Optimum Substrate Temperature for SnO2Thin Films Suitable for Photovoltaic Devices

S.Sivathas [1], S. Murugan [2], A. Victor Babu[3]*, S. Ramalingam[4], N. Mahendran[5], D.C. Bernice Victoria[6]

Abstract

In this work, the investigation is made to study the peculiar properties of SnO2 material thin films using the most popular spray pyrolysis technique with respect to substrate temperature in steps of 100 oC from 300 oC to 600 oC. The structural, morphological, and optical analyses on deposited thin films are carried out to explore the intellectual application of the films. The well-defined crystallinity appears on the crystal at a substrate temperature of 500oC which explicit tetragonal rutile phase structure configuration on the basis of added lattice configuration; it is proved by the observation of the selected diffraction peak. The optical transmittance increase with the improvement of substrate temperature and very high intensity is attained at 500oC. The threshold level of crystal formation begins at 450ºC and the breakdown of the structure happens at 600ºC. In addition, the quantity of light radiation absorbed by the sample surface is high even in the ultraviolet region of the spectrum. The observed frequency shows the consistent formation of the bond structure of Sn-O. The functional bandgap with respect to the temperature is estimated and the wide bandgap is measured at a sample with a temperature of 500ºC.

Key Words: SnO2 Thin films, Spray pyrolysis, Morphology, Optical Properties

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Introduction

For Photovoltaic (PV) devices, materials have been selected to act as transparent electrodes, and transparent conductors (TCs) are an acceptable material for this purpose. Transparent conductive oxides (TCOs) are the most frequently used transparent conductors[1]. The thin film technological field proved the way for preparing fantastic and trustable materials. The nano-made Transparent semi-conducting oxide materials are being used in optical energy formulated electronic devices; solar panels, light conversion displays, and optoelectronic interfaces of their efficient properties such as electrically induced conductive ability with semi-transmittance of light [2]. The attainment of such specific properties is made possible by additive doping of adverse bandgap semiconductors (Eg > 3.1eV) and pivoted advanced technologies in making use of these materials [3–8]. Indium tin oxide (ITO) is a well-known thin film material with the highest conductivity and lowest optical absorption. Though this material is high-cost, it may be replaced by viable use of applications such as transparent thin film solar modules. The other accepted TCO is an organic blended (fluorine)-doped tin oxide (FTO). This TCO plays a major role in thin film silicon solar modules as it has the property of native substrate roughness, which is essential for light handling in non-amorphous silicon. Apart from the other technological made thin film solar cell, a thin film of silicon requires a rough surface to absorb light at large angles without affecting the growth efficiency of a silicon surface with sharp edges. In addition to such properties, high optical transparency and high electronic response [9,10] are needed to work at maximum efficiency.

Corresponding author: A. Victor Babu
Address: [1,2,3,4]Department of Physics, A.V.C. College (Autonomous) (Affiliated to Bharathidasan University, Trichy) Mannampandal, Mayiladuthurai, Tamil Nadu-609305. India, [5]PG and Research Department of Physics, Idhaya College for Women (Affiliated to Bharathidasan University, Trichy), Kumbakonam, Tamil Nadu-612001, India, [6]TELC. Higher Secondary School, Pudukkottai, Tamil Nadu, India
E-mail: victornicy@gmail.com

Another accepted class (AZO) provides an environmentally free, low-cost-effective, low-intensity light-transparent conductive front contact for several photonic and industrial electronic applications[11]. In this work, a high class of transparent conductive oxides-based tin oxide has been processed and analyzed. Tin oxide is economically cheaper compared to indium oxide which is a very high cost. It is an ideal nanomaterial candidate for adverse applications and for instance, its high electro-chemical stability is mainly considered to be a remarkable advantage of SnO2 [12,13]. It enables SnO2 suitable for applications in challenging environments, for instance, it is used as a transparent and conductive electrode on photo devices or as a selectively solar transmitting coating material [14]. The rutile-based lattice structure of SnO2 is shown in figure 1. The center of the tin atom has surrounded by six oxygen atoms and Sn is placed nearly on the corners of a quasi-regular octahedral. Three tin atoms are surrounded by each oxygen atom, forming an equilateral triangle arrangement that has the lattice parameters a = b = 4.737Å and c = 3.186Å [15,16]. So many techniques have been used for the deposition of thin films, such as linear spray pyrolysis [17], induced sol-gel process [18–21], and wide chemical vapor deposition [22,23], sputtering [24,25], and pulsed-laser deposition [26]. In the present investigation, thin films of SnO2 are prepared using the spray pyrolysis technique as this technique is the low cost [27–30], and a large amount of the works are concerned with semiconductors, metals, and transparent conductive oxides are reported using this technique [31]. The Substrate temperature effect plays a major role in determining the properties of the films formed. It is generally observed that higher substrate temperatures result in the formation of better crystalline films. Composition and thickness are affected by changes in substrate temperature, which consequently affect the properties of deposited films. In this paper, an attempt is made to find the optimum substrate temperature for the deposition of SnO2 films.

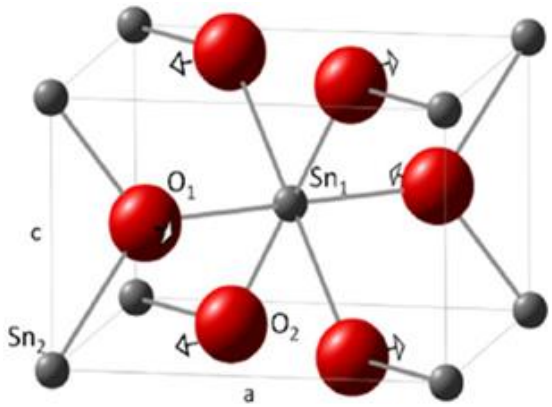

Figure 1. The rutile structure of SnO2[32].

Experimental Methods

2.1 Synthesis Of Sno2 Thin Films

On the aqueous solution, 0.1M of tin chloride pentahydrate (SnCl4•5H2O) (as a precursor) is dispersed. SnO2 thin films are deposited under the compressed air atmosphere and as a carrier gas by the self-assembled spray pyrolysis technique. Formerly, the base of substrates is cleaned with hydrochloric acid, and acetone, and sterilized with water with a suitable ultrasonic cleaner to remove organic substances. Four sets of SnO2 thin films are deposited by maintaining the substrate temperatures at 300oC, 400oC, 500oC, and 600oC, respectively.

The microcontroller spray pyrolysis setup is presented in figure 2. It basically consists of a syringe needle at the top and having a diameter of 0.6mm, connected to the reservoir. Its height can be adjusted in order to reach up to 0.002mL/minute at the needle tip. For efficient spray, a distance of 8cm is reserved between the needle and the base surface of the substrate. Usually, the substrate absorbs the precursor solution at a high voltage of about 9.65KV which is applied between the needle and the hot plate of the instrument. To optimize the thickness (500nm), different volume of precursor solution is sprayed, and the dimensions between the two surfaces of all the sprayed samples are physically verified by using a standard method. An auto-timer is connected to a high-voltage power supply through a relay and a hot plate. The timer is set in such a way

that when the hot plate is on, the high voltage will be off and vice-versa. The charge is present in the substrate holder and syringe needle. The substrate temperature and heat inside the chamber are normally proscribed using a relay, thermocouple arrangement, and exhaust fan. The distance between the syringe needle and the film is adjusted such that up and down to obtain a continuous spray to make a thin film. The FC framed alternating spray process is employed for this study at a rate of spray for 5 seconds and 10 seconds for each and every cycle.

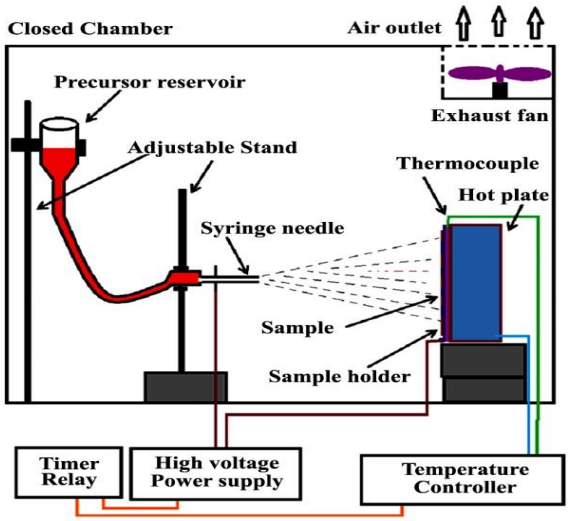

Figure 2. Schematic diagram of the Homemade spray pyrolysis setup.

Characterization Techniques

The SEM images are observed from a scanning electron microscope (CARL ZEISS) and displayed formally to analyze the properties. The molecular and material structural properties of the coated SnO2 thin films are examined by a diffused X-ray diffractometer (SHIMADZU-XRD 6000 diffractometer with X-ray (Cu–Ka) of wavelength 1.5306Å). The normal optical absorption and surface transparency of the films are measured in the wavelength range of 300–1100 nm using a Unico 4802 UV–Visible double beam spectrophotometer. Fourier transform infrared spectra are recorded by using the SHIMADZU1800-UV model FT-IR spectrometer.

Results And Discussion

3.1 Morphological Analysis

For obtaining the surface morphological data of thin films, a scanning electron microscope is used and it is a highly efficient technique that gives remarkable validation on data about the nano particle‘s shape and size. For getting useful applications of optoelectronic devices, significant physical factors such as optical and electrical properties are very much important to quantify the surface morphology of the novel TCO films. So the valuable analytical examination of the surface morphology of the film becomes most significant[33]. Figure 3. explicits the SEM micrograph image of SnO2 films at the substrate temperatures; 300oC, 400oC, 500oC, and 600oC, respectively. The acquired SEM images revealed the surface morphology is considerably affected by developing the substrate temperature [34]. The morphology of the deposited films at the substrate temperatures of 300oC and 400oC (lower temperature) reveals a rather amorphous surface texture in which the atoms and molecules are not organized properly into a definite lattice. This is consistent with the findings of Johny T. Abraham et al., [35], and only a small number of granules are observed. The polycrystalline nature of the structure of the sample at 500oC is very stable and free from molecular and atomic disorders and supported by P. K. Saikia etal.,[36]. Hence a sudden increase in substrate temperature leads to agglomeration of the particles and resulting in increased size.

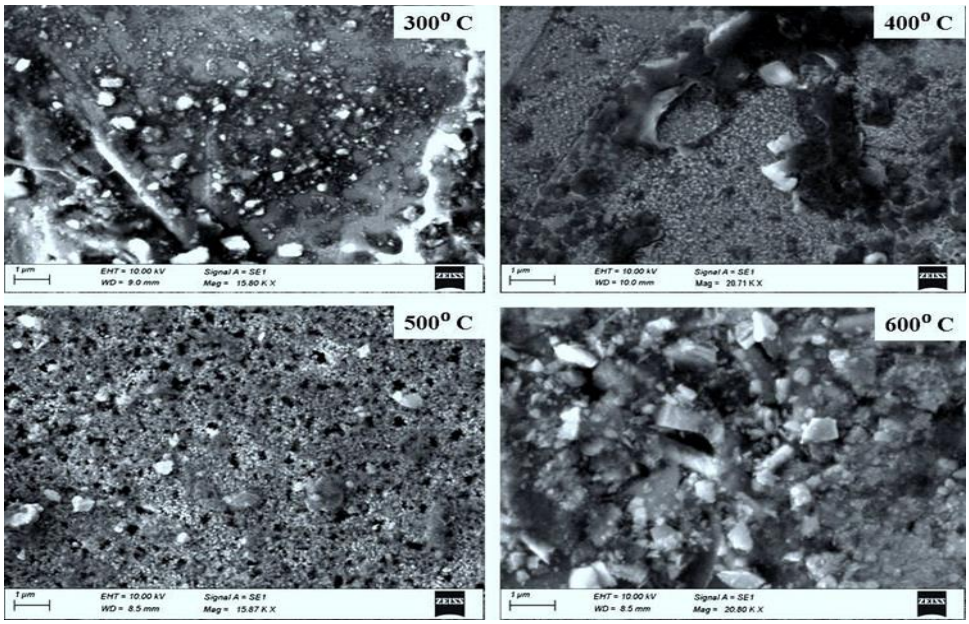

Figure 3. SEM images of SnO2 thin films for different substrate temperatures.

3.2 Structural Analysis

In order to analyze crystal structure and to estimate the average grain size, SnO2 samples deposited at various required substrate temperatures are subjected to PXRD. The XRD modulated patterns are shown in figure 4. where in which, at the substrate temperatures of 300 oC and 400 oC, no sharp peaks are observed in the patterns of the deposited films. This indicates that the films are amorphous, which is evident from morphological studies. All the peaks in the pattern of the film deposited at a substrate temperature of 500oC are sharp and clear, and they are well-matched with JCPDS card no. 41-1445[37], indicating the polycrystalline SnO2 crystals are in the tetragonal rutile phase. The peaks are not well-defined in the pattern of the deposited film at 600oC. The average particle size(D) was estimated using Debye-Scherrer's formula.

$$D = 0.9\lambda / \beta \cos\theta \quad (1)$$

Whereas usual D is the average particle size, β is the full width at half the maximum of the XRD peak in radians, and θ is the position of the sharp diffraction peak. The X-ray analysis data is listed in the table.1 the data confirms the pattern of SnO2 thin film deposited over a glass substrate with calculated h, k, and indices (110), (101), (200), and (211) corresponding to peak positions of 26.793, 34.073, 38.0856, and 51.9303, and the average crystalline size calculated from diffraction peaks is 36.245nm.

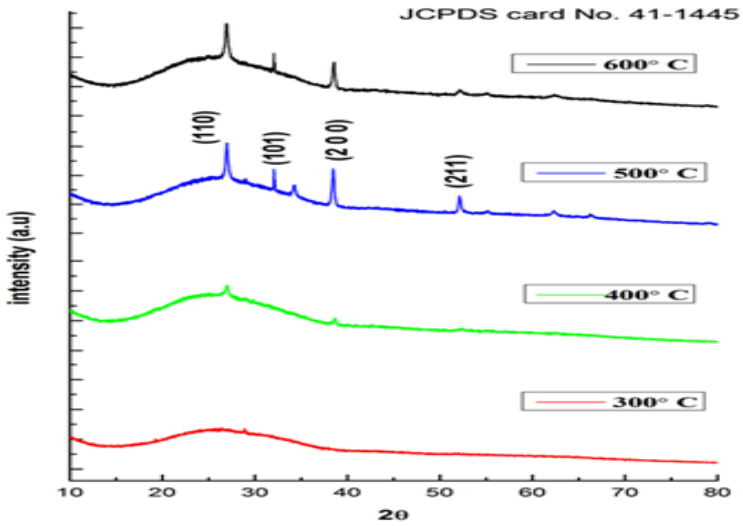

Figure 4. X-Ray Diffraction spectra of SnO2 thin films deposited at various temperatures.

Table 1. X-Ray Diffraction analysis of SnO2 thin films for 500oC substrate temperature.

(h k l) planes	Angle,2θ (degree)	d spacing (Å)	FWHM	Crystalline size (nm)
(110)	26.793	3.3248	0.204	41.83
(101)	34.073	2.6292	0.271	32.04
(200)	38.0856	2.36088	0.243	36.14
(211)	51.9303	1.75937	0.264	34.97

3.3 Optical Properties

The instant variation of transmittance peak with a specific wavelength of SnO2 deposited at various substrate temperatures is depicted in figure 5. Depending upon the substrate temperature, the average transmission in the visible region has been found to vary from 65 to 80%. For lower temperatures, the transmittance is minimum, and maximum transmittance is observed for the film deposited at a substrate temperature of 500oC. Many factors, such as the presence of mixed phases, increased thickness, the presence of defects, and the presence of oxygen vacancies, can reduce the transmittance of a film [38], but in this case, the minimum transmittance may be due to the rough surface of the films and the maximum transmittance may be due to the nature of the microstructure and surface morphology. An increase in the substrate temperature leads to a decrease in transmittance [39–42].

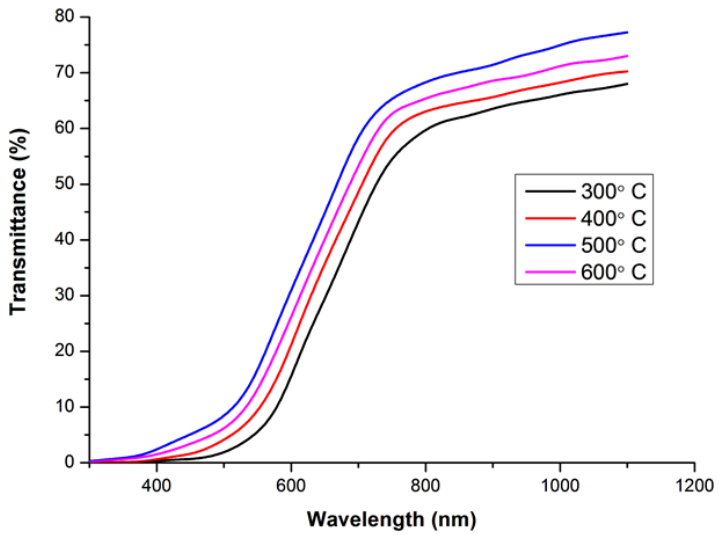

Figure 5. Transmittance spectra of SnO2 thin films.

The physical factors related to structure affecting absorption are the received photon's energy, the type of material, the crystalline structural nature, and the adoptions of impurities [43,44]. Fiureg.6 shows the optical absorption spectra of SnO2 thin films deposited at a substrate temperature of 300oC–600oC, which are almost the same. This implies that their optical band gaps are almost the same. The light energy absorption coefficient is very huge in the UV region for all the films, and it gradually low in the visible region and becomes transparent in the IR region. The behavior of the absorption spectra shows the opposite trend to that of the transmittance spectra [45].

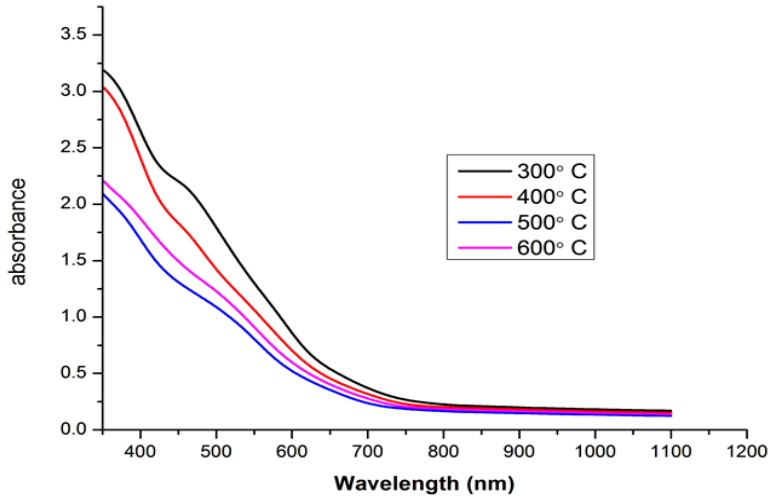

Figure 6. Absorption spectra of SnO2 thin films.

In the surrounding of the base absorption edge, the analysis of transmission and linear absorption spectra explicits the absorption coefficient which could be varied by the following relationship, which indicates the direct transition [46].

$(\alpha h\nu)2 = A(h\nu - Eg)$ (2)

Where α is a linear absorption coefficient, h is the usual Planck constant, ν is the wavenumber, and hν is the incident active-photon energy. The direct bandgap (Eg) is estimated from the absorption coefficient and it is as a function of wavelength using the Tauc relation. Thereby plotting a straight line of the (αhν)2 vs hν plot to intercept on the horizontal photon energy axis. Figure 7. depicts the plot between (αhν)2 and eV of the deposited films at the substrate temperatures of 300oC, 400oC, 500oC, and 600oC, respectively. In the SnO2 thin films deposited at a substrate temperature of250oC, 300oC, and 350oC, they exhibit direct transitions with band gap energies ranging from 3.85eV to 3.94eV [47], but in this present case, the bandgap energies range from 3.15eV to 3.50eV. The high value of the bandgap energy of 3.50eV may be due to the very smooth surface of the film.

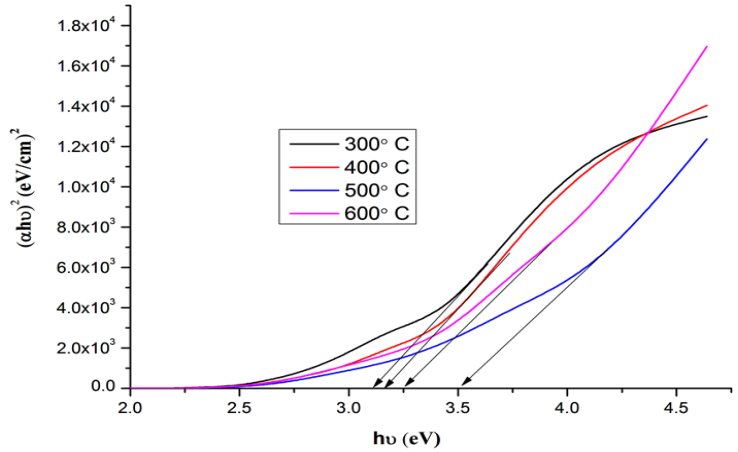

Figure 7. Plot of (αhυ)2 vs photon energy (hυ) for SnO2 thin films.

3.4 FTIR Analysis

Figure 8. shows the typical FT-IR spectrum of the SnO2 samples deposited at different substrate temperatures. Usually between 300cm−1 and 800cm−1 Sn-O stretching vibrations have been detected. For all cases, the main absorption bands appear in the range between 500cm−1 – 700cm−1, indicating Sn-O and O-Sn-O bond vibration. In all of the samples, there are some weak peaks between 1600 cm−1 and 2000 cm−1, which are associated with surface absorbed water molecules.

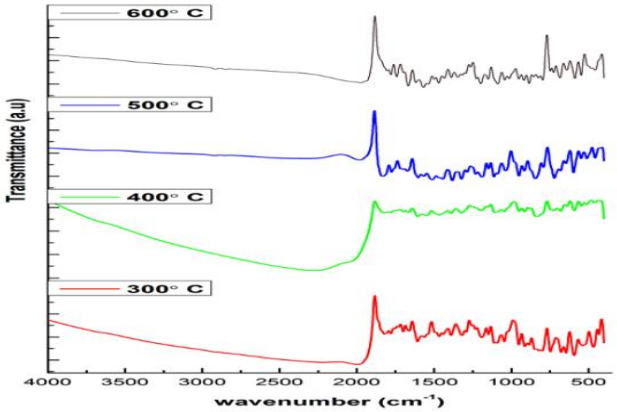

Figure 8. FTIR spectra of SnO2 thin films.

Conclusion

The nano SnO2 thin film material was successfully deposited by the low-cost self-assembled spray pyrolysis method under a suitable atmosphere in order to make efficient quality. The molecular and material structural, surface morphological, and induced optical properties of the Tin oxide thin films are investigated. The SEM reported analysis expressed that the polycrystalline nature of the structure of the present sample prepared at the substrate temperature of 500oC is very effective and surface viable for receiving light. The XRD pattern of the prepared nano compound at a substrate temperature of 500oC indicates the polycrystalline crystals which are having a tetragonal rutile phase. From the transmission spectra, it is observed that maximum transmittance is observed for the film deposited at 500oC. The light energy absorption coefficient is maximum in the UV region for all the films, moderate in the visible region, and transparent in the IR region. The enlarged bandgap energy of 3.5eV is mainly by the suitable arrangement of molecular setup in the crystal and also it validates the active morphology. The vibrational analysis confirms the active presence of strong bonds such as Sn-O and O-Sn-O. From the characterization studies of SnO2 thin films, a thin film deposited at a substrate temperature of 500oC can be suggested as a suitable candidate for photovoltaic applications.

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