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Thermoelectric Power and Structure of Amorphous $Ge_{100-X}sb_x$ Alloys

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Abstract: *Bulk samples of the system $Ge_{100-x}Sb_x$, where x = 5, 50 and 95 at. % were prepared by the melt quenching technique. The disorder-order transition has been investigated for the alloys annealed at 323, 348, 373 and 393 K for different times. X-ray and thermoelectric power were recorded for the annealed samples and the results revealed crystalline Ge, Sb and Ge-Sb phases. It was found that the values of the thermoelectric power decreased as a result of raising the annealing temperature while increasing the Sb content leads to a an increase in the thermoelectric power.*
Keywords: *Thermoelectric power, structure, amorphous alloys and x-ray.*

I. Introduction

In some materials the predictable long range order characteristic of crystalline solids breaks down. Such materials are the non-crystalline (amorphous) or glasses. When such bulk materials are cooled from the melt, even at low rates, the more random atomic positions that associated with a liquid are frozen in place within the solid. Amorphous semiconductors have excited wide-spread interest when the switching devices based on amorphous chalcogenide were fabricated [1]. Chalcogenide glasses prepared by melt quenching, in general, are p-type semiconductor [2]. Different techniques have been used to study the structure of amorphous glasses, e.g. electron microscopy, X-ray diffraction and scanning calorimetry [3-5]. Amorphous semiconductors are suitable for studying the nature of randomness by comparison with crystals of the same material. Bulk glassy samples are chemically ordered and comprise well- defined structure units. The aim of this work is to study the annealing time dependence of the thermoelectric power, the effect of antimony content on the thermoelectric power and structure of bulk samples of amorphous $Ge_{100-x}Sb_x$.

II. Experimental

Bulk amorphous alloys of Ge_{100-x} Sb_x (x = 5, 50 and 95 at. %) were prepared by the melt quenching technique. The Proportions of 99.999 % purity elements in appropriate atomic percentage were weighted using an electronic balance. The weighed elements were mixed together in silica ampoules which were sealed under vacuum at 10^{-4} mbar. Each ampoule was heated in a furnace up to 1100 K for about 10 hours. The ampoules were frequently agitated to intermix the constituents and to increase the homogeneity of the melt. These ampoules containing molten samples were then quenched rapidly in ice-cooled water and then the samples were crushed into fine powder. Cylindrical disks of thickness 2.5 mm and diameter of 13 mm were pressed by die using a pressure machine at a load of 5 tons. Using X-ray diffraction, the bulk samples were found to be in amorphous state. The thermoelectric power (S) was measured for the prepared disks of systems Ge_{100-x} Sb_x in the temperature range 323-393 K. Each specimen was tightly inserted between two copper rods holder placed in an electric furnace. The temperature gradient ΔT established across the specimen is measured by two similar copper-constantan thermocouples. The Seebeck voltage ΔV was picked up by a Digital–Multimeter model Peak Tech® 2010. The thermoelectric power (S) was calculated according to the equation:

$$S = \frac{\Delta V}{\Delta T} \quad \ldots\ldots (4.1)$$

Where ΔV is the thermo electromotive force (e. m. f) (in m v) developed across the sample due a temperature difference ΔT. The change in voltage was recorded using a digital multi-meter.

III. Results And Discussion

Figs. 1, 2 and 3 show the behavior of the thermoelectric power (S) as a function of the annealing time at different temperatures for the glassy system $Ge_{100-x}Sb_x$, (where x = 5, 50 and 95 at. %). It is seen from these figures that all compositions at all selected temperatures show approximately the same trend with increasing the annealing time. The curves illustrate that the thermoelectric power increases until a certain point with increasing the annealing time which is due to amorphous –crystalline transformation, then it remains approximately constant for longer annealing times due to the stopping of nucleation and growth processes.

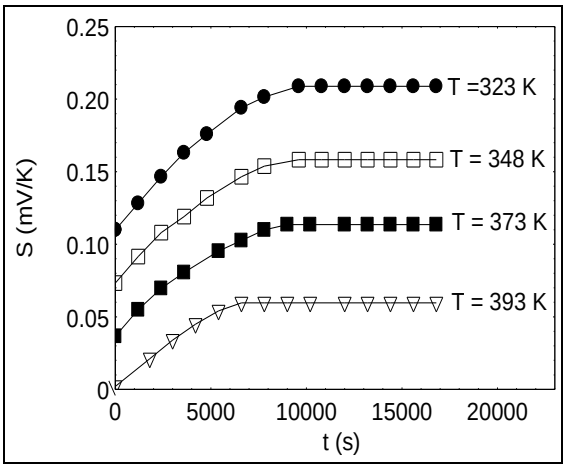

Fig. 1. Thermoelectric power versus annealing time at different temperatures for $Ge_{95}Sb_5$ alloy.

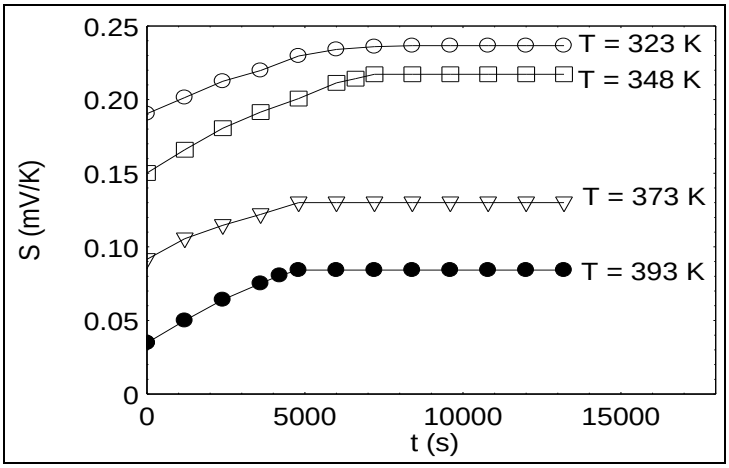

Fig. 2. Thermoelectric power versus annealing time at different temperatures for $Ge_{50}Sb_{50}$ alloy.

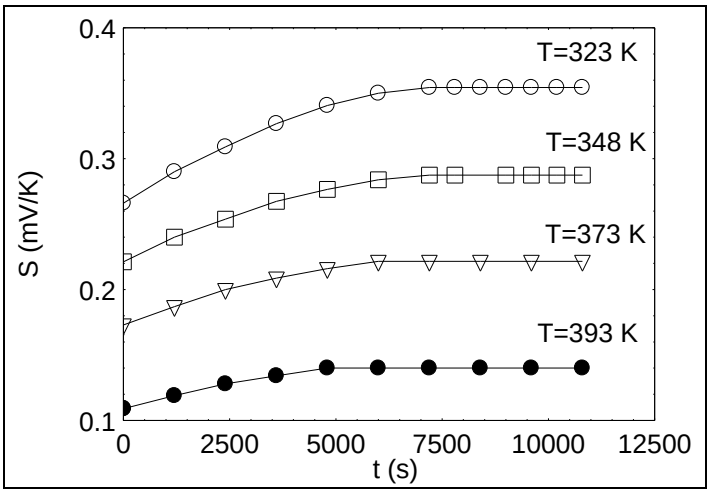

Fig. 3. Thermoelectric power versus annealing time at different temperatures for Ge_5Sb_{95} glassy sample.

In semiconductors, a charged particle must first be excited across an energy gap E_G. In this case the thermo-power is approximated by:

$$S \approx \frac{C_{el}}{Q} \approx \left(\frac{k_B}{e}\right)\frac{E_G}{k_B T} \quad \text{..... (1)}$$

Where k_B is the Boltzmann constant, (e) electron charge and E_G is the energy gap. Since the energy gap for ordered semiconductors is higher than that of amorphous one, the increase of thermoelectric power (S) of the

investigated amorphous alloys with the annealing time is due to the crystallization effects according to equation (1). Fig.4 shows the temperature dependence of thermoelectric power in the temperature range 323-393 K for the glassy system Ge_{100-x} Sb_x, (where x = 5, 50 and 95 at. %). It is seen from this Figure that all compositions show approximately the same behaviour with rising the temperature where the thermoelectric power are observed to decrease with temperature. The results reveal that the material have a positive thermoelectric power for the studied compositions overall the temperature range 323-393 K. In typical p-type thermoelectric materials, positive charge carriers (holes) accumulate at the cold end, causing a positive potential [2, 6].

In general, the behaviour of the thermoelectric power as a function of temperature (Fig.4) can be explained as follows: It has been reported that the thermoelectric power of semiconductors is larger than the characteristic value $87\mu VK^{-1}$ and increases with decreasing temperature [7]. In a semiconductors S is typically a few millivolts per degree Kelvin [8]. It is seen from Fig. 4 that the thermoelectric power of Ge_{100-x} Sb_x alloys decreases with increasing the temperature and have positive values larger than the value $87\mu VK^{-1}$ in agreement with the above statements [7, 8].

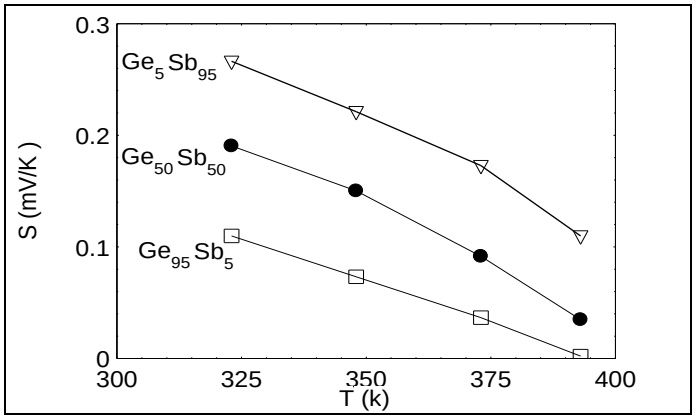

Fig. 4: Temperature dependence of thermoelectric power of Ge_{100-x} Sb_x glassy system.

In order to show the variation of thermoelectric power (S) with the Sb content, the results have been plotted in Fig.5. It is shown that the thermoelectric power (S) increases with increasing the Sb content. This increase may be due to increase of ionicity which leads to the increase of the optical energy gap according to equation (1).

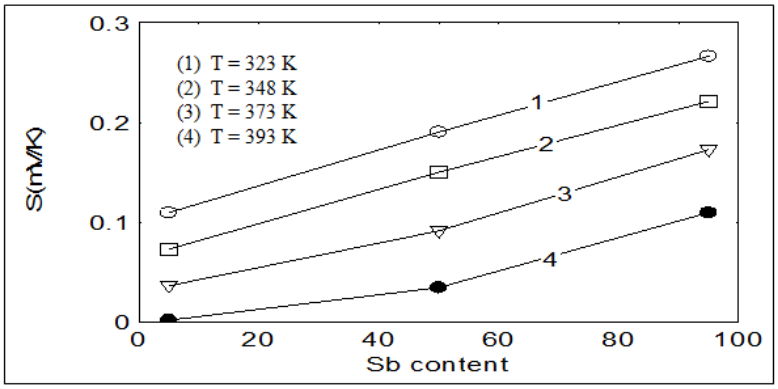

Fig. 5. Variation of thermoelectric power (S) with Sb content at different temperatures.

The effect of annealing on the structure of Ge_{95} Sb_5 alloy annealed at 423 K and 593K for different times was investigated by X-ray (for brivity) as shown in Figs. 6 and 7, respectively. The detected peaks in the X-ray charts reveal that some partial crystallization occurs after annealing. The growth of these peaks with increasing the annealing time shows the growth of the crystalline phase on the expense of the amorphous one. The detected crystalline phases were found to be Ge in the tetragonal form and Sb in the hexagonal form. Also, a new crystalline phase in the hexagonal form was appeared and it may be due to Ge-Sb phase as shown in Table 1 for Ge_{95} Sb_5 alloy . The crystalline Ge_{100-x} Sb_x phases obtained by annealing showed the presence of Ge crystalline phase in Ge-Sb matrix [9].

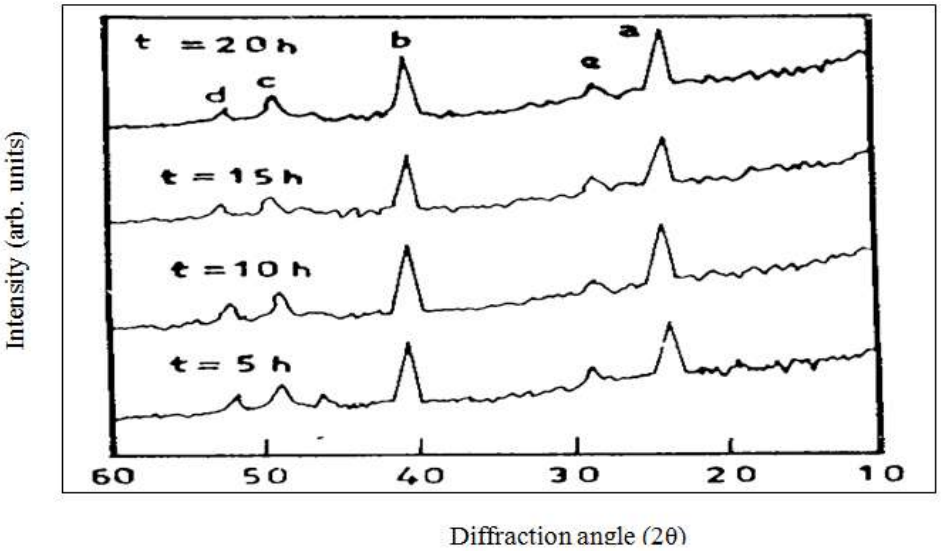

Fig. 6 . X-ray diffraction records for $Ge_{95}Sb_5$ alloy annealed at 423 K for different annealing times

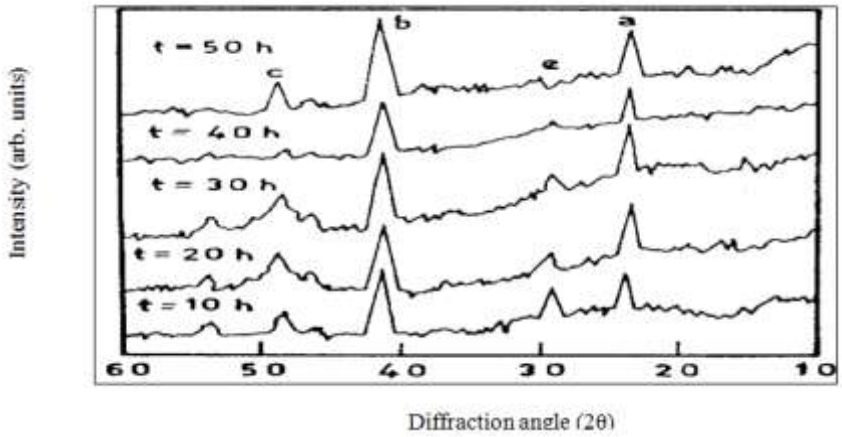

Fig. 7: X-ray diffraction records for amorphous $Ge_{95}Sb_5$ bulk sample annealed at 593 K.

IV. Conclusion

The thermal stability of Ge_{100-x} Sb_x alloys was investigated by measuring the thermoelectric power as a function of annealing time and annealing temperature. The results revealed that increasing the thermoelectric power with increasing the annealing time or rising the Sb content is due to transition from amorphous to crystalline forms which leads to increasing the optical energy gap.

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