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Synthesis of Antimony Telluride Thin Films by Thermal Annealing

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***Abstract:** Antimony telluride (Sb_2Te_3) has high thermoelectric performance at room temperature. It has high electrical conductivity and high thermoelectric power with low thermal conductivity. This work reports effect of annealing on optical band gap of thin films of Sb_2Te_3 compound obtained by thermal annealing of bi-layers of Antimony (Sb) and Tellurium (Te). These films having thickness of 28nm were grown on Si substrate using vapor deposition technique. The films were annealed at different temperatures of 100°C, 150°C and 200°C and the variation in optical, morphological, structural, electrical and thermo electrical properties were studied. Results indicate that good quality films of crystalline compound Sb_2Te_3, having optimum properties have been formed.*

***Keywords:** Antimony telluride, ADXRD, SEM, thermoelectrics.*

I. INTRODUCTION

The electronic systems are becoming denser hence thermal management has become necessary part of the design process [1]. Large area thermoelectric devices are now using lateral configuration (thin film) of thermoelectric materials having performance similar to that of bulk materials which is due to their low dimensional quantum size effect [2-4]. This option has a superior possibility of using them right at the source of the heat generation hence it is a promising area of research and development [5-9]. Tellurium based thermoelectric materials have been proven to be very effective in the room temperature range. Nano-scaled tellurium-based materials are expected to make a breakthrough in the present era technology [10-12]. Hicks and Dresselhaus reported that low-dimensional materials have better efficiency than bulk because of low-dimensional effects on charge carriers and lattice waves [13]. Since then a large number of deposition techniques have been employed to obtain the thermoelectric thin films, such as flash evaporation, ion-beam sputtering, pulse laser deposition, sputtering, electrochemical deposition, metal organic chemical vapor deposition, molecular beam epitaxy, and electrochemical deposition [14-15].

Sb_2Te_3 is an inorganic compound with solid crystalline structure having 580°C melting point and 6.50 gm/cm^3 density. Its melting point, color and density depend upon crystalline form it adopts. Sb_2Te_3 can be transformed into both n-type and p-type semiconductor by appropriate doping [16-17], in fact, heavily doped Sb_2Te_3 and its alloys are the best performing thermoelectric materials at room temperature [18]. These interesting properties of Sb_2Te_3 grabbed the attention of researchers for its wide range application in the field of thermoelectrics [19-20].

In this paper the synthesis of Sb_2Te_3 compound thin films by thermal annealing of bi-layers of elements Sb and Te has been reported. These films have been annealed at different temperatures i.e. 100°C, 150°C and 200°C. Typical measurements of optical, structural, morphological, electrical and thermoelectric properties of these films have been carried out using ADXRD, SEM, UV-VIS, Thermoelectric measurements and I-V methods.

II. EXPERIMENT

Powders of Te (Aldrich, 99.8%, 200 mesh) and Sb (Aldrich, 99.5%, 100 mesh) were used as received. Si wafers were carefully washed by acetone. The deposition of bi-layer thin films on silicon substrate was carried out in a vacuum chamber using vapor deposition technique. Sb was placed in a boat located downstream to Te boat and deposition done at a pressure of 10^{-5} mbar. Bi-layers (Sb-Te) were prepared for 28nm which were annealed at 100°C, 150°C and 200°C.

The bi-layer thin films on Si substrate were directly used for ADXRD and SEM measurements. The Angle dispersive X-ray diffraction was performed at beam line 12, INDUS-2, RRCAT, Indore, India. Beam line-12 is a bending magnet based, high resolution XRD beam line having 2.5GeV, 300mA with a photon energy range of 5-25 KeV. The size and morphology of the particles in the thin films were characterized using Field emission gun- scanning electron microscopes (FEG- SEM) (JEOL JSM-7600F) at INUP, IIT Mumbai, Bombay, India. The measurement of resistivity was carried out by one of the standard and most widely used four probe apparatus. The experimental set up consists of probe arrangement, sample, oven 0-200°C, constant current

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generator, oven power supply and digital panel meter (measuring voltage and current). Voltage and current readings were measured for films annealed at different temperatures. For optical study, a spectrometer (UV-VIS -NIR Spectrometer - Lambda 750, make PerkinEelmer) at IIT Bombay was used.

III.RESULT & DISCUSSION

The optical absorption was measured in the wave range 500-3600 cm^{-1}. The absorption coefficient was calculated as a function of photon energy from absorbance v/s wavelength curve while the optical band gaps were obtained from graphs between $(\alpha h\nu)^2$ v/s hν curves. Figure 1 shows plots for thin films of thickness 28nm at RT, annealed at 100, 150 and 200°C. Linear behavior of plots can be seen near the fundamental absorption edge representing the direct band gap characteristic of the compound. The band gap obtained for various films are also shown by extrapolating the linear part to the zero of the ordinate. This comes out to be 0.21eV for 100°C, 0.235eV for 150°C and 0.239eV for 200°C annealing. It can be seen that with the increase in the annealing temperature the value of the observed band gap approaches to the reported values for Sb_2Te_3 in the earlier works.

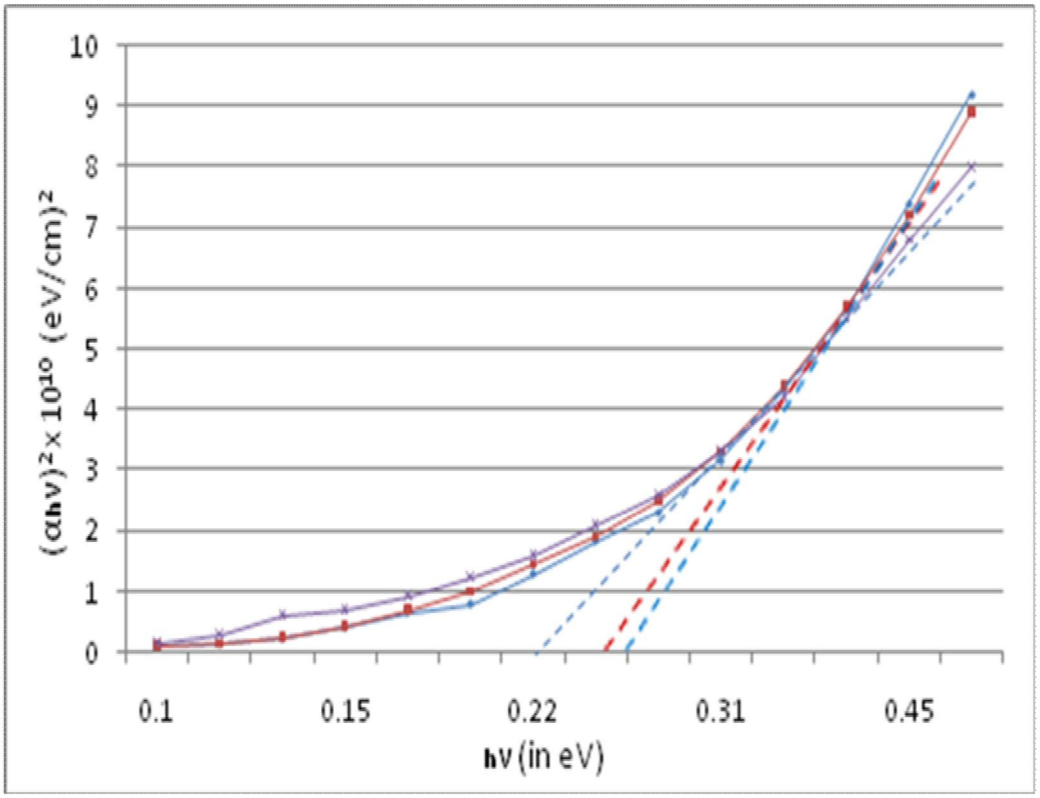

Figure 1: Plot of $(\alpha h\nu)^2$ v/s hν for films annealed to different temperatures.

Figure 2 shows the ADXRD pattern obtained for the thin films of Sb_2Te_3 for insitu annealing temperatures. The bi-layer films grown at room temperature were annealed to different temperatures and in-situ measurements were taken. In figure 2 it is revealed that the bi-layer thin films of Sb and Te annealed at 100°C, 150°C and 200°C were having crystalline nature. The graph indicates the characteristics of XRD peaks corresponding to antimony and tellurium which are observed at specific angle, corresponding to the orthorhombic face of thin films. All peaks of Sb and Si are shown in XRD and no peak of Te is shown due its non crystalline behavior after heat treatment. Also some small peaks of Sb_2Te_3 also report due to merging of Te into Sb after annealing. As annealing increase these peaks become more visible. No additional peaks have been observed after annealing which shows that film exhibit a Sb rich composition and there are no compositional variations occur after the annealing treatment of Sb and Te bi-layer thin films. The intensity of Individual peaks has been observed increasing after annealing which may be due to fact that after annealing the Sb-Te thin films are arranged in more crystalline manner due to heat treatment. From figure 2 it can be seen that there are only a few faint peaks which show almost amorphous nature of the film. As the annealing temperature is increased, the (002), (003) and (004) reflections are strengthened which represents the increase in the crystalline nature of the films.

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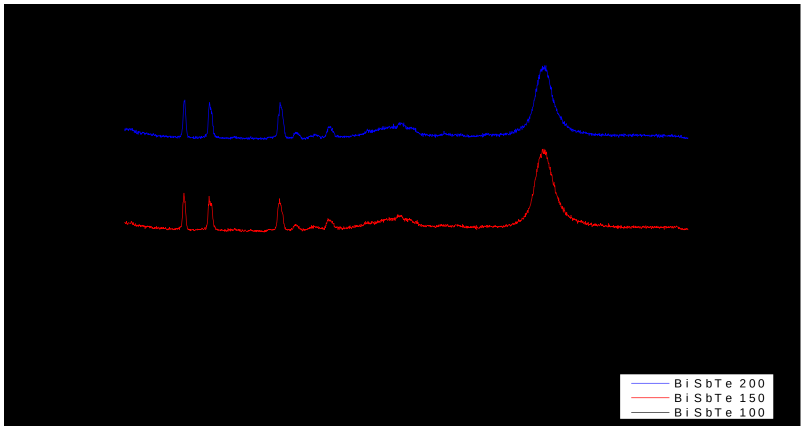
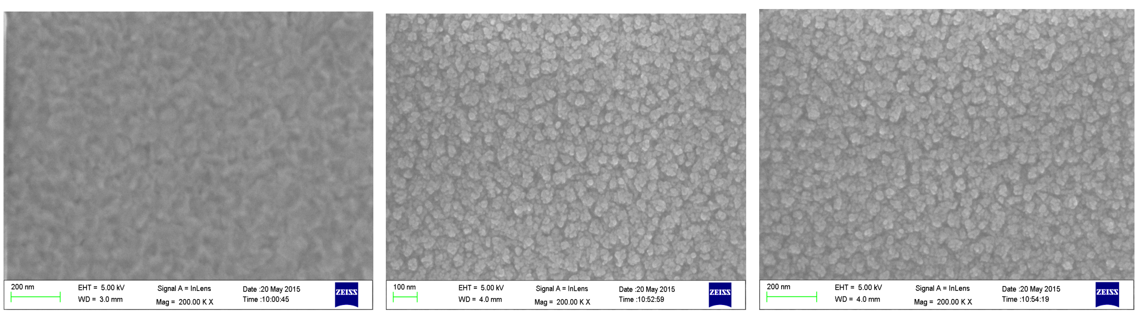


Figure 2: ADXRD measurements of thin films annealed at various temperatures.

It can be observed from scanning electron micrographs (shown in figure 3) that the surface of the films becomes more crystalline with the increase in the annealing temperature. It can be seen that at 100°C temperature surface of the thin film contains flakes type structure which when annealed to a higher temperature 150°C (b) and 200°C (c) becomes more crystalline with a particle size of about 40nm.



(a) (b) (c)

Figure 3: SEM micrographs annealed at different temperatures

The variation in the current v/s voltage (I-V) characteristics (figure 4) were taken for samples of bi-layer thin films as deposited at room temperature and annealed at various temperatures. The plot ‘a’ in figure 4 is for bi-layer film at room temperature while plots b-d are for the films annealed at 100, 150 and 200°C. The I-V curves indicate ohmic conduction throughout the voltage (from lower voltages to higher voltage region) for RT while for annealed films slop of the plots are increased; this confirms the presence of injected space charge due to high temperature.

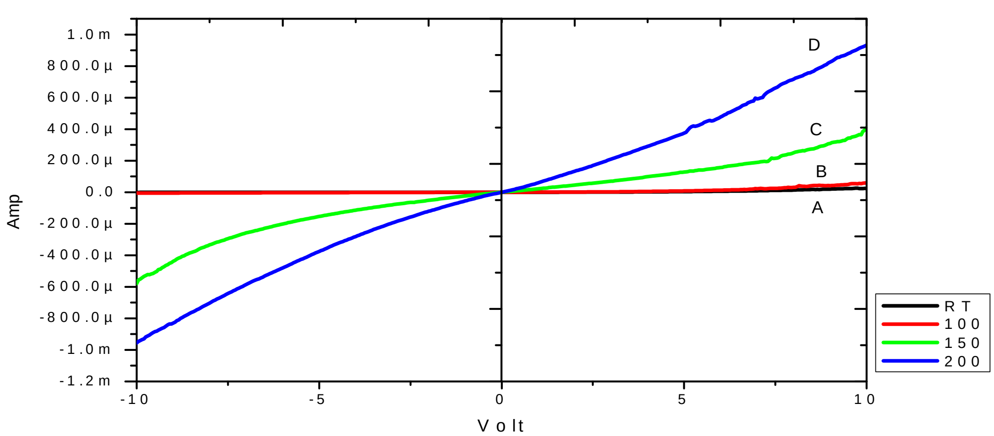


Figure 4: I-V characteristics for the various thin films.

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Figure 5 shows the result of resistivity measurement by four-probe apparatus within the range of 40-100°C. It reveals that all three types of thin films have low electrical resistivity in the range of $4X10^{-5}$ Ω-m. for thermoelectric semiconductors low electrical resistivity is essential to achieve high ZT value. For all three types of thin films the electrical resistivity decrease with the annealing temperature. It means it leads to enhance the contact between the particles in the annealed samples which is evident from the SEM observations.

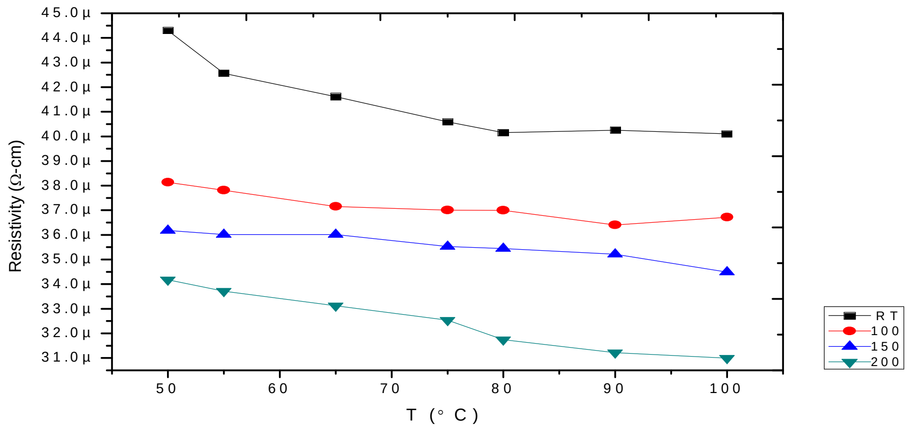

Figure 5: Resistivity measurements for the various thin films.

Dependence on the temperature of the Seebeck coefficient has been shown in figure 6. In the extrinsic conductive region, the increase in thermoelectric power with temperature is due to the increasing number of thermally excited carriers. The Seeback coefficients of the films increase with the increment in temperature. It can be realized that the Seebeck coefficient value is with positive sign suggesting P-type charge conduction. It also shows that with increase in the annealing temperature the thermoelectric efficiency of the films increases.

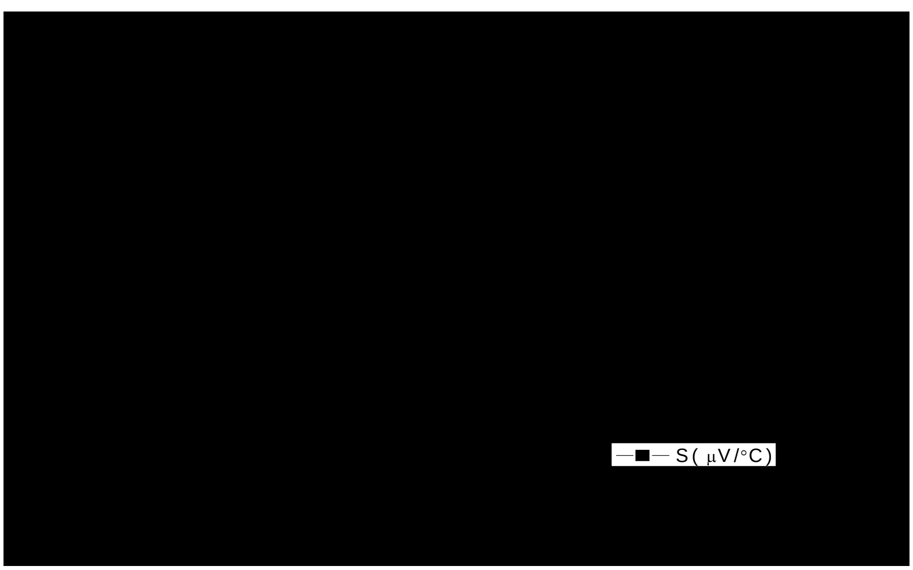

Figure 6: Variations in the Seebeck coefficient with temperature.

IV. CONCLUSIONS

Sb_2Te_3 compound thin films were deposited by vacuum evaporation deposition method on silicon substrate. ADXRD and SEM results show the crystalline and homogenous structure of films annealed for various temperatures. Optical data results are also in coherence with the reported values. The thermoelectric properties of the thin films have been evaluated via Seebeck coefficient, and electrical resistivity measurement. These results indicate that good quality compound Sb_2Te_3 thin films were grown by vacuum evaporation technique.

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V. ACKNOWLEDGMENT

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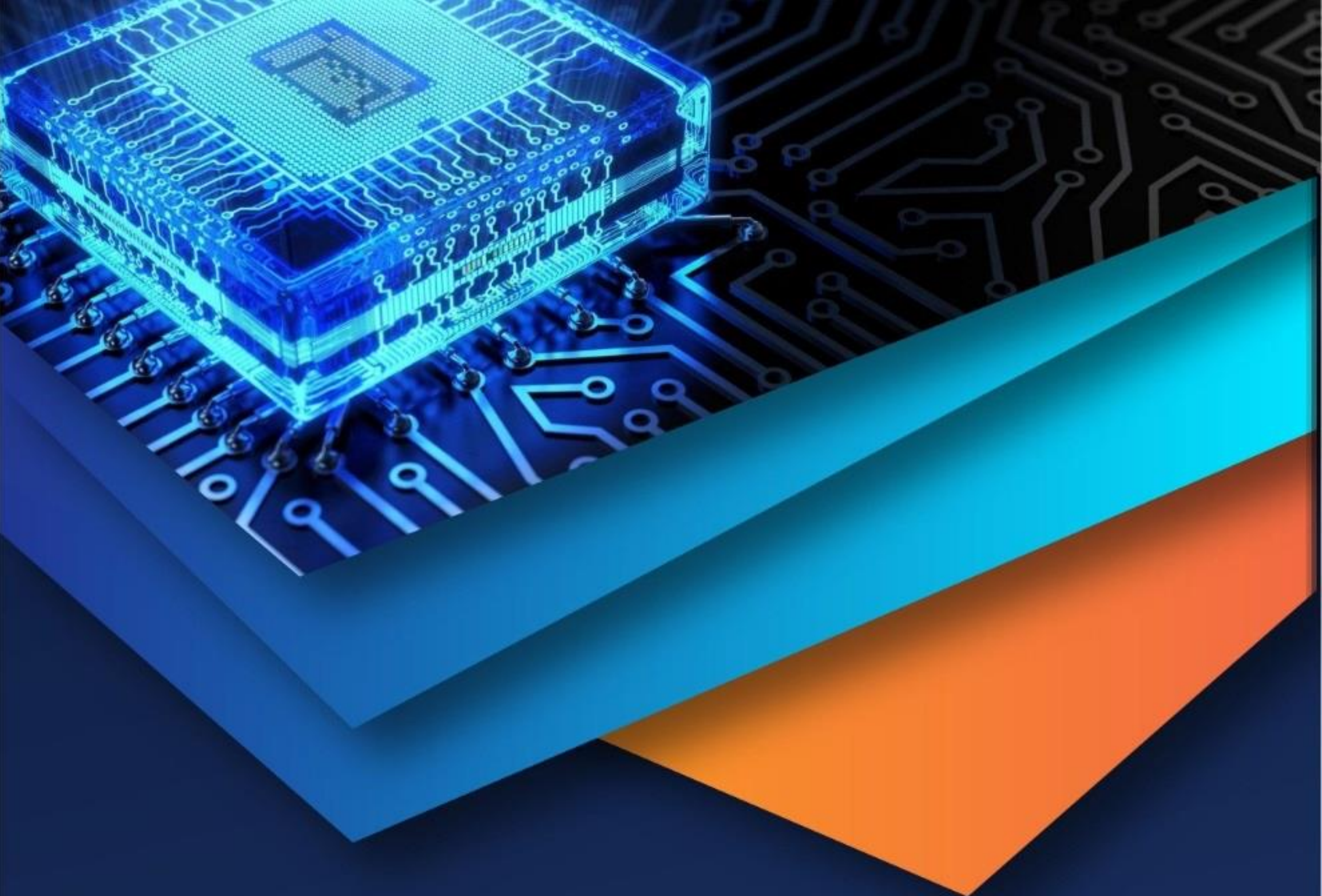

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