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Effect of substrate temperature and film thickness on the thermoelectric properties of In_2Te_3 thin films

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ABSTRACT

Herein, the thermoelectric properties of vacuum deposited In_2Te_3 thin films were investigated by varying the substrate temperature and the thickness of the films. The thermo-electro motive force of the prepared films was found to increase with an increase in the substrate temperature up to 423 K and then decrease at 473 K due to the presence of mixed-phase structure. The maximum thermoelectric power of 220 μV/K was observed for the films deposited at 423 K substrate temperature, which was found to decrease with increase in thickness. The films deposited at 423 K with 150 nm thickness showed maximum power factor of 27 $\mu Wm^{-1}K^{-2}$ at 450 K. These observations are explained on the basis of structural, morphological and compositional changes.

1. Introduction

Thermoelectric materials are the clean energy sources because of their unique ability to harvest electricity directly from the waste heat. The efficiency of a thermoelectric material can be expressed in terms of a dimensionless quantity called figure of merit [1,2] as given by $ZT = \frac{S^2\sigma T}{k}$, where S, σ, k and T indicate Seebeck coefficient, electrical conductivity, thermal conductivity and absolute temperature, respectively. A good thermoelectric material should have high power factor ($S^2\sigma$) at lower thermal conductivity [3]. The thermal conductivity (k) comprises the combination of heat carried by both phonon and charge carriers. Significant progress has been made in the recent years to increase ZT, especially by controlling the dimensions of a material to nanoscale. At nanoscale regime the reduction in the thermal conductivity due to the increased phonon scattering at grain boundaries and distortion in electron density of states, leads to the enhancement in the thermoelectric power [4].

A good thermoelectric material should have the electrical properties of crystalline material and thermal properties of an amorphous material [5,6]. Hence, the metal chalcogenides, such as Bi_2Te_3, PbTe, and SnSe have been broadly exploited for thermoelectric application. Metal chalcogenide thermoelectric materials exhibiting a superior power factor compared to oxide counterparts, because of their low electronegativity. In addition, the heavy atomic weight associated with these compounds reduces the thermal conductivity [7]. Indium telluride (In_2Te_3) is also a metal chalcogenide with defective zinc blend structure. This material has defect density of the order of 10^{21} cm^{-3} with one-third of the cation sites vacant due to valence mismatch between cation and anion [8,9]. These vacancies are orderly distributed in α-In_2Te_3 whereas randomly in β-In_2Te_3 [10–13]. However, such vacancies in crystal sites causes a significant reduction in the lattice vibrations due to strong phonon–vacancy scattering. Reportedly, the thermal conductivity of In_2Te_3 crystal is 1 W m^{-1} K^{-1} at 450 K [14]. This value can be tailored by reducing one of bulk dimensions to nanoscale regime i.e., depositing nanostructured thin films, because of the expected enhancement in the phonon scattering at grain boundaries. Moreover, In_2Te_3 has high radiation stability of electronic parameters after ionization fluences which was examined experimentally up to 10^{18} fast neutrons per cm^2 [15]. Hence this material can be used in thermoelectric power generators in the vicinities of nuclear reactors. Despite of its ample characteristics to exhibit good thermoelectric properties, research interest invested in this direction on In_2Te_3 thin films is still meager. Lakshiminarayan et al. (2002) [16] investigated the thermoelectric power in stoichiometric In_2Te_3 thin films. They observed that the

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thermoelectric power is nearly independent on the hot-end temperature (300–420 K) and dependent on the thin film thickness predicted by the size effect theories.

In the present work, an attempt has been made to find the best possible condition to enhance the thermoelectric power factor of In_2Te_3 films. The influence of substrate temperature and film thickness on the generation of thermo-emf of In_2Te_3 thin films has been studied and discussed in detail.

2. Materials and methods

2.1. Preparation of In_2Te_3 thin films

In_2Te_3 thin films were grown on glass substrates using a thermal evaporation system (SMART COAT 3.0 Hind High Vac. Co, India). In_2Te_3 pellets (ACI alloys, USA) of 99.999% purity were used as precursor material and the substrate was maintained at different temperatures ranging from 298 to 473 K. To ensure the enhancement of the thermoelectric property, the thin films were grown at higher deposition rate (~11.67 nm/s) rather than lower rates. The thickness of the films was varied in the range of 150–500 nm (estimated from gravimetric method).

2.2. Characterization

The crystalline structure of the prepared films was examined using X-ray diffraction (XRD) patterns (RIGAKU MINIFLUX 600, CuK_α radiation) recorded in the diffraction angle (2θ) range of 20–65°. The surface morphology and the quantitative analysis of the films were studied by field emission scanning electron microscopy (FESEM, CARL ZEISS) and energy dispersive analysis (EDAX, CLASS ONE SYSTEM), respectively. The optical absorption spectra of the films were recorded using UV-IR spectrophotometer (SHIMADZU UV-3600) in the wavelength range of 500–2500 nm. The thickness of the films was estimated from gravimetric method and the error was 5% compared with FESEM image cross section view measurement. The electrical resistivity of the prepared films was measured by two-probe method using Keithley multimeter-2002.

In the present study, a lab designed experimental setup (Fig. 1) has been used to investigate the thermoelectric properties of In_2Te_3 films. The experimental setup consists of two aluminum clamps to hold the sample and one of the clamps was connected to the heater. Here, the sample size (In_2Te_3 thin film) was maintained such that the length-to-width ratio as 6:1 and the ohmic contacts at the both ends of the thin film were made with deposition of silver (thermal evaporation method) [17]. To maintain the temperature gradient in sample, the heater temperature was raised gradually. The temperature indicators were connected to the hot and cold ends of the sample to monitor the temperature gradient maintained. This temperature gradient induces the thermo emf in the sample and it can be explained as follows: at the hot probe, the thermal energy of the majority carrier is higher than at the cold probe, hence carriers will tend to diffuse away from the hot probe driven by the temperature gradient. As they diffuse away from the hot probe, they leave behind the oppositely charged immobile donor atoms, which results in a current flow towards the hot probe and the magnitude of voltage across two ends will give the generated thermo emf values. The thermo emf of the prepared films was measured in the temperature gradient range of 5–170 K. Further, the current flow is towards hot probe for p-type material and away from the hot probe for n-type material, this allows to find the material type [18].

3. Results and discussion

Fig. 2 shows the XRD patterns of In_2Te_3 thin films deposited at different substrate temperatures (298, 373, 423 and 473 K). All films are observed to be polycrystalline in nature. For the as-grown In_2Te_3 films at substrate temperature till 423 K (i.e., 298, 373 and 423 K), four peaks are observed, which correspond to (1 1 0), (5 3 1), (2 1 0) and (7 6 3) planes of cubic phased In_2Te_3 (JCPDS Cord No. 33-1488). With further increase in the substrate temperature to 473 K, the minor contribution of tetragonal phased InTe (JCPDS Cord No. 00-007-0112) is observed in the predominant In_2Te_3 phase. The (5 3 1) plane corresponding to In_2Te_3 phase is diminished, while the (2 1 1) plane associated with the InTe phase is pronounced in the as-grown films. This is reasonable because, according to the indium telluride compound phase diagram, InTe phase is next stable phase

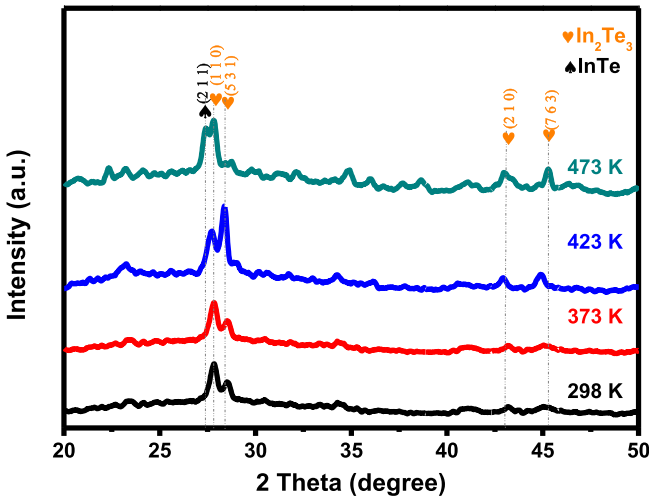

Fig. 2. XRD patterns of Indium telluride thin films grown at different substrate temperatures.

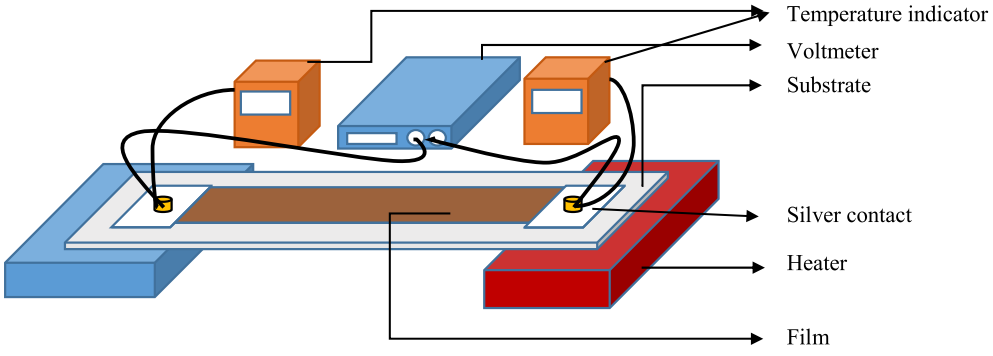

Fig. 1. Schematic diagram of the themo emf measurement set up.