Optoelectronic properties of evaporated antimony tin sulfide thin films for solar cell applications

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ABSTRACT
The absorber layer antimony tin sulfide thin film was deposited by physical vapor deposition (PVD) using two source simultaneous deposition techniques for solar cell applications. The photoconductivity was measured with a photoconductivity spectrometer which confirms the photo-activity of antimony tin sulfide. Thickness, band gap, refractive index and transmittance were calculated by using ellipsometry techniques. The transmittance of the material was above the visible range which decreases with the increase of wavelength. The material has n-type conductivity measured with hot probe technique.

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Introduction
Electrical energy is the most consumable energy worldwide and new methods are needed for the exploration of electrical energy. The solar cell is the best source of renewable energy which gives us energy in the form of electrical energy, as its raw material is free and abundant (Wagner and Baber, 2011). Fuel expenses are increasing day by day due to fossil fuel depletion, creating energy crises all over the world. Since the invention of the first solar cell, different materials have been used to fabricate solar cells. Foremost among these materials has been silicon, crystalline as well as the amorphous form. Silicon materials are good and efficient but the power generated from these cells is very costly. After silicon, compound semiconductors have been used to fabricate thin film solar cells. Compounds such as copper indium disulfide, copper indium gallium diselenide and cadmium telluride produce solar cells with excellent efficiencies. The approach of using these compounds for solar cell applications was ineffective however, due to the toxic nature of cadmium and non availability of indium metals (Versavel and Haber, 2007; Bonnet and Meyers, 1998; Green, Emery et al. 2011; Andersson, 2000). Researchers working on new absorbing materials with an optimum band gap and absorption coefficient need them to be cheap and abundant as well. A good absorber material can be beneficial to us as it can reduce the amount of material needed for solar cells (Tanuševski and Poelman, 2003).

For the last couple of decades, sulfosalts have been found to be the most promising materials for solar cell applications and commercialization (Gassoumi and Kanzari, 2009; Dittrich, Bieniok et al. 2007). These are low-cost and nontoxic materials of great importance for solar cell applications. We are therefore focusing our research on inventing new materials for solar cell fabrication. Tin sulfide and antimony sulfide thin film solar cell are under study for the last few years; however, they are not efficient material. So we have attempted to combine their properties in a single material called tin antimony sulfide.

Experimental
Tin antimony sulfide (SnSbS) thin films were synthesized with the thermal evaporation technique, including deposition on simple soda glass substrate using SnS and Sb2S3 as the initial materials. Sb2S3 used in the experiment was of analytical grade and 99.99% pure. SnS was obtained from powdered tin and sulfur (Massalski, H. Okamoto, 1990).

Both the powders, Sb2S3 and SnS were mixed well in a crucible and annealed at 500°C for 10 hours and evaporated from AlO2 crucibles via resistive basket simultaneously in a vacuum chamber. The chamber pressure was maintained approximately at ~10⁻⁴ mbar. After deposition, the thin films were at 85°C, 150°C and 200°C in sealed glass ampoule containing argon gas.

The elemental composition of the film was confirmed by energy-dispersive X-ray spectroscopy (EDX). Optical properties including band gap, thickness, absorption coefficient, transmittance and refractive index were evaluated by using J. A. Woollam variable angle ellipsometry (VAE). Hot point probe methods were applied to identify the type of conductivity of the film material. The photoconductivity spectrometer was used to measure the photoconductivity of the films.
Results and analysis

Energy-dispersive X-ray spectroscopy (EDS) analysis was done for the assessment of elemental composition of the thin film after annealing. Table 1 summarizes the results. Figure 1 shows the EDS spectrum of the as deposited thin film.

Table 1. Elemental composition of tin antimony sulfide thin films (atomic percentage)

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<th>Sn%</th>
<th>Sb%</th>
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<td></td>
<td>60</td>
<td>10</td>
<td>40</td>
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The XRD pattern of tin antimony sulfide thin films shows that it is polycrystalline when annealed at low temperature while the deposited film is in amorphous state. All films have strong diffraction peaks at 32° corresponding to (640) plane with strong SnSbS₄ phase (Figure 2).

It is evident from the XRD graphs that the grain size increases with annealing temperature. The crystalline size was calculated using Scherrer’s formula (Kaelble, 1967; Gassoumi and Kanzari, 2011).

\[ D = \frac{0.9\lambda}{\beta \cos \theta} \]  

(1)

Where D is the average crystallite size, \( \lambda \) is the wavelength and \( \beta \) is the full-width at half maximum (Jager-Waldau, 2011; N. Ali, M. Iqbal et al., 2012). The average grain size of our crystalline sample was 10-15 nm for the annealed films.

The photoconductivity of the film is shown in Figure 3. It shows that the annealed films have good photoconductive response and photoconductivity increases with annealing temperature. The figure shows that the thin film is highly photoactive at 200°C annealing. The film at 200°C annealed has photoconductivity extending in the near-infrared region.

The photoconductivity measurement of SnSbS₄ thin film

Figure 2. XRD of SnSbS₄ thin films

Figure 3. Photoconductivity measurement of SnSbS₄ thin film

The transmittance of the as deposited and annealed film showing that transmittance decreases with the increase of annealing temperature.

The film has short maxima and minima in the transmittance curve indicating decreases in transmittance at higher wavelength.

The refractive index can be calculated by plotting \( \lambda \) vs. n from ellipsometry data (Figure 5). It can be seen from graph that the refractive index has no observable variation on the wavelength. The refractive index is in the range 3.0-4.5.

The absorption coefficient of tin antimony sulfide thin film. The absorption coefficient starts from 4×10⁵ cm⁻¹, which is the best possible value for absorbing layers in solar cells.

The band gap can be calculated by extrapolating the straight section of the (\( \alpha h \nu \))² vs. \( h \nu \) curve to the horizontal axis (N. Ali, Z. Ali et al., 2012; Gassoumi and Kanzari, 2009).
Figure 6. Absorption coefficient vs. wavelength

The energy band gap was calculated from extrapolating the curves corresponding to the valance-conduction band transition are in the range 2.05-2.3 eV as in Figure 7.

Figure 7. Band gap calculation

The type of conductivity of the thin film was calculated by hot point probe technique. Initially, the instrument was calibrated for n-type Silicon (Figure 8(c) as a reference) and then the conductivity type of tin antimony sulfide thin film was calculated (Golan, Axelevitch et al., 2006; Plummer, Deal et al., 200). We found the similar spectrum in the graphs in Figure 8(a, b, c). It is inferred therefore that SnSb$_2$S$_4$ thin film has n-type conductivity.

Figure 8. Hotpoint probe measurement (a) as it is (b) 200°C annealed (c) Reference data of n-type silicon

Conclusion

It is concluded from this study that tin antimony sulfide thin film is a new material for solar cells that has potential for use as an absorber layer. The material is photoactive in the visible and NIR region. Its optical properties were calculated by modeling the ellipsometry data to match with the reported absorber layers. The transmittance decreases, in both as-prepared and annealed films, with good absorbance. The absorption coefficient is above $4 \times 10^5$ cm$^{-1}$ and its value is wavelength-dependent. The band gap was found to be in the range of 2.05-2.3 eV. The 200°C annealed film has good properties such as being a low temperature annealed film. The material has n-type conductivity as measured by hot point probe method.

Acknowledgements

We wish to thank the Higher Education Commission of Pakistan for the financial support of this research work and the University of Cranfield for access to the research facility. We are also thankful to the National Centre for Physics, Islamabad, Pakistan and University of the Punjab, Lahore for their financial support.

References


