

PREPARATION OF TIN OXIDE THIN FILMS BY CHEMICAL BATH DEPOSITION TECHNIQUE

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Abstract

To improve the performance of the perovskite solar cells (PSCs), the bottom Electron Transport Layer (ETL) is plays a critical role in extracting and transporting the photo-generated electrons as well as blocking holes to reduce charge recombination. In the present work, SnO₂ thin films were prepared by chemical bath deposition method. The optical properties and surface properties of resulted films were characterized by using UV-vis spectrophotometry and water contact angle measurements. The optical band gap of SnO₂-CBD (180°C) and SnO₂-CBD (500 °C) are found to be 3.42 eV and 3.88 eV respectively. Excellent transmittance of SnO₂-CBD (180 °C) film showed over 90 % than that of SnO₂CBD annealed at 500 °C. Hydrophobic properties of the surface are closely related to the crystalline behavior of the SnO₂ films at 180 °C confirmed by Water Contact Angle measurement.

Keywords: Chemical bath deposition, thin films, SnO₂, Energy Gaps, Transmittance

Introduction

Perovskite solar cell (PSCs) have rapidly popular in photovoltaic field due to its high efficiency, easy fabrication and low cost. The power conversion efficiency (PCE) of PSCs swiftly raised from $\approx 3.8\%$ in 2009 to 26.7% in 2023 due to their remarkable photovoltaic properties (Liu et al., 2023). Basic structure of perovskite solar cell consists of transparent conductive oxide (TCO), electron transport layer (ETL), perovskite absorber layer, hole transport layer (HTL) and the electrode.

Currently, numerous n type metal oxide such as TiO₂, SnO₂ and TiO₂ have been explored as ETLs fabricated in PSCs. Low-temperature processed semiconductor metal oxides, such as zinc oxide (ZnO), cadmium selenide (CdSe) and ternary metal oxides have been explored as a prospective contender to replace TiO₂ since it must be post-treated at high temperature (Sugapriya et al., 2013). Among them, the interests of SnO₂ thin film in fabrication of ETL are composed of the primary factors such as appropriate energy matching, high optical transparency, good electron mobility and high electrical conductivity, which can enhance the efficiency, stability due to reduce the recombination loss (Xiong et al., 2018).

In order to obtain SnO₂ thin films, has been many different methods such as hydrothermal method, spray-coating, spin-coating, blading coating, slot-die coating, physical vapor deposition, chemical bath deposition and atomic layer deposition have been commonly operated (Hoang Huy & Bark, 2023). Different preparation methods for ETL exhibit different morphologies which influencing the performance of solar cells. The optical properties of thin films depend on film thickness, deposition method, surface properties and substrate temperature (Park & Zhu, 2022).

Thanks to chemical bath deposition method has advantages such as low cost of production, low temperature processing, easy to handle and large area deposition.

In the present work, SnO₂ thin films has been prepared by chemical bath deposition method. Optical properties of SnO₂ film showed the highest transmittance at 180 °C than that of

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500 °C fabricated in ambient air. Energy gap was increased with increasing temperature from 3.42 to 3.88 eV. Morphology of SnO₂ annealed at 180 °C showed relatively close to hydrophobic nature measured by water contact angle measurement. It may be attributed to improve morphology due to compact and thin SnO₂ layer was formed. Hence, low temperature processed SnO₂ films prepared by Chemical Bath Deposition Method are potential candidate for efficient electron transport layer for perovskite solar cells.

Materials and Method

Materials

Urea (CH₄N₂O, ACS reagent, 99.0%), Tin (II) chloride dihydrate (SnCl₂.2H₂O, ACS reagent, 98%) and Deionized water were purchased by Sigma Aldrich. Concentrated hydrochloric acid (37 %HCl) and Sodium Hydroxide were bought by Aladdin Scientific. FTO conductive glasses (TEC7 Glass Plates 2.2 100mm x 100mm) were purchased by Great Cell Solar, Austria.

Preparation of SnO₂ Thin Films at 180°C by Chemical Bath Deposition Method

In this procedure, SnO₂ thin films were fabricated for low temperature using chemical bath deposition method (Bu et al., 2018). In this method, 625 mg of urea (CH₄N₂O) was firstly dissolved into 50 ml of deionized water, followed by the addition of 625 µl hydrochloric acid. Finally, 137 mg of tin (II) chloride dihydrate power (SnCl₂.2H₂O) was dissolved in the solution at a concentration of 0.012 M and then stored in fridge for 3 days before use. The ascleaned FTO glass was soaked into the diluted SnCl₂.2H₂O solution (0.012M) by chemical bath deposition and then annealing 70°C at 2h. SnO₂ thin films washed by deionized water and dried. The films were annealing at 180°C on the hot plate for 1h. The preparation steps of SnO₂ via CBD method at 180 °C were showed in Figure 1.

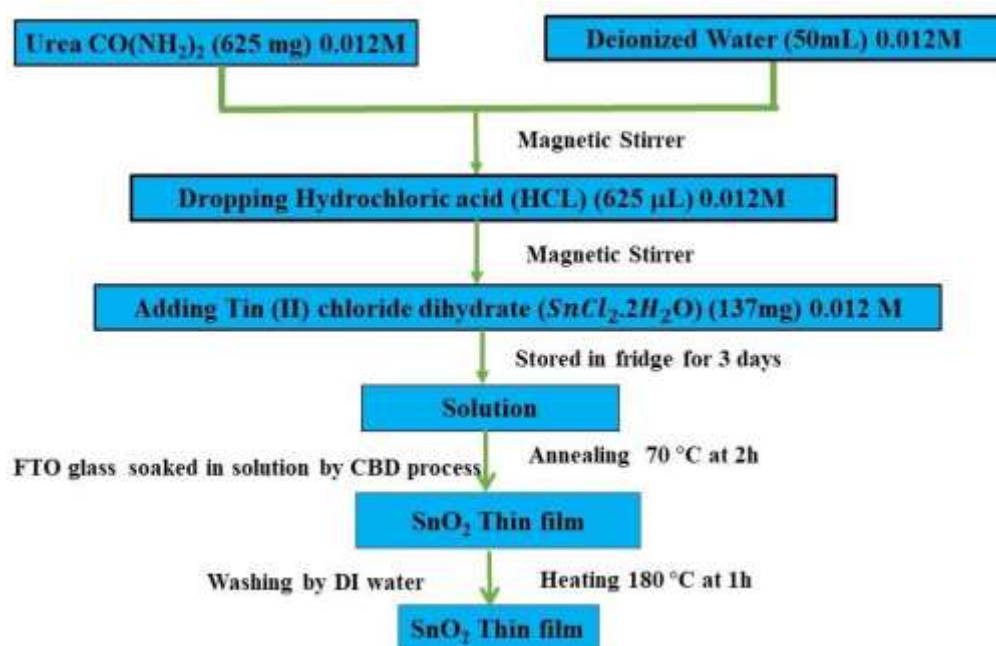


Figure 1. Preparation steps of SnO₂-CBD thin film at 180 °C

Preparation of SnO₂ Thin Films at 500 °C by Chemical Bath Deposition Method

In this procedure, SnO₂ thin films were prepared for high temperature by using chemical bath deposition method (Rahman et al., 2021). In this method, 113 mg of tin (II) chloride dihydrate (SnCl₂·2H₂O) was dissolved into 200ml of deionized water and 60 mg of urea (CH₄N₂O) was dissolved into 15ml of deionized water. These two solutions were mixed together and stirring 30 minutes and 1M of sodium hydroxide (NaOH) was added drop wise to the solution until the PH value reached at 12, stirring the solution. The cleaned FTO glass immersed in solution at room temperature for 24 h by chemical bath deposition and then annealed at 50°C at 1h. SnO₂ thin films were washed with deionized water and dried as a twice. The films were annealed at 500°C for 2h. The preparation steps of SnO₂ via CBD method at 500 °C were showed in Figure 2.

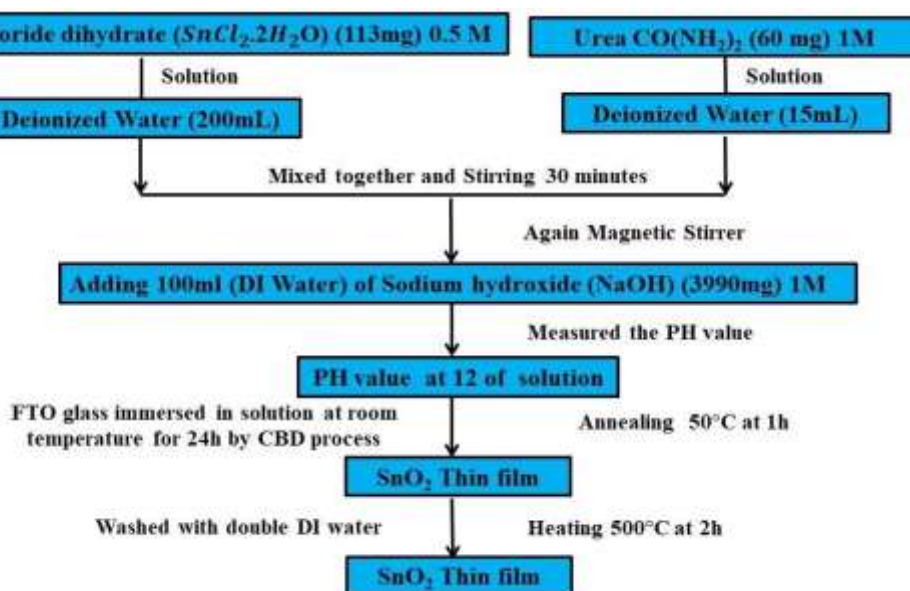


Figure 2. Preparation steps of SnO₂-CBD thin film at 500 °C

Characterization

The SnO₂-CBD thin films were characterized using a UV-vis spectrophotometry (Genesys 10S UV-vis) at wavelength range from 200 to 900 nm, with spectral resolution of 0.1 nm. In addition, the SnO₂-CBD thin films were investigated using a Contact Angle Goniometer (L2004A1, Ossila).

Results and Discussion

Optical Properties and Band Gap Energy of SnO₂ Thin Films

The optical absorption and transmission spectra of SnO₂ thin films were measured between 200 nm to 800 nm by UV- vis spectrophotometry. The UV-Vis spectra for SnO₂ thin films using different temperature were shown in figure (1), (2), (3) and (4). SnO₂-CBD (180 °C) film exhibits highest transmittance than rigid glass substrate which has high roughness surface also leads to light scattering increasing the path of the incident light and leading to a higher probability of photon absorption. SnO₂-CBD annealed at 500°C film exhibits lower transmittance and higher band gap than low temperature (180°C) annealed film. It may be attributed the presence of cracks in high-temperature growth. In order to find out, the optical band gap energy of rigid glass substrate (FTO) and SnO₂ thin films were investigated from Tauc's plot equation, $(\alpha h\nu)^n = A(h\nu - E_g)$, where α is absorption coefficient, $h\nu$ is the energy of absorbed photons, A is the proportionality constant and E_g is the band gap of the material. The exponent 'n' inspects the

transition type of the material. The value of n is 2 for direct transition of SnO_2 thin films. Figure 2 and figure 4 shows the plot of the $(\alpha h\nu)^2$ versus energy (eV) graph. The value of the band gap energy for FTO, SnO_2 film at 180°C and SnO_2 film at 500°C were found 3.89 eV, 3.42 eV and 3.88 eV.

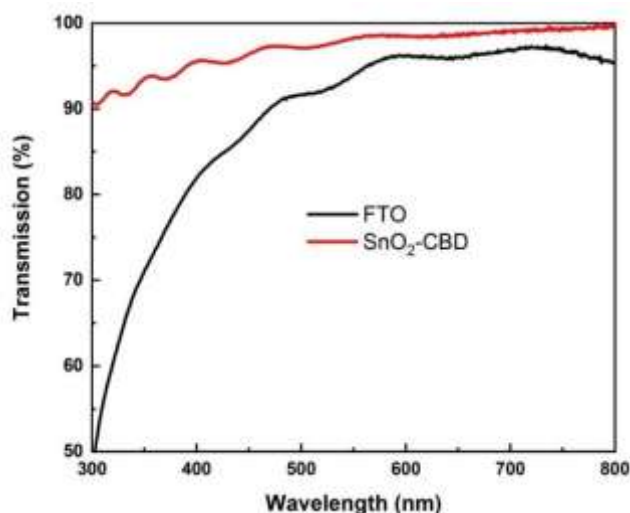


Figure 3. UV-Vis transmission spectra of FTO substrate and SnO_2 films at 180°C

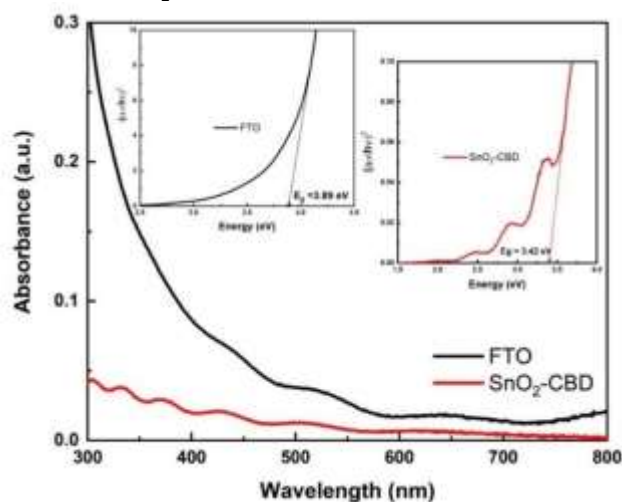


Figure 4. UV-Vis absorption spectra under temperature at 180°C of SnO_2 thin films. Zoom images showed energy gaps of FTO and SnO_2 films.

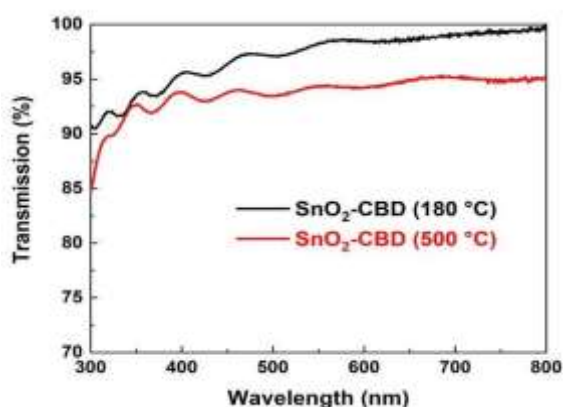


Figure 5. Transmission spectra of SnO_2 thin films at 180°C and 500°C

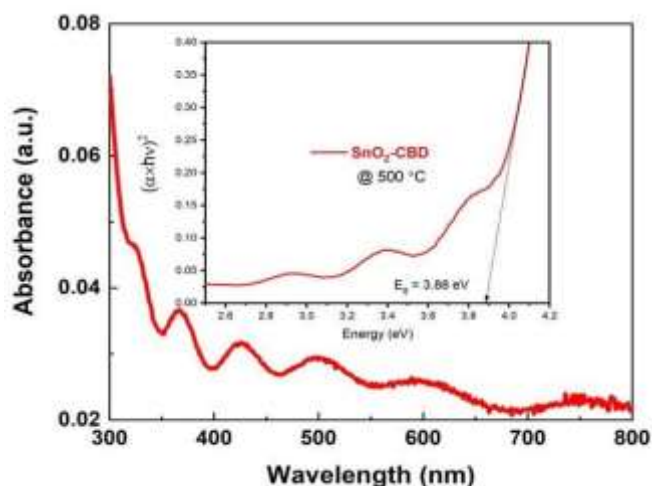


Figure 6. UV-Vis absorption spectrum of SnO₂ thin films at 500 °C. Zoom image showed a energy gap of SnO₂ at 500 °C

Water Contact Angle Measurement of SnO₂ Thin Films

The water contact angle (WCA) is the angle formed between a water droplet and a solid surface. The water contact angle of less than 90° and greater than 90°, the surface was determined hydrophilic and hydrophobic. The SnO₂-CBD at 180 °C film shows an average contact angle of 69.89° shown in Figure 6, which is higher than pure FTO film which has an average contact angle of 44.74° as shown in Figure 5. WCA confirm that smooth surface (SnO₂) may contribute to reducing the formation of large clusters and decreasing defects whereas, pristine fluoride doped tin oxide (FTO) substrate is responsible for reducing surface wettability.

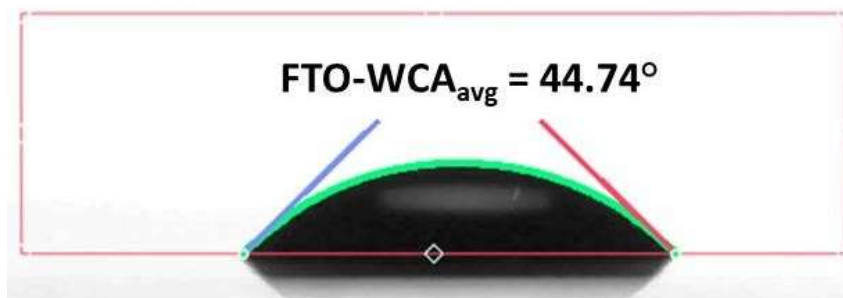


Figure 7. WCA measurement of FTO glass

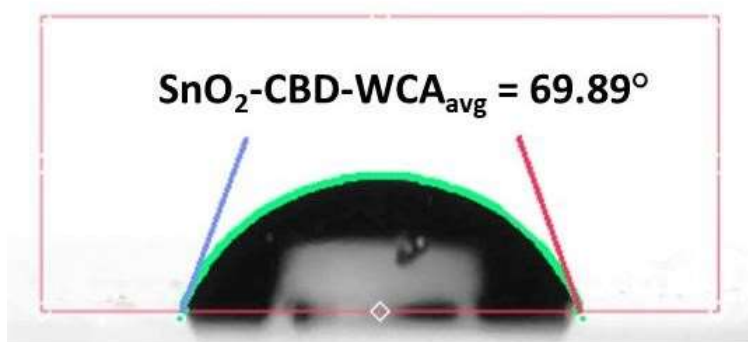


Figure 8. WCA measurement of SnO₂-CBD film at (180°C)

Summary and Conclusion

Excellent transmittance of over 90% of SnO₂ thin films in FTO glasses at 180 °C and 500 °C were prepared by Chemical Bath Deposition Technique. It was found that higher transmittance and lower energy gap were achieved in low temperature processed SnO₂-CBD film. Hydrophobic property of the surface was closely related to the crystalline behavior of the SnO₂ films at 180 °C confirmed by water contact angle measurement. Furthermore, post treatment spin coated technique via CBD will be applied for efficient SnO₂ films.

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