

Composition Dependent Thermoelectric Power (α) of Zn-Te Thin Films as a Function of Temperature and Thickness.

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Abstract:- Thin films of Zn-Te compound of varying thicknesses for different compositions have been deposited on glass substrates in a vacuum of the order of 10^{-5} torr. The composite films used for characterization were annealed at a constant temperature for 6 to 8 hours. Thermo-electric power (α) has been evaluated as a function of thickness, composition and temperature of films. The Zn-Te thin films showed three distinct mechanisms. They show n-type behavior in the temperature range of 273 to 289 °K and p-type behavior in the temperature range of 289 to 433 °K. Thermoelectric power slowly increases with thickness of the Zn-Te films. The maximum thermoelectric power observed nearly at 50 at.wt.% of Zn in Zn-Te thin films can be attributed to defect free stoichiometric phase.

Keywords-- Zn-Te, thin films, thickness, composition, transition temperature, thermoelectric power (α)

I. INTRODUCTION

Thin films of Zn-Te have been made by several workers from the point of view of crystallization [1-3] and application considerations [4-5]. The Zn-Te is a II-VI semiconducting compound of a direct band gap 2.26 eV at room temperature. The polycrystalline compound semiconductor films are of considerable technological importance and play a major role in the fabrication of electronic devices, both from purely scientific and application points of view, it is used in detectors, IR filters, Solar cells, Switching devices, etc. The crystallite size in evaporated films can be improved by employing a higher deposition temperature for the reduction of cracking and pinhole effects [6].

Thermoelectric power of Zn-Te crystals were studied by Davis-Mott model [7]. Many amorphous materials [8-9] when heated to a relatively high temperature undergo an irreversible change in structure. Thermal energy becomes large enough to initiate a very fast crystallization process and amorphous materials switch to a polycrystalline state.

Seebeck coefficient measurements over entire composition range of Zn-Te films are made using differential method [10-14]. The temperature of hot end (T_H) is varied from 273 to 433 °K with constant difference of 2 °K and 10 °K for temperature regions of 273 to 303 °K and 303 to 433 °K respectively.

The TEP (α) is found negative in sign when the temperature of hot end varied from 273 to 289 °K, however it is positive in sign for a temperature range of 289 to 433 °K. The change of sign of TEP (α) from negative to positive is composition dependent. For stoichiometric ZnTe (Zn ~ 50 at. wt. %) films, ' α ' changes its sign negative to positive at 289 °K, and for films with Zn < 50 at wt %, and Zn > 50 at. wt %, the transition temperature changes by $\sim \pm 2$ °K. This fact reveals that Zn-Te deposits are n-type in the temperature range of 273 to 289 °K and p-type in the temperature range of 289 to 433 °K. It seems that in Zn-Te deposits the majority carriers are p-type.

II. EXPERIMENTAL

The work reported here Zn-Te thin films were prepared by the three temperature method [13,14,17,18]. Zn-Te films of different compositions and different thickness were prepared by vacuum deposition of the constituent elements Zn (99.99% pure) and Te (99.99% pure). Zinc and Tellurium powders were evaporated from two different pre-heated conical mica baskets which in turn were heated externally by nichrome wire. The films were prepared mostly on glass substrate kept at room temperature in a vacuum of the order of 10^{-5} torr, after adjusting the flux rates from two sources by varying the source current, films of varying compositions were obtained. Overcoming the experimental difficulties in adjusting and maintaining evaporation rates of the individual components to obtain films of different compositions having nearly same thickness and films of different thicknesses with same composition. The films obtained were annealed at ~ 433 °K for 6 to 8 hours for the purpose of uniform distribution of the components in the deposits. The method employed to determine the composition, thickness and uniformity of the film were similar to those reported earlier [13,14,17-19]. The composition of the film was determined by employing absorption spectroscopy [15] at a wavelength of 350 m μ with an accuracy of ± 1 at.wt.%.

The film thickness (d) was measured by using gravimetric method [13,14,18,19] ($\pm 100\text{\AA}$) using the relation

$$d = M / (g A) \text{ cm} \quad \text{----- (1)}$$

Where A - surface area of the film
M - Mass of the film
g - density of the film material,

expressed as

$$g = x_1 g_1 + x_2 g_2 \quad \text{----- (2)}$$

Where g_1 , g_2 and x_1 , x_2 are densities and atomic fractions of Zn and Te elements respectively.

Electrical measurement such as thermoelectric power of annealed Zn-Te films of various thicknesses and compositions were made at temperature of hot end ranging from 273 to 433 °K in vacuum of 10^{-2} torr using digital multimeter as described before [20].

III. RESULTS AND DISCUSSION

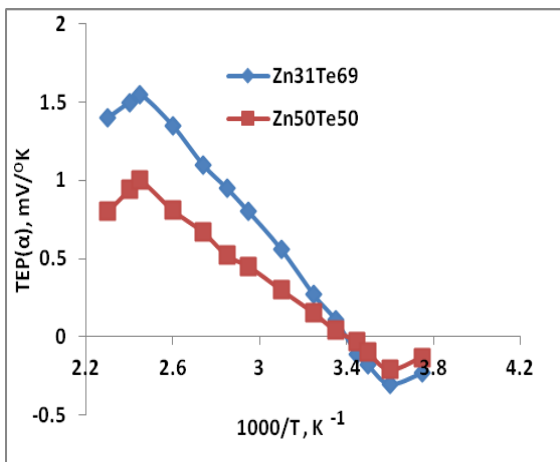


Fig.1: Plot of thermoelectric Power (α) versus $1000/T$ of Zn-Te thin films in the temperature range of 273 to 433 °K.

The thermoelectric Power (α) variation with temperature is expressed by an expression

$$\alpha = \Delta V / \Delta T, \text{ mV} / ^\circ\text{K} \quad \text{----- (3)}$$

Fig.1 shows ' α ' versus $1/T$ will yield straight line with different slopes for Zn-Te thin films in the temperature range of 273 to 433 °K, It is seen that probably in the temperature range of 303 to 433 °K, ' α ' increases with increase of temperature up to 410 °K linearly and after 410 °K, ' α ' slowly decreasing with further increase of temperature.

From the observations of fig.1 it can be said that three distinct mechanisms for conduction in Zn-Te films are possible (1) Extended state conduction, (2) conduction is in the localized state at the band edge and (3) conduction mechanism is the excitation of carriers from the localized states at the Fermi level to the localized states at the valence band edge plus the hopping between these states or excitation of carriers from the Fermi level to the extended states beyond the mobility shoulder. In the present investigation ' α ' does not vary linearly with temperature (fig 1). Hence Zn-Te deposits are not purely metallic or degenerate semiconductor.

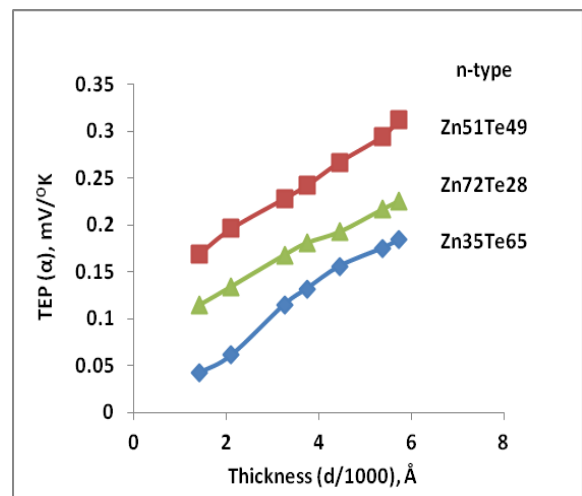


Fig.2: Plot of thermoelectric Power (α) versus thickness (d) of Zn-Te thin films at 281 °K from 273 to 289 °K.

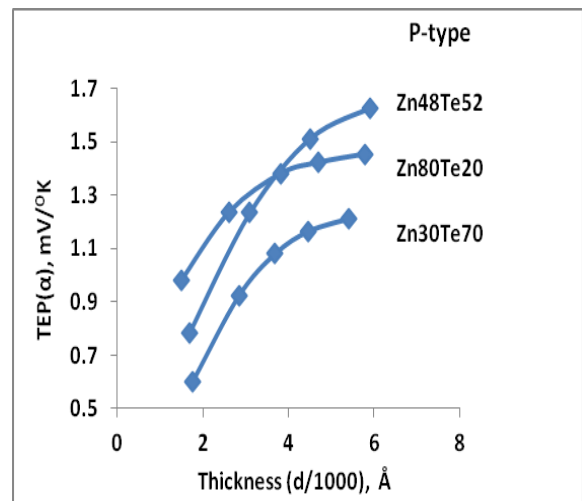


Fig.3: Plot of thermoelectric Power (α) versus thickness (d) of Zn-Te thin films at 333 °K from 290 to 433 °K.

Fig. 2 shows the variation of TEP (α) with thickness (d) of n-type Zn-Te thin films below the temperature 289 ⁰K and fig.3 shows variation of TEP (α) with thickness (d) of p- type Zn-Te films above the temperature 289 ⁰K upto to 433 ⁰K. It is seen that ' α ' of thinnest film is lowest in magnitude and increases rapidly with increase of thickness (d). The rate of increase of TEP (α) with thickness (d) is greatly influenced by the stoichiometry of the film. It has been observed further that for stoichiometric ZnTe films, the rate of increase of ' α ' with 'd' is maximum, probably due to removal of dislocations and grain boundaries with increase of thickness of stoichiometric ZnTe films.

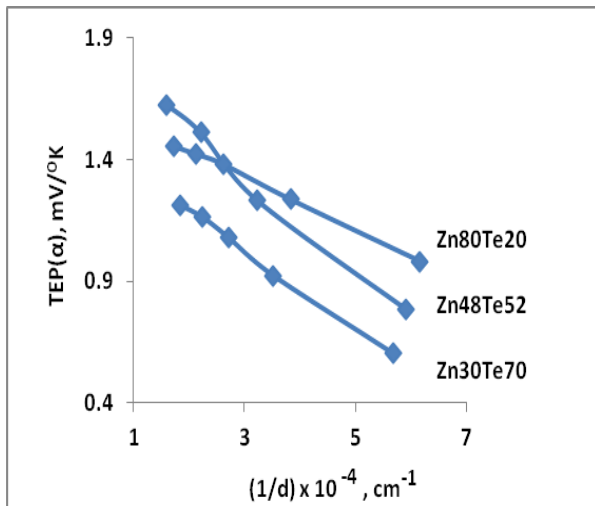


Fig.4: Plot of thermoelectric Power (α) versus (1/d) of Zn-Te thin films at 333 ⁰K from 290 to 433 ⁰K.

Fig. 4 shows plot of α versus 1/d. It is seen that all the curves are almost linear indicating an inverse relationship between ' α ' and thickness (d). This dependence of ' α ' on 'd' indicates dimensional effects in these films. The intercept on y-axis of ' α ' versus '1/d' plots gives the bulk thermoelectric power (α_B) or grain boundary TEP (α_g). (α_B) is TEP of bulk sample with very large grain size, where the grain boundary scattering effects are insignificant. The value of bulk thermoelectric power (α_B) or grain boundary thermoelectric power (α_g) obtained from the intercept of the α_F versus 1/d plots at 333 ⁰K, for the compositions of Zn-Te films at Zn₃₀Te₇₀, Zn₄₈Te₅₂ and Zn₈₀Te₂₀ are 1.45 mV/⁰K, 1.93 mV/⁰K and 1.64 mV/⁰K respectively. From these three values of α_B , it is concluded that the thermoelectric power of bulk state stoichiometric ZnTe films is highest than that of other compositions.

In free electron approximation and for a spherical Fermi surface, the thermoelectric power of metal and/ or degenerate semiconductor is given by [20]

$$\alpha_B = [(\pi^2 K_B^2 T) / (3e E_F)] (U+V) \text{ ----- (4)}$$

Where

α_B - bulk thermoelectric power, $U = [(d \ln \lambda_B / d \ln E)_{E=E_F}]$ the rate of change of mean free path with the energy evaluated at the Fermi energy, $V = (d \ln A / d \ln E)_{E=E_F}$ (the rate of variation of the Fermi-surface area with the energy evaluated at Fermi energy), e - charge of carrier, K_B - Boltzmann constant and T-temperature in ⁰K.

IV. CONCLUSIONS

Thin films of Zn-Te compound of varying thicknesses and compositions deposited on glass substrates in a vacuum show three distinct mechanisms. In the present investigation ' α ' does not vary linearly with temperature hence Zn-Te deposits are not purely metallic or degenerate semiconductor they shows n-type behavior in the temperature range of 273 to 289 ⁰K and p-type behavior in the temperature range of 289 to 433 ⁰K. Thermoelectric power slowly increases with thickness of the Zn-Te films, the maximum thermoelectric power observed nearly at 50 at.wt.% . It is seen that ' α ' of thinnest film is lowest in magnitude and increases rapidly with increase of thickness (d). The rate of increase of TEP (α) with thickness (d) is greatly influenced by the stoichiometry of the film. It has been further observed that, for stoichiometric ZnTe films the rate of increase of ' α ' with 'd' is maximum, probably due to removal of dislocations and grain boundaries with increase of thickness.

The value of bulk thermoelectric power (α_B) or grain boundary thermoelectric power (α_g) obtained from the intercept of the α_F versus 1/d plots at 333 ⁰K, for the compositions of Zn-Te films at Zn₃₀Te₇₀, Zn₄₈Te₅₂ and Zn₈₀Te₂₀ are 1.45 mV/⁰K, 1.93 mV/⁰K and 1.64 mV/⁰K respectively. From these three values of α_B , it is concluded that the thermoelectric power of bulk state stoichiometric ZnTe films is highest than that of other compositions.

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