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Buffer layer of antimony doped tin disulphide thin films for heterojunction solar cells

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ABSTRACT

Antimony doped tin disulphide (SnS_2:Sb) thin films have been prepared by the spray pyrolysis technique at the substrate temperature of 300 °C. The properties of the films were studied as a function of antimony dopant concentration (up to 10 at%). The XRD analysis revealed that the films were polycrystalline in nature and having hexagonal crystal structure with a preferred orientation along (002) direction. The optical energy band gap values were decreased from 2.50 eV to 2.05 eV with increase in Sb concentration and the PL spectra showed strong emission peak around at 470 nm. The film has the lowest resistivity of $1.088 \times 10^2\ \Omega$ cm while higher carrier concentration was obtained at 8 at% of Sb.

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1. Introduction

Tin disulphide (SnS_2) is considered to be one of the most useful IV–VI group semiconducting tin chalcogenides which has found applications in opto-electronic devices, a part of solar collectors, etc. It was first synthesized some 200 years ago [1] and it has more than 70 polytype structures [2]. It is a layered semiconductor with CdI_2 type structure composed of sheets of tin atoms sandwiched between two close-packed sheets of sulphur atoms. SnS_2 thin films have high optical absorption coefficient ($>10^4\ cm^{-1}$) [3], wide band gap (2.4–2.8 eV) [4], n-type electrical conduction and possession of strong photoconducting behaviour [5–7]. These characteristics make it suitable for 'buffer layer' in the fabrication of heterojunction thin film solar cells [3]. In addition, the elemental constituents of this compound are non-toxic and abundant in nature.

Thin films of SnS_2 were deposited by various techniques like atmospheric pressure chemical vapour deposition [8], SILAR [9], vaccum evaporation [10], chemical vapour deposition [11], dip deposition [5] and spray pyrolysis [3,12]. To reduce the production cost for large uniform coatings, a variety of methods are used. Among them, the spray pyrolysis is a principal, simple and promising technique to obtain uniform coating with large surface area. To improve the electrical and optical properties of the films, some dopant elements were introduced. Therefore, it is necessary to study doped SnS_2 thin films for various applications. Our group already reported the undoped and indium doped SnS_2 thin films and represented the reported work in Ref. [13]. Indium acts as acceptor ion (In^{3+}) to Sn^{4+} in SnS_2 thin films and hence the electrical resistivity has increased. We searched for another suitable doping material to improve the electrical properties and reduce electrical resistivity. In this aspect, antimony is identified as a suitable dopant with pentavelant impurity to improve the properties SnS_2 thin films. In our knowledge, no reports are available on Sb-doped SnS_2 thin films, so we tried to investigate the properties of Sb-doped SnS_2 thin films prepared by spray pyrolysis technique and to estimate their suitability for buffer layer in heterojunction solar cells.

2. Experimental details

Thin films of antimony doped tin disulphide (SnS_2:Sb) were deposited onto glass substrates by the spray pyrolysis technique. A detailed description for the preparation of SnS_2 thin films and an automated spray pyrolysis technique has been given in elsewhere [13]. The prepared solution was sprayed onto microscopic glass substrates of $75 \times 25\ mm^2$ dimensions at the substrate temperature of 300 °C. Antimony was doped with SnS_2 thin films using $SbCl_3$ (2–10 at%) as a dopant source.

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The structural characterization of the deposited films was carried out by X-ray powder diffraction technique using JEOL JDX-803 diffractometer ($\lambda = 1.5406$ Å). The surface topography of the films has been studied by Atomic Force Microscopy (AFM) using Nano-Surf Easy Scan2. Optical measurements were carried out by using a JASCO V-670 UV–vis–NIR double-beam spectrophotometer in the wavelength range of 400–1200 nm. The photoluminescence (PL) spectra were recorded at room temperature using a FluoroLog spectrofluorometer with an excitation wavelength of 255 nm. The electrical resistivity, carrier concentration and mobility were measured by automated Hall Effect measurement system (ECOPIA HMS-3000) at room temperature.

3. Result and discussion

Structural properties: Fig. 1 showed the XRD patterns of antimony doped SnS_2 thin films with different dopant concentration. All the films exhibited polycrystalline nature with preferential orientation along (002) plane. From these patterns, it was identified that no other diffraction peaks corresponding to antimony or antimony sulphides could be detected while doping. It means that Sb-incorporation does not change the crystal structure; all the SnS_2 thin films have a hexagonal crystal structure and they were preferentially oriented along the *c*-axis perpendicular to the surface of the substrate. While increasing the doping concentration up to 8 at%, the intensity of (002) peak was enhanced and the full width at half-maximum (FWHM) value decreased compared to that of undoped SnS_2 thin films [13]. This behaviour suggested that the crystalline quality of SnS_2 thin film was improved on increasing the doping concentration. It was probably due to the creation of new nucleating centres from the dopant atoms and it was favourable for the growth of SnS_2 crystals [14]. However, when increasing the Sb-doping concentration above 8 at%, the intensity of (002) peak was reduced and the FWHM value increased, which means the crystalline quality of the films was weakened. It was probably connected with the following two factors: (i) the newer nucleating centres reach the saturation; (ii) due to the difference in ionic radius between Sb^{5+} (0.65 Å) and Sn^{4+} (0.71 Å), when more Sb^{5+} ions reach the lattice sites in the place of Sn^{4+}, the lattice distortion is intensified, resulting in larger strain in the films and consequently affecting the normal growth of SnS_2.The observed peak positions and the calculated lattice parameter values were compared with the standard data (JCPDS 89-3198).

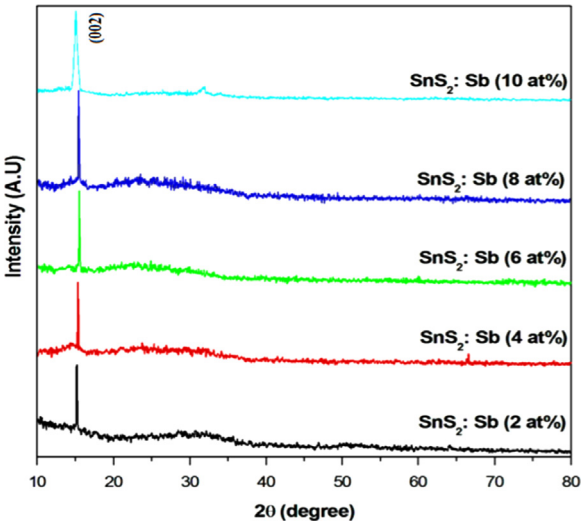

Fig. 1. XRD spectra for antimony doped SnS_2 films prepared at different doping concentrations.

The micro-structural parameters of the deposited films were calculated from the XRD data and the values have been given in Table 1. The crystallite size was found to increase with increasing the doping concentration up to 8 at% and then decreased with further doping concentration. Correspondingly, the micro-strain and dislocation density values were found to decrease with increasing Sb concentration which may be due to the improvement of crystallinity as well as specific orientation along the (002) direction. The minimum values of micro-strain and dislocation density lead to the carriers to move freely in the lattice. It can be concluded that the amount of dopant modifies the film growth process and consequently the microstructure.

The 3D view AFM image of antimony doped SnS_2 thin film (8 at% of Sb) was depicted in Fig. 2 and the image was taken in the area of 15×15 μm^2. From the figure, it was observed that the broad hills like structure of grains, which grows preferentially along *c*-axis orientation perpendicular to the surface of the substrate. The average surface roughness value was found to be 19.62 nm. The surface roughness value was higher than that of undoped and indium doped films [13]. Thus the incorporation of Sb in the films changes the surface topography. It could be attributed to the well crystallization of the films which has also been confirmed by XRD analysis.

Optical properties: The optical transmittance spectra of antimony doped SnS_2 thin films have been recorded in the wavelength range of 400–1200 nm. The energy band gap of the films was evaluated from the relation [15]. Fig. 3 showed the variation in band gap for antimony doped SnS_2 thin films with different doping concentration. It was observed that as Sb content increases, the band gap energy decreases from 2.75 eV [13] to 2.05 eV for undoped and 8 at% of Sb doped films, respectively. The decreasing trend in band gap with antimony doping can be explained by the fact that the crystallite size increased from 120.0 nm to 127.8 nm [16] and also it might be attributed to the band shrinkage effect because of increasing carrier concentration. When the doping concentration is increased above 8 at%, the band gap value increased slightly. This observation may be attributed to an effective incorporation of dopant into the SnS_2 lattice, since more

Table 1
Variations in micro-structural parameters and Hall Effect values for antimony doped SnS_2 films prepared at doping concentrations.

Sb (at%)	2θ	Micro-structural parameters			Hall Effect		
		Crystallite size (nm)	Dislocation density ($\times 10^{14}$)	Micro-strain ($\times 10^{-4}$)	Resistivity (Ω cm)	Carrier concentration (cm^{-3})	Hall mobility (cm^2/V s)
2	15.2402	122.3	0.9605	2.8096	1.368×10^3	7.451×10^{14}	89.42
4	15.2518	125.7	0.9209	2.7664	7.126×10^2	6.753×10^{15}	148.85
6	15.2613	126.5	0.9014	2.7232	3.773×10^2	1.636×10^{16}	206.89
8	15.2647	127.8	0.8783	2.6799	1.088×10^2	6.895×10^{16}	397.21
10	15.2695	124.4	0.9316	2.7674	5.618×10^2	8.060×10^{15}	270.53

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