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Thickness Dependent Thermoelectric Power (α) of Ag-Te Thin Films

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ABSTRACT

Thin films of Ag-Te compound of varying thicknesses at fixed composition have been formed on glass substrates in a vacuum of the order of 10-5 torr. The films were annealed at constant temperature for 6 to 8 h and then thermoelectric power (α) of annealed films has been evaluated. The thermoelectric power shows composition dependent p-type and n-type behavior. The measurements were carried out with constant temperature difference of 2 K and 10 K employing three temperature methods for low and high temperature regions respectively. Thermoelectric power (α) for both p-type and n-type materials increase with film thickness.

Keywords: Ag-Te, Glass substrate, Thin films, Thickness, Thermoelectric power

INTRODUCTION

Studies on the I-VI semiconducting compounds have received much attention because of their potential applications in semiconductor technology. The compound Ag_2Te is a narrow band gap semiconductor with high electron mobility and low lattice thermal conductivity, It exists in two modifications; a low temperature monoclinic modification and at high temperature cubic modification [1]. The Das and Karunakaran [2] reported electrical conductivity and thermoelectric power measurements as a function of temperature have been carried out on Ag_2Te thin films of different thicknesses on glass substrates at room temperature in a vacuum. It is found that the phase transition temperatures located by steep change in resistance and thermoelectric power with temperature during heating and cooling are different, there by showing a thermal hysteresis during the phase transition, the magnitude of hysteresis as a function of thickness.

The level of interest in thermo magnetic and magneto thermoelectric transducers has increased greatly in recent years. The reason for this trend is that in certain cases, these devices have advantages over thermoelectric transducers, in particular solid state cooling devices and IR radiation pickups [3,4].

The electrical and structural properties of silver telluride films have been investigated by several workers in stoichiometric phase, as a function of temperature. However these compounds are less investigated in the form of thin films of different composition and thicknesses. From this point of view and considering application in electronic devices the effect of thickness and composition of Ag-Te films are discussed. We report the measurement of thickness dependent thermoelectric power for various compositions of Ag-Te thin films.

MATERIALS AND METHODS

Preparation of tin thin films

Thin films of Ag-Te for the measurement of thermoelectric power were prepared by three temperature method [5-11] for various thicknesses. The constituent elements of Ag (99.999% pure) and Te (99.99% pure) used for thin films preparation in metal and powder form respectively. They were evaporated from two different preheated conical mica baskets which in turn heated externally by nichrome wire. The films were prepared on glass substrate kept at room temperature in a vacuum of the order of 10^{-5} torr with an IBP TORR-120 vacuum unit. After adjusting the flux rates from two sources by varying the source current, films of varying thicknesses were obtained by overcoming the

experimental difficulties in adjusting and maintaining evaporation rates of the individual components. The films obtained were annealed at ~423 K for 6 to 8 h for the purpose of uniform distribution of the components of the deposits. The method employed to determine the composition of the film were similar to those reported earlier. The composition of Ag from Ag-Te films was determined by employing absorption spectroscopy at 350 nm [12].

Measurement of thickness

The film thickness (d) of Ag-Te thin films was measured by multiple beam interferrometry [13] and gravimetric method [5-11] using the relation,

$$d = \frac{M}{g \times A} cm \quad (1)$$

Where

A=surface area of the film

M=Mass of the film

g=the density of the film material = $x_1 g_1 + x_2 g_2$

where g_1, g_2 and x_1, x_2 are densities and atomic fractions of Ag and Te elements, respectively.

Thermoelectric properties

The thermoelectric power (α) was measured by integral method [9,14]. In the integral method, one end of the sample was heated while other end kept at constant temperature. The temperature difference (ΔT) between two ends of the film causes the generation of thermo emf.

Thermoelectric power was calculated from the relation:

$\alpha = \Delta V/\Delta T, \mu V/K$ (2)

To eliminate the effect of adsorbed moisture, the measurements were made at reduced pressure of ~10^{-2} torr keeping the film inside a glass vacuum tube connected to rotary pump.

RESULTS AND DISCUSSION

Thermoelectric power (α) studies were made on Ag-Te deposits of different compositions and thicknesses in the temperature range of 273 to 433 K, with constant temperature difference of 2 K and 10 K, in low temperature region (273 to 303 K) and high temperature region (303 to 433 K) respectively (Figures 1 and 2).

Figures 1 and 2 show plots of p-type and n-type thermoelectric power (α) verses thickness (d) of Ag-Te thin films at 323 K from 273 to 433 K, respectively. To analyze dimensional effects, the TEP (α) is plotted as a function of film thickness at two different compositions (p-type and n-type), it is seen that TEP (α) of thinnest film is the lowest in magnitude in both cases, and it increases rapidly with increase of thickness (Figures 3 and 4).

Figures 3 and 4 shows the plots of TEP (α) against reciprocal thickness (1/d) for p-type as well as n-type deposits. It is seen that both the curves from certain extent are linear, indicating an inverse relationship between 'α' and 'd'. The intercept on y-axis gives bulk TEP (α_B) or grain boundary TEP (αg). The TEP of bulk sample with very large grain size, where the grain boundary scattering effects are insignificant, while 'αg' is defined as the TEP of bulk sample having the same grain size as the films, so that the grain boundary scattering effects are insignificant.

It is well known that the transport properties of thin films may be quite different from those of the bulk material, particularly if the film thickness is small. This is because, as the thickness of the film becomes comparable in magnitude with mean free path. Physical effects which appear as a consequence of the reduction of the mean free path due to small dimensions are referred to as classical size effects.

According to classical size effect theory for a free electron model was worked out by Fuchs [15] for a spherical Fermi surface and later Sondheimer [16] extended this to include galvanomagnetic effects. Their treatment is a statistical analysis based on the Boltzmann equation for the distribution function of conduction electrons. One of the basic assumptions made in their theory was that the electros are scattered from the film surfaces either diffusely

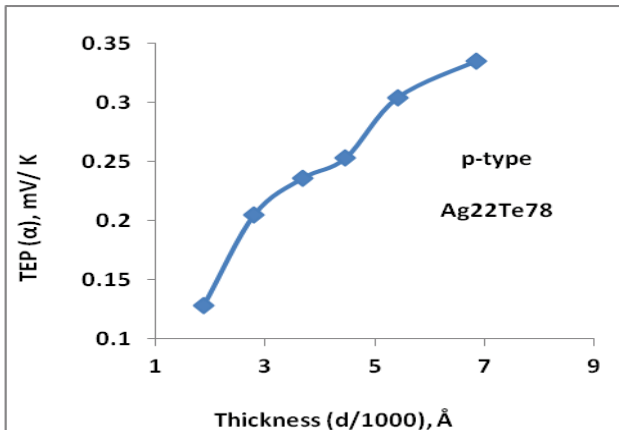

Figure 1: Plot of p-type thermoelectric power (α) verses thickness (d) of Ag-Te thin films at 323 K from 273 to 433 K

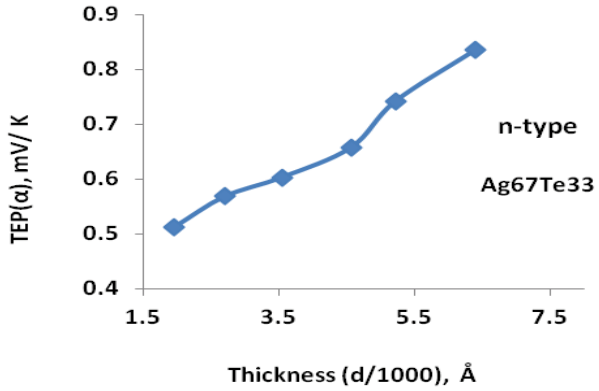

Figure 2: Plot of n-type thermoelectric Power (α) verses thickness (d) of Ag-Te thin films at 323 K from 273 to 433 K

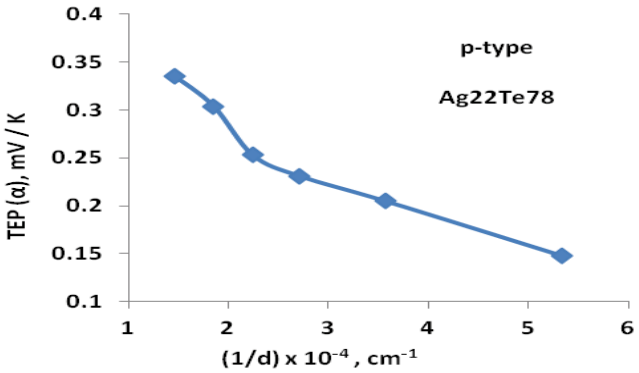

Figure 3: Plot of p-type thermoelectric power (α) verses (1/d) of Ag-Te thin films at 323 K from 273 to 433 K

or specularly, which are the two extreme possibilities to consider. For a sufficiently rough surface the former would apply and for a mathematically plane surface the later would be more likely. In the former case, every free path of the carriers is terminated by collisions at the surface, so that the distribution function of the carriers leaving, such surface is independent of direction. In the latter case, the carriers (electrons) are elastically scattered from both the surfaces of the film with the reversal of the velocity component normal to the film surface. It was also assumed that a portion 'p' of the incident electrons was specularly scattered and the reminder diffusely scattered. The relaxation process for surface scattering was essentially taken for bulk scattering.

According to the classical size effect theory mentioned above, the thermoelectric power 'α_F' of a thin film of thickness 'd' was given by [17].

$$\alpha_F=\alpha_B\{1-(3/8)(1-p)[U/(1+U)]\ (\lambda_B/d)\} \quad (3)$$

Where 'p' is the specularity parameter, giving the fraction of electrons specularly scattered from the surfaces.

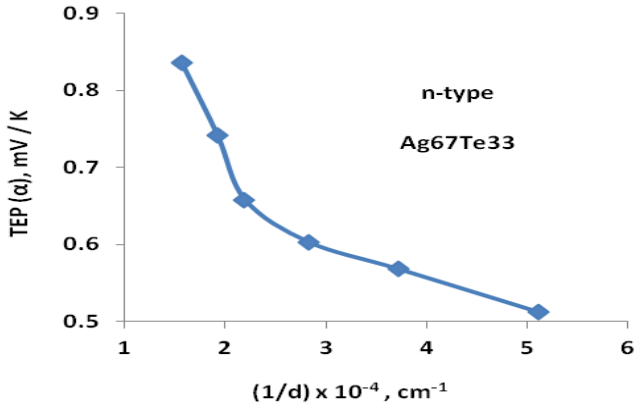

Figure 4: Plot of n-type thermoelectric power (α) verses (1/d) of Ag-Te thin films at 323 K from 273 to 433 K

The above expression is valid for mono crystalline films, as it does not taken into account for the contribution to thermoelectric power due to grain boundary scattering. Now taking into account the effect of grain boundary scattering along with background scattering Pichard et al. [18] have derived an expression for thermoelectric power of polycrystalline thin films as

$$\alpha_F=[\pi^2 K_B^2 T/3e E_F][V+U(\sigma g/\sigma_B)-(3/8)(1-p) (\lambda_B/d) U(\sigma g/\sigma_B)^2] \quad (4)$$

Where σg and σ_B are the grain boundary conductivity and bulk conductivity, respectively and 'e' is the charge of carriers.

It is seen from the above equations that 'α_F' is a linear function of reciprocal thickness (1/d), if 'p' is assumed to be constant in both cases .Hence a plot of 'α_F' verses 1/d will be straight in either case. However, the intercept on the y-axis gives the bulk thermoelectric power 'α_B' or the grain boundary thermoelectric power 'α_g' which is the thermoelectric power of a bulk specimen having the same micro structure as the films, according to the above equations (equations 3 and 4), respectively.

Further the slopes of 'α_F' versus 1/d contain different parameters according to the above two equations (additional parameters according to equation 4). Thus the nature of the two equations is the same, except for the difference in the constant parameters involved in functional relationship.

CONCLUSION

The value of 'α_B' or 'α_g' obtained from the intercept of α versus 1/d plot is ~-814 μV/K at 333 K of $Ag_{67}Te_{33}$ n-type thin films. Similarly for p-type Ag-Te semiconducting films, ([Ag]<54 at. wt. %), α_B of $Ag_{22}Te_{78}$ films is 332 μV/K. The values reported for bulk samples of β-Ag_2Te is about -120 μV/K and for thin films of Ag_2Te is -60 μV/K.

The large difference between the thermoelectric power values obtained by us in the case of thin films on the bulk polycrystalline samples can be because of the following two reasons. Firstly , thermoelectric power as with any other transport property exhibits anisotropy as the crystal structure of β-Ag_2Te is monoclinic and/or orthorhombic and other the thermoelectric power measured in different crystallographic directions will be different.

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