Electrical and thermal transport properties of binary chalcogenide indium polytelluride crystals

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Abstract: In the present work, single crystals of In_2Te_5 were grown by the modified Bridge man technique. An investigation has been carried out on the influence of temperature on the electrical conductivity, Hall effect and thermoelectric power. The energy gap calculated to be 0.88 ev, the ionization energy of acceptor was determined to be 0.14 ev. The conductivity throughout the entire temperature range was found to be of P-type. The electrical conductivity, Hall coefficient, carrier concentration ration at room temperature were estimated to be 1.47 $x10^{-2}$ (Ω^{-1} cm^{-1}), 4.6 $x10^4$ cm^3/C and 1.3 $x10^{14}$ cm^{-5} respectively, the electron and hole mobility are found to be 8.53 $x10^3$ cm^2/v.sec and 6.78$x10^3$ cm^2/v.sec respectively. The effective masses of charge carriers are 1.59 $x10^{-39}$ kg and 2.42 $x10^{-38}$ Kg for electrons and holes respectively. The diffusion coefficient for both majority and minority carriers was estimated to be 177.6 cm^2/sec and 221.3 cm^2/sec respectively. The diffusion length as well as the relaxation times of holes and electrons are found to be Lp= 4.29$x10^{-7}$ cm, L_n= 1.368$x10^{-7}$ cm, τ_p = 1.63 x 10^{-15} sec and τ_n = 8.4 $x10^{-17}$ sec respectively. In addition to these pronounced parameters, the efficiency of the thermoelectric element (figure of merit) was checked, which leads to better application in many fields keywords; crystal growth, In_2Te_5, electrical conductivity, Hall effect, thermoelectric power.

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1. Introduction

Research on binary semiconducting compound formed by the elements from groups (III) and (VI) of the periodic table as a collective group of materials have been and are still the subject of much intensive investigation. In the last few years widespread attention has been paid to the semiconductors of the ($A^{III}B^{VI}$) group. This interest has been driven by their possible device application.(1) Great attention has been paid(2) to Ga, In and Te chalcogenides. In particular the study of the $A_2^{III}B_3^{VI}, A_2^{III}B_5^{VI}$ and $A_4^{III}B_3^{VI}$ compounds is quite attractive during the last few decades(3).

Indium tellurides are useful as materials for electronics and for optical recording. The phase diagram of the In Te system studied by many authors(4-7), and according to the last study it is obvious that four compounds in the In-Te system, InTe, In_2Te_3, $IaTe_5$ and In_4T_5 are stable. In_2Te_5 is a member of this family, has interesting properties. Among $A^{III}B^{VI}$ compounds not much attention has been paid to investigations of indium telluride, there are some works devoted to InTe and In_2Te_3, (8-19), but only very few works devoted to structure and some physical properties of In_2Te_5(20-25). The search for new semiconducting materials has led us to the present investigation on In_2Te_5, As far as we know from the published literature up to now, there is still insufficient data to throw a clear light upon the actual behavior of this compound, also some of the results given in the literatures show some discrepancies. In view of this, the present work aims to prepare In_2Te_5 in single crystal form and investigate the main transport properties of this compound. This study is a timely one in view of the recent interest in this compound.

2- Experimental

2.1. Preparation of sample

In_2Te_5 samples were prepared using a modified Bridgman technique for growing crystal from melt. Indium polytelluride monocrystals was prepared from high purity indium (6N) representing 26.4672% and tellurium (5N) representing 73.5328%, stoichiometeric of the elements was used as starting material in the growth experiments. At the beginning of the growth run, the ampoule with its charge was held in the hot zone of the furnace at 783k for 10 h for melt homogenization the charge was shaken during heating several times to accelerate the diffusion of contaminates through each other. Then the ampoule was moved into the middle zone of the furnace with a temperature of 740k corresponding to the crystallization temperature according to the phase diagram(4), Afterwards, the ampoule was cooled down slowly in the third zone of the furnace, then the furnace was switched. Details of the experimental equipment for crystal growth and preparation procedures are described elsewhere[27]. The resulting ingots had a plate-like habit with metallic bright color. The producing ingot showed good agreement with the obtained data reported early [21]. The single crystallinity of the compound was checked using X-ray diffraction technique. From the X-ray studies it was evident that the crystal has a high degree of crystallinity with the required phase without any secondary phase.

2.2. Experimental arrangement

Measurements of the electrical conductivity and Hall effect were done with the help of a Pyrex glass cryostat, which was designed[28] for this purpose. The cryostat is used as a holder evacuated container for liquid nitrogen (for low-temperature measurement) and support to the electric heater (for high-temperature measurements. Copper-constantan thermocouple was used for measuring the temperature of the sample. Silver paste was used for the ohmic contact. Typical dimension for rectangular sample were (8.5×2.7×1) mm^3. The Hall measurements were made in a magnetic field of 0.5 tesla and were performed using the conventional DC potentiometer method. For thermoelectric power (TEP) measurements, an evacuated working chamber was used to protect the sample from oxidation and water vapor condensation at high and low temperature respectively. The outer heater discharge its heat slowly to the specimen environment. The inner heater was attached to lower end of the crystal in order to control the temperature and its gradient along the specimen. More details about the apparatus and technique of measuring have been published.[29,30]

3- Results and discussion:

3.1.Temperature dependence of electrical conductivity and Hall effect for In_2Te_5

Fig.1 shows the variation of electrical conductivity σ versus inverse temperature for In_2Te_5 single crystal. The electrical conductivity and Hall coefficient measurements were performed in the temperature rang extend from 198 up to 558K. The complete temperature range can be subdivided into three regions, below the transition, the transition region and above the transition. These regions are quite clearly shown in fig.1. With increase of temperature the electrical conductivity at first increases gradually, then it reaches the transition region at 373k, then the lnσ vs 1/ T curve passed through an intermediate region (373-473k) in which the carrier concentration is not actually constant, and in the third region, σ rises rapidly. This pattern of changes in the electrical conductivity is due to the appeance of impurity and intrinsic conductivity, respectively, and to the variation of the carrier mobility and concentration with temperature. In the intermediate region where the carrier density (N_A-N_D) = constant, until the intrinsic region is reached. The decrease of the value of σ in this region may be due to the increase of intensity of lattice vibration which leads to decrease in the carrier mobility. This discussion is acceptable, since the conductivity decrease in this region and this extended to full ionization of impurity at the end of impurity at the end of exhaustion region. At temperatures above the transition point the conductivity increases rapidly (473-558K). The temperature dependence exhibits a transition from a region of lower slope to one of higher slope. The slopes of the curve increase with increasing temperature, and are higher at higher temperature because of the carriers being excited from the extended state of the valance band into the conduction band. The width of the forbidden gap as calculated from the slope of the curve in the high- temperature region is found to be ΔE_g =0.88 ev. Also the ionization energy as deduced in the impurity region was evaluated to be ΔE_a = 0.14 ev.

The room temperature conductivity of In_2Te_5 single crystal equal to $1.47x10^2$ $(\Omega\ cm)^{-1}$. The Hall coefficient (R_H) variation with temperature and a positive sign of (R_H) indicated the major contribution to the conductivity by holes.

Fig.2. Shows the temperature dependence of ($R_H T^{3/2}$), it is the usual type for semiconductors.

Assessment of the forbidden band width from this graph and found have a value close to that determined from electrical conductivity and that reported early.[26]. It was found also the depth of the acceptor centre is 0.14 ev. The variation of the Hall mobility with temperature is shown in fig 3.

It was found that the exponent(n) in the relation $\mu \sim T^n$ below 373k is equal to 1.112, while in the high temperature range (T>373) the mobility decreases according to the low $\mu \sim T^{-1.92}$ from this relation, it seems that the value of n close to impurity and lattice scattering mechanism, in the low and high temperature respectively. The room temperature mobility was found to be 6.866 $x10^3$ cm^2/V.sec. The variation of carrier concentration with temperature is shown in fig 4.

As the, In_2Te_5 sample exhibiting intrinsic behavior above 473K the expected value for the intrinsic concentration will be given as

$$n_i = 2\left(\frac{2\pi k}{h^2}\right)^{3/2}\left(m_n^* m_p^*\right)^{3/4} T^{3/2} \exp\left(\frac{-\Delta Eg}{2kT}\right)$$

One can see that the carrier concentration varies sharply with increasing temperature. The room temperature concentration is $1.3x10^{14}$ cm^{-3}

3.2. Temperature dependence of TEP for In_2Te_5 single crystal.

The thermoelectric power (TEP) measurements were performed in a wide, temperature range (153-450k).

Fig5 illustrates the general mode of variation of TEP with temperature. This was done by plotting the relation between α and ln T in the low temperature range. Fig (5) shows a straight line relation in this region of temperature.

In the impurity region the following formula could be applied[31].

$$\alpha = \frac{K}{e}\left[2 - \frac{\ln Ph^3}{2(2\pi m_p^* KT)^{3/2}}\right]$$

Thus the effective mass of holes is evaluated to be2.42x10^{-38} kg.

Some features of these results may be pointed out

(1) Our sample shows P-type conductivity within the temperature range of investigation, which is in quantitative agreement our previous data of the Hall coefficient and another published data[26].

(2) The room temperature thermoelectric power value for In_2Te_5 mounted to be 7.47 μv/K.

(3) The figure shows that the value of α decreases as the temperature rises. This may be due to the presence of some crystal defects or trapping centers in the direction of the carrier flow.

(4) With further rise of temperature α decreases i.e in the whole extrinsic range of temperature α decreases with T.

As follows from the expression for TEP of a semiconductor in the intrinsic region [32] .

$$\alpha = \frac{-k}{e}\left[\frac{b-1}{b+1}\left(\frac{\Delta Eg}{2KT}+2\right)\right]+\frac{1}{2}\ln\left[\frac{m_n^*}{m_p^*}\right]^{3/2}$$ Where

k is the Boltzmann constant, b is the ratio of the electron to hole motilities, ΔE_g is the gap width and (m_n^*, m_p^*) are the effective masses of electrons and holes respectively.

Fig 6 shows the relation between the thermoelectric power and the inverse of temperature. This relation show that a plot of α in the intrinsic range, as a function of reciprocal of absolute temperature is a straight line.

The measured thermoelectric power in conjunction with the previously obtained Hall effect data are used to calculate electron to hole mobility ratio and also the ratio of effective masses of both electrons and holes. The slopes of the curve are used to estimate the ratio of the electron and hole mobilities. Taking ΔE_g =0.88 ev, the ratio $b=\mu_n/\mu_p$ is found to be 1.245, since μ_p =6.86x10^3 cm^2/v.sec, then we can evaluate μ_n =8.53x10^3 cm^2/v.sec.

Another important parameter can be deduced with the aid of the obtained values of μ_n and μ_p using Einstein relation, that is the diffusion coefficient for both majority and minority carriers at room temperature can be evaluated to be 177.6 and 221.3 cm^2/sec respectively.

The ratio between the effective masses of both electrons and holes can be estimated from the intersection of the curve. We evaluate this ratio as $(m_n^*/m_p^* = 0.065)$

Combining the value of affective mass of holes with that obtained for the ratio m_n^*/m_p^* , one obtains an effective mass of minority carriers of the value m_n^* = 1.57x10^{-39} kg. By using the effective mass values of electrons and holes, the relaxation time for both current carriers can be determined. Its value for holes comes to be 1.038 x10^{-15} sec, while for electrons it is equal to 8.4x10^{17} sec. Using the values of diffusion coefficient and relaxation time, the diffusion length for both charge carriers can be determined. The values of the diffusion length for electrons and holes are found to be L_n=1.36x10^{-7} cm and L_p= 4.29x10^{-7} cm, respectively. Our results are in good agreement with each other, since the mobility 0f holes is smaller compared with that of electrons,and its effective mass is larger than that of electrons. Its relaxation time will be larger than that of electrons. Fig.7 represent the dependence of α on carrier concentration for a given In_2Te_5 sample, as we have seen α increases sharply and linearly with the decrease of carrier density.

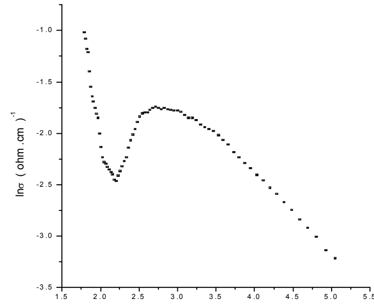

Fig.1:The temperature dependence of electrical conductivity for In_2Te_5 single crystal.

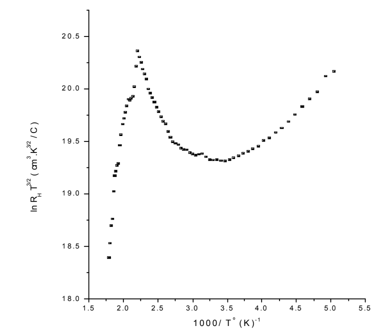

Fig.2:The dependence of ($R_HT^{3/2}$) against the temperature for In_2Te_5 single crystal.

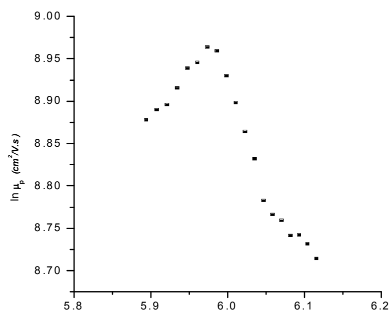

Fig3: behavior of the Hall mobility with temperature.

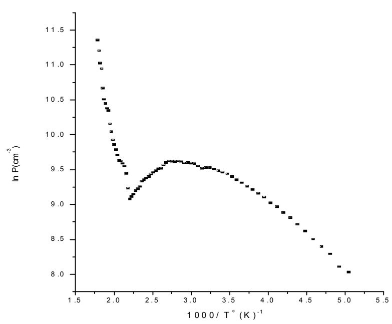

Fig4: Variation of carrier concentration with temperature

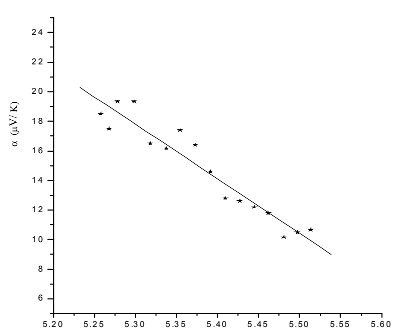

Fig5: Relation between TEP for In_2Te_5 and ln T.

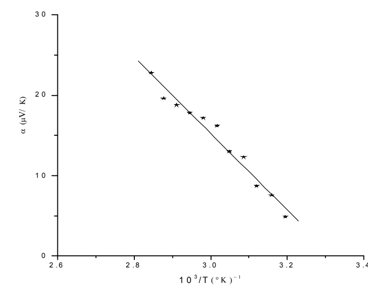

Fig 6: Plot of α against 10^3 /T for In_2Te_5 crystal.

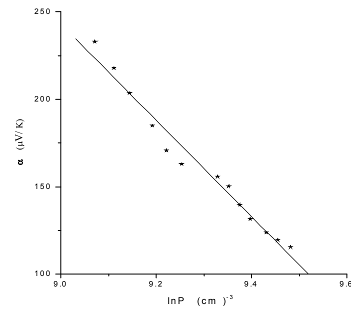

Fig.7 Represent the dependence of TEP on carrier density

From this behavior we realize the effect of the charge carriers is a strong factor governing the variation of α.The same behavior was observed when we plotted α vs Ln σ for In_2Te_5 sample as in Fig.8

This figure shows the dependence of thermoelectric power coefficient α on the natural logarithm of electrical conductivity according to the published formula[33]

$$\alpha = \frac{K}{e}\left[A + \frac{lin2\left(2\Pi m_p^* KT\right)^{3/2} eM}{\left(2\Pi h^2\right)}\right] - \frac{K}{e} lin\sigma$$

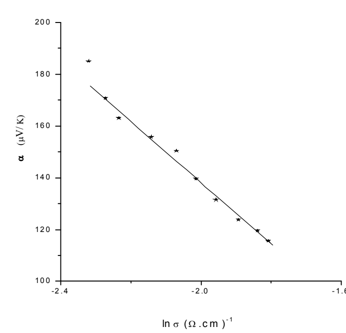

Fig. 8 The dependence of TEP on the natural logarithm of σ for In_2Te_5

It is seen from the curve that the thermoelectric power decrease rapidly as the electrical conductivity increased. From figures 7 and 8, we can deduced that the variation of α with the environmental temperature is not a mobility effect, but is dependent on the variation of concentration. The choice of material for thermoelectric generators and refrigerators is based on the efficiency parameter Z defined by the relation;

$$Z = \frac{\alpha^2 \sigma}{K}$$

where K is the thermal conductivity of semiconductor and σ is the electrical conductivity. K value of In_2Te_5 was determined previously [(34)]. This parameter was deduced and its value was found to be $6.2\times10^{-10}\ K^{-1}$.

The proposed treatment of the experimental data sheds newlight on the main physical parameters in In_2Te_5 single crystal.However those pronounced parameters are found to be sufficient to give complete information about the physical behavior of our best compound. This gives the chance of practical application in different fields.

4-Conclusion

Measurement of electrical conductivity, Hall coefficient and TEP of as grown In_2Te_5 single crystals are performed over wide range of temperature. The conductivity was found to be P-type. The energy gap was calculated to be 0.88 eV, while the depth of the acceptor level is 0.14 eV. The experimental data gives us the chance to determine the following pronounced parameters, carrier mobility, effective masses, diffusive coefficient, diffusive length as well as the relaxation time of both types of charge carriers.Also the efficiency of the material as thermo-element was checked. This mode of investigation (crystal growth, electrical conductivity, Hall effect and thermoelectric properties study) is an ideal was for finding out the possibility of making application for this semiconductor compound especially in the field of energy conversion, semiconductor devices and electronic engineering.

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