (ETL) in perovskite solar cells (PSCs) [6,7]. However, the obstacle to utilizing this effective ETL material in commercial PSCs lies in costly deposition techniques like physical vapor deposition (PVD) and molecular beam epitaxy (MBE) [2].

The objective of this study is to make SnO_2 thin films using a cost-effective method and decrease the post-annealing temperature considerably below 200 °C to achieve comparable properties of high-temperature-annealed SnO_2 thin films [2,6]. In our research, we used the spin coating technique for film deposition because it is inexpensive and simple, and the film thickness is controllable. Additionally, the effect of post-annealing at low temperatures (130–180 °C) was performed, and the effect of post-annealing on the optical, electrical, and structural properties of SnO_2 thin films was studied to make it more suitable for application in PSCs as a low-temperature-processable ETL.

2. Materials and Methods

We synthesized the SnO_2 nanoparticle (NP) dispersion as per the previously reported method via the sol-gel reflux method [8]. The synthesized NP dispersion was then spin-coated on a cleaned glass substrate of 10 by 10 mm at 3000 rpm and subject to post-annealing at different temperatures, e.g., 130 °C, 150 °C, and 180 °C. It then underwent glass cleaning, i.e., sequential bath sonication in soap water, DI water, acetone, and ethanol for 15 min each, followed by 30 min of UV ozone cleaning.

3. Results and Discussions

The XRD analysis (Figure 1a) of the SnO_2 thin films was studied for all three samples. As per the XRD results, the crystallinity was altered from the amorphous phase because of the annealing treatment with the increasing peak intensity. The annealing temperature influences the properties of the thin film of SnO_2. When the temperature increases, the XRD pattern becomes sharper and less noisy. Decreasing peak broadening shows crystallite size enhancement. Varying annealing temperature also affects other film characteristics such as absorbance in the visible region, as evident from the given data in Figure 2b. The minor change in UV transmittance spectra also indicates the alteration in the electronic structure of the material that possibly happened due to the improved crystallinity of the material.

Scanning electron microscopy (SEM) (Figure 2) revealed the surface morphology of post-annealed films of the SnO_2 thin films. SEM analysis is one of the most prominent tools for topographical analysis; hence, the surface morphology of all three different temperature-treated thin films was analyzed at various resolutions to reveal that varying annealing temperatures cause major effects on the grain size, surface roughness, and surface coverage.

The alteration in the surface morphology could be attributed to the heightened thermal enrgy, enabling the atoms to move more freely, consequently boosting electron mobility. These freely moving electrons fill the gaps/defects, promoting grain growth. Due to the reduction in the defects, the surface becomes smoother. But at higher temperatures (180 °C), the grain growth is non-uniform and additional phases are evident in the SEM graph (Figure 1c), possibly due to the phase transition of SnO_2 from rutile to cassiterite. On the other hand, good surface coverage and highly crystalline film show improved charge transport and extraction capabilities, along with films treated at 150 °C (Figure 1b), which show a good surface area and grain size.

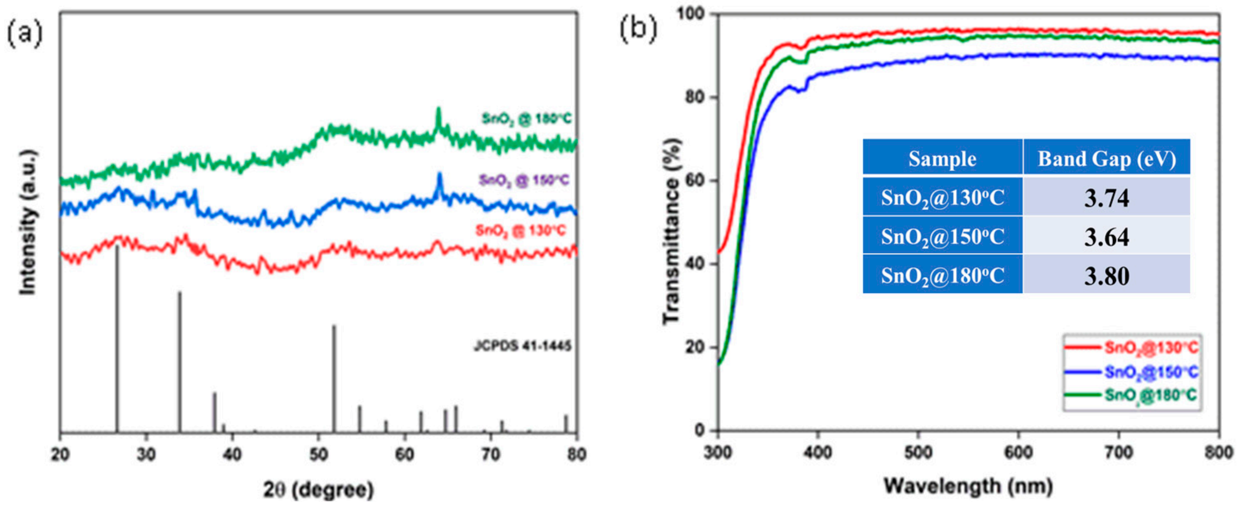

Figure 1. (**a**) XRD analysis of the SnO_2 thin films post-treated at various temperatures. (**b**) UV-Vis absorbance spectra of the SnO_2 annealed thin films.

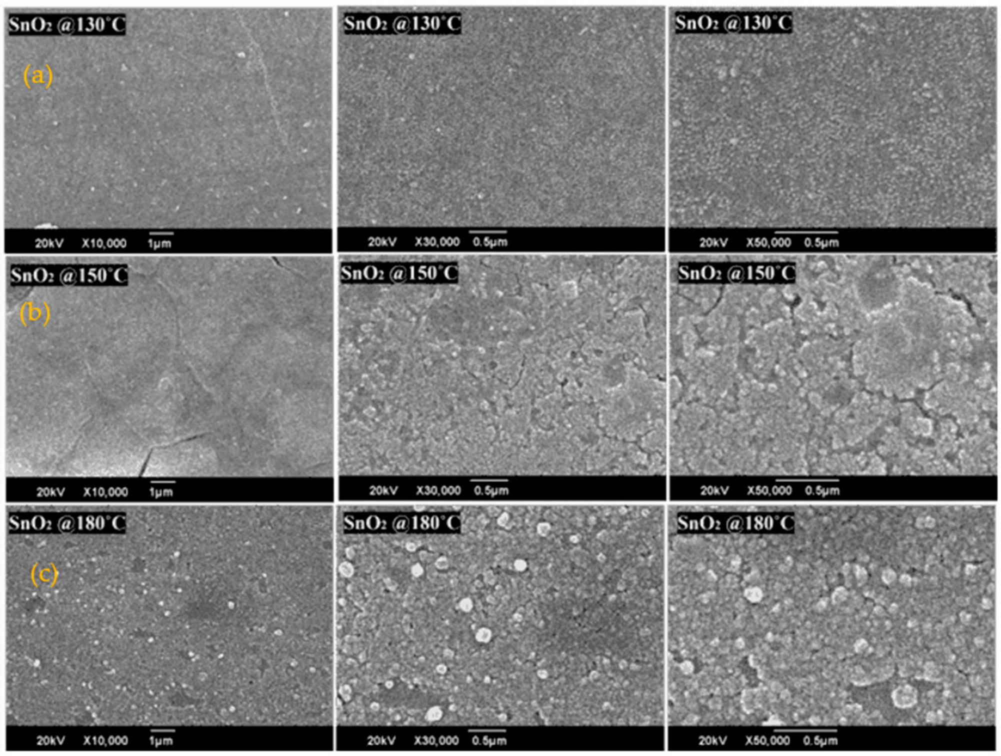

Figure 2. SEM images of the SnO_2 thin film post-treated at (**a**) 130 °C; (**b**) 150 °C; and (**c**) 180 °C.

Additionally, good crystallinity will also improve conductivity, as evident from the hall effect measurements (Table 1). Hall effect data reveal that 150 °C-annealed film shows enhanced sheet resistivity and high charge carrier mobility, possibly due to an increase in grain size.

Table 1. Hall effect measurements of the SnO_2 thin films at varying temperatures.

Parameter	SnO_2@130 °C	SnO_2@150 °C	SnO_2@180 °C
Sheet Resistance (Ohm/cm^2)	1.909×10^9	4.151×10^8	5.277×10^8
Charge Carrier Mobility (cm^2/Vs)	8.037×10^{-2}	6.932×10^1	3.824×10^0
Bulk Concentration (/cm^3)	-4.068×10^{15}	-2.169×10^{13}	-3.093×10^{14}

4. Conclusions

SnO_2 thin films exhibited enhanced charge carrier and optical properties when subjected to treatment at 150 °C. A low-temperature-processable electron transport layer (ETL) was fabricated, demonstrating a high surface area, high transmittance in the visible region, good film coverage, and high charge carrier mobility observed at 150 °C. Fabricated SnO_2 thin films showcase promising potential for application as the ETL in PSCs. However, further performance evaluation is required specifically in a functioning solar cell to comprehensively assess their efficacy and suitability.

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