

Determination of continuous electrical conductivity parameters and their influence on Partial Replacement Of Antimony With Sn In The $\text{Ge}_{20}\text{Te}_{72}\text{In}_8$ Chalcogenide Glass

Laheeb A. Mohammed ^{a*}, Zainab J. Neamah ^a, Anaam W. Watan ^a, Kareem Ali Jasim ^a and , Auday H. Shaban ^b

^a Department of Physics-College of Education for Pure Sciences Ibn Al-Haitham, University of Baghdad, Baghdad 10071, Iraq

^b Department of Remote Sensing & GIS, College of Science, University of Baghdad, Baghdad 10071, Iraq

* Corresponding author. Tel.: +9647818253987; e-mail: laheeb.a.m@ihcoedu.uobaghdad.edu.iq

Received 11 November 2022, Revised 2 December 2023, Accepted 7 March 2024

ABSTRACT

In the present work, the changes in electrical conductivity upon partial replacement of antimony by Tin (Sn) in the ternary alloy prepared by the molten cooling method were synthesized and studied. The electrical measurements were performed on $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ Chalcogenide glass alloy with $x = 0, 2, 4,$ and 6 . The dark conductivity (σ_d) increases with the increase in temperature in all the samples under the experiment. Observations of data were made at low (300 – 335K), medium (335 – 365K), and high temperatures (365 – 400K) across three distinct regions regarding electrical conductivity. For each of the three conduction regions, the factor that precedes the exponent and the effective energies were calculated using electrical conductivity measurements as a function of temperature. It was found that all of them were affected by an increase in the value of Sn. A numerical analysis of the conductivity equation was also carried out for the densities of the energy for the extended states, localized states, and at the Fermi level. It has been observed that all of them change with the change in the value of Sn.

Keywords: *Electrical conductivity, partial replacement, dark conductivity, conduction regions, extended states*

1. INTRODUCTION

Chalcogenides are substances that are made up of many layers, and the layers are held together by weak van der Waals bonds. As in the case of many organic isotopes, glassy chalcogenides with a random structure have the ability to change their electronic properties due to internal structural rearrangement. However, it has been shown that reshaping these materials via partial grafting or composite addition is difficult in terms of structural integrity due to the flexibility of the amorphous structure, which allows the formation of highly favorable coordination defects called valence and alternating states [1]. The influence of semiconductors amorphous materials on their physical, structural, and electronic properties containing one or more chalcogens have often been considered controversial, but lithium, but lithium and bismuth proven to mix with other elements such as lead, change conductivity in molten glass [2, 3].

The doping of a semiconductor remains stable if the type of semiconductor is p-type or n-type. In the case of Bi and Pb doping, it is inversely proportional to the carrier type, from p-type to n-type, in a chalcogenide semiconductor with a random structure with balanced steroids. The origin of n-type conductivity in bismuth-modified bulk chalcogenide glass was explained by the high polarizability of bismuth, which promotes the formation of partially ionic double bonds of the chalcogen. From theoretical considerations, filtration-controlled conduction by n-type impurities has been proposed in heterogeneous amorphous

chalcogenides [5, 6]. It has been recommended that the 8-N rule figured out by Mott does not sufficiently depict the contamination band hole conditions of formless chalcogenides, whose compound pieces are to a great extent inhomogeneous. In this case, the electrical properties of the indistinct grid are not entirely settled by the nuclear construction of the nanostructured unpredictable units, which can oblige the rising coordination of particles around the current pollution. The debasement iota inside the nanoscale consideration acts as it would in a glasslike semiconductor [6].

The electrical properties of glasses based on one or more chalcogen elements are fascinating, as these materials are key components in electronic devices like optical memory and optical shading [7, 8] By analyzing their electrical behavior, we can learn more about them. Uncoated chalcogenide glasses have low electrical conductivity in general, which could limit their technological and electrometric applications significantly. To improve these qualities, several additives are utilized. Due to their high hardness, high light sensitivity, and high crystallization temperature, selenium, and tellurium alloys have sparked a lot of attention. [9, 10], and smaller aging actions compared to pure amorphous selenium [11-13]. When germanium is added to the Se-Te system, it causes structural changes in the material, which modulate the band structure and consequently the material's electrical characteristics. The goal of this research is to present

various electrical parameters, assess the effect of tin content on the $\text{Ge}_{20}\text{Te}_{72}\text{In}_8$ system's continuous electrical conductivity, and define its temperature dependency. The work focuses on four samples with the following compositions: $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ ($x = 0, 2, 4,$ and 6). That is, tin replaces part of the $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ glass alloy composition (Sn). It will determine the density of the energy states inside and outside the energy gap.

2. MATERIALS AND METHODS

The $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ glass alloy ($x = 0, 2, 4,$ and 6) is prepared by the molten quenching technique. Substances with pure chemical elements (99.999% pure) are weighed according to their atomic proportions and then these components are mixed using a hermetic electric mixer and then packaged and sealed in quartz ampoules in a vacuum of $\sim 5 \times 10^{-4}$ bar. The sealed ampoules are kept in an oven where the temperature is raised to 950 K at a heating rate of 5°C/min. The ampoules are shaken repeatedly for 18 hours at the highest temperature to make the dissolution homogeneous. Cooling is done in ice water. The ampoules are then broken and the ingots are re-grinded separately in an electric mixer and then re-pressed in the form of discs with radii of one and a half cm and a thickness of 2 mm with a 7-ton/cm². The bulk conductivity of the $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ glass system ($x = 0, 2, 4,$ and 6) is evaluated using the conductivity technique as a function of temperature (measuring voltage and current with an increase in the temperature of the samples), the measurements were made in the temperature range 300-400K.

3. RESULTS AND DISCUSSIONS

Figure 1 shows the total dark conductivity (σ_d) measured as a function of the exchange temperature at different tin concentrations of the a- $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ glass alloy with $x = 0, 2, 4,$ and 6 . The curves $\log \sigma_d$ versus $1000/T$ divided into three straight lines { Low (300 – 335K), medium (335 – 365K), and high temperatures (365 – 400K)} in the measurement temperature range (300–400 K). This indicates that the conduction mechanisms in these glasses operate through an active process, each with a distinct activation energy within specific temperature ranges: low conduction occurs between 300-335 K, medium conduction between 335-365 K, and high conduction between 365-400 K. Thus, the conductivity (σ_d) can be expressed using Mott's formula [12].

$$\sigma = \sigma_{01} e^{\left(\frac{-\Delta E_1}{KT}\right)} + \sigma_{02} e^{\left(\frac{-\Delta E_2}{KT}\right)} + \sigma_{03} e^{\left(\frac{-\Delta E_3}{KT}\right)} \quad (1)$$

where ($\sigma_{01}, \sigma_{02}, \sigma_{03}$) are previous pre-exponential factor parameters, $KB =$ Boltzmann's constant, ($\Delta E_1, \Delta E_2, \Delta E_3$) are the activation energy of each term and $T =$ the absolute temperature.

The values of activation ($\Delta E_1, \Delta E_2, \Delta E_3$) and ($\sigma_{01}, \sigma_{02}, \sigma_{03}$) at the temperature range (300–400 K) were calculated from the curves drawn in Figure 1. By taking the slope of each region of the curve to calculate the values

of ΔE . As for the values of σ_0 , they were obtained from the intersections of the extension of the curves procedure with the y-axis depending on the limits of equation (1) [14]. The results are listed in Table 1. It is clear from Table 1 that the value of ΔE changes as follows 0.099, 0.195, 0.221, 0.198 in the region of high temperature (for extended states from 365 to 400K), 0.691, 1.093, 0.976, 1.093 (for localized states from 335 to 365K), 0.314, 0.562, 0.617 and 0.600 (Fermi states from 300 to 335K) when the Tin (Sn) values increase from 0 to 6 for samples $\text{Ge}_{20}\text{Te}_{72}\text{In}_8, \text{Ge}_{20}\text{Te}_{72}\text{In}_6\text{Sn}_2, \text{Ge}_{20}\text{Te}_{72}\text{In}_4\text{Sn}_4,$ and $\text{Ge}_{20}\text{Te}_{72}\text{In}_2\text{Sn}_6$, respectively which is illustrated in Figure 1 with multi regions highlighted with red line to separate the regions. The values of σ_0 are shown in Table 1.

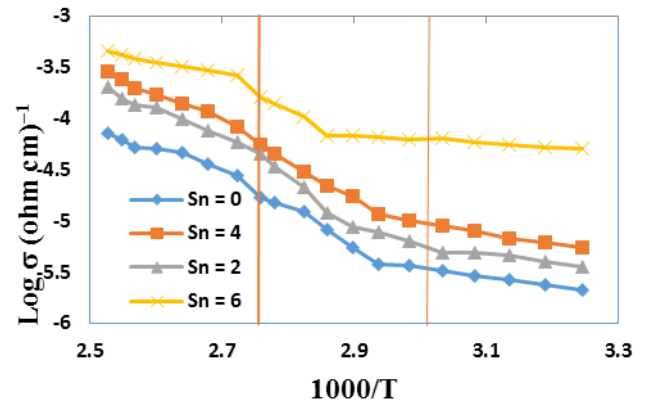


Figure 1 The plots of $\log(\sigma)$ versus $1000/T$ for $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ ($x = 0, 2, 4$ and 6) glassy system

Table 1 The composition dependence of activation energies and pre-exponential factor σ_0 for three regent states with different Tin (Sn) concentrations of $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ ($x = 0, 2, 4$ and 6) glassy system

x	ΔE_1 (eV)	$\sigma_{01} \times 10^{-2}$	ΔE_2 (eV)	σ_{02}	ΔE_3 (eV)	σ_{03}
0	0.099	18.3	0.691	3.6×10^6	0.314	2.4
2	0.195	25.1	1.093	4.4×10^6	0.562	453
4	0.221	4.87	0.976	4.7×10^7	0.617	292
6	0.198	3.25	1.135	3.4×10^7	0.600	997

Figure 2 addresses the connection between the enactment energies in the three districts (broadened, restricted, and the closeness of the Fermi level) as an element of Sn fixation, It should be visible from this figure that the initiation energies in the lengthy area and close to the Fermi level increment with the expansion in the grouping of Sn, while the actuation energies of the confined states increments at the centralization of tin 0.2 and diminishes marginally when filled it 0.4, yet increments to the most extreme worth at the convergence of tin 0.6 and that the adjustment of the initiation energies in the three locales might result from a few Reasons [14-16], including:

- 1- The change in the value of the kinetic energy gap.
- 2- Move the Fermi level from the center (up or down).

- 3- A change in the values of the width of the tails of the beams ΔE .
- 4- The shift in the type of charge carrier as a result of partial substitution of type n or type p.
- 5- An increase or decrease in the concentration of carriers.

The use of Table 1 values in the equations [15, 16],

$$a = 0.026 \frac{e^2}{\hbar\sigma_0} \text{ and } R = 0.7736 \left[\frac{\Delta E a^{-1}}{N(E_c)(KT)^2} \right]^{0.25}$$

Where $\Delta E = E_1 - E_2$ is width of energy tail and α^{-1} is optical absorption coefficient is 10^{-7}cm

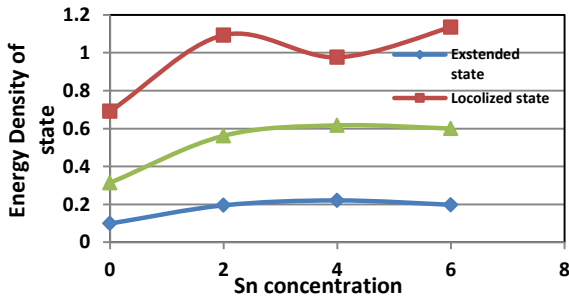


Figure 2 The plots of ΔE_1 , ΔE_2 , and ΔE_3 as a function of Tin concentration for $\text{Ge}_{20}\text{Te}_{72}\text{In}_6\text{Sn}_2$ glassy system

the atomic distance (a), hopping distance (R), and tail width (ΔE_T) were computed, and the findings were published in Table 2.

The interatomic distance (a) was estimated using the equation $a = 0.026 [e^2/\hbar\sigma_0]$ using Table 2's hopping distance R and tail state width (ΔE_T).

The relationship between the inter-atomic distance (a), the hopping distance (R), and the width of the tails (ΔE_T) as a function of the tin element concentration in the samples is shown in Figure 3, where the increase in the values of a becomes extremely obvious with the increase in concentration.

Table 2. Values of the electronic characteristics with respect to Cu concentration for $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ (x = 0, 2, 4 and 6) glassy system

x	Tail Width ΔE_T (eV)	R(A)	a(A0)	N(Eext) $\text{eV}^{-1}\text{cm}^3$	N(E_{loc}) $\text{eV}^{-1}\text{cm}^3$	N(E_F) $\text{eV}^{-1}\text{cm}^{-3}$
0	0.299	16.86	59.61	5.92×10^{18}	2.97×10^{15}	3.8×10^{11}
2	0.898	18.461	43.47	8.12×10^{18}	3.03×10^{15}	3.34×10^9
4	0.755	22.666	224.05	1.575×10^{18}	2.15×10^{16}	1.47×10^{11}
6	0.952	30.90	355.72	1.051×10^{18}	8.34×10^{15}	4.26×10^{10}

Figure 3 represents the relationship between the Atomic distance (a), hopping distance (R), and tail width (ΔE_T) as a function of Tin concentration for the $\text{Ge}_{20}\text{Te}_{72}\text{In}_6\text{Sn}_2$ glassy system. it was found that all those parameters changed with increasing Tin concentrations.

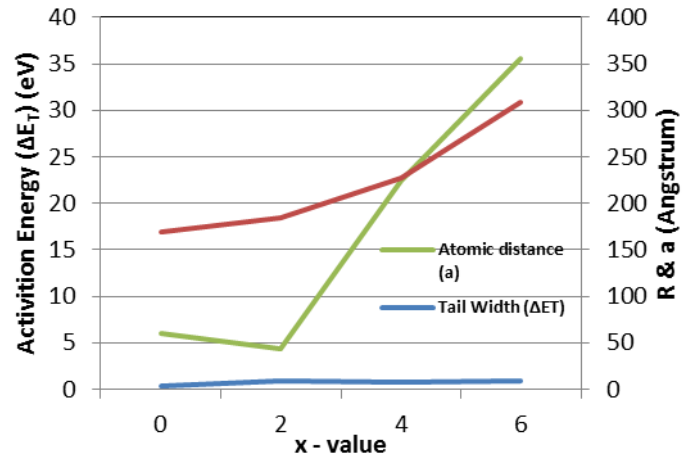


Figure 3. The plots of The Atomic distance (a), hopping distance (R), and tail width (ΔE_T) as a function of Tin concentration for the $\text{Ge}_{20}\text{Te}_{72}\text{In}_6\text{Sn}_2$ glassy system

As a function of Sn concentration, Figure 4 depicts the link between the density of extended $N(E_{ext})$ and localized energy states $N(E_{loc})$ and the Fermi level's vicinity $N(E_F)$. The density of local and extended states decreases as the concentration of Tin (Sn) = 0.2 increases, peaking at 0.4, whereas the density of energy states near the Fermi level exhibits the opposite behavior, due to changes in activation energies in the three regions and for the reasons stated above. All or some of these factors could lead to varying energy levels in the body.

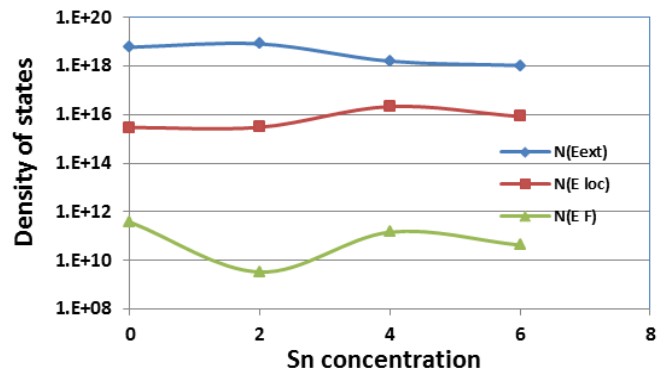


Figure 4 The density of states and the proximity of the Fermi level with respect to the Tin concentration for the $\text{Ge}_{20}\text{Te}_{72}\text{In}_6\text{Sn}_2$ glassy system

4. CONCLUSION

In the work, four samples were synthesized from chalcogenide glass alloy with the chemical formula $\text{Ge}_{20}\text{Te}_{72}\text{In}_{8-x}\text{Sn}_x$ glass alloy with different substitution ratios x = 0, 2, 4, and 6 of elemental tin by molten quenching method. The changes in electrical conductivity were studied upon partial replacement of antimony with

tin at values of tin 0, 2, 4, and 6. Electrical measurements were performed on the alloy samples. The dark conductivity (σ_d) increases with increasing temperature in all samples under the experiment. From the results above, it can be concluded that the pre-exponential factor parameters σ_0 and activation energy ΔE which were calculated using electrical conductivity measurements for all samples under experiment changes with increasing Tin concentration. It is clear from the obtained results that the ΔE values change as follows 0.099, 0.195, 0.221, 0.198 in the high temperature region (for extended cases), 0.691, 1.093, 0.976, 1.093 (for localized cases), 0.314, 0.562, 0.617 and 0.600 (Fermi states) when Tin (Sn) values increase from 0 to 6 for samples $\text{Ge}_{20}\text{Te}_{72}\text{In}_8$, $\text{Ge}_{20}\text{Te}_{72}\text{In}_6\text{Sn}_2$, $\text{Ge}_{20}\text{Te}_{72}\text{In}_4\text{Sn}_4$ and $\text{Ge}_{20}\text{Te}_{72}\text{In}_2\text{Sn}_6$, respectively. The density of the state was calculated in the localized and extended regions. It was found that the density of local and extended states decreases with the increase in the Tin concentration (Sn = 0.2), and increases to the greatest value when the concentration of Tin equal to 0.4, while the density of energy states near the Fermi level is the opposite of the behavior and this is due to the change in the activation energies in the three regions.

ACKNOWLEDGMENTS

The authors are thankful to Baghdad University, (<https://uobaghdad.edu.iq>), College of Education for Pure Science (Ibn-AL-Haitham), and the Physics Department for the award of Major Research Project.

REFERENCES

- [1] R. A. Street and N. F. Mott, "States in the Gap in Glassy Semiconductors," *Phys. Rev. Lett.*, vol. 35, pp. 1293-1296, 1975.
- [2] B. A. Khan and D. Adler, "Chemical modification of chalcogenide glasses by lithium," *J. Non-Cryst. Solids*, vol. 64, pp. 35-42, 1984.
- [3] M. Frumar and L. Tichý, "N-type conductivity in chalcogenide glasses and layers," *J. Non-Cryst. Solids*, vol. 97, pp. 1139-1146, 1987.
- [4] S. R. Elliott and T. A. Steel, "Mechanism for Doping in Bi Chalcogenide Glasses," *Phys. Rev. Lett.*, vol. 57, pp. 1316-1319, 1986.
- [5] L. Tichy, H. Iicha, A. Triska, and P. Nagels, "Is the n-type conductivity in some Bi-doped chalcogenide glasses controlled by percolation?," *Sol. St. Commun.*, vol. 53, pp. 399-402, 1985.
- [6] T. F. Mazec and K. D. Tsendin, "On Mechanism of Doping of Chalcogenide Glassy Semiconductors," *Physica i Tekhnika Poluprovodnirov*, vol. 24, pp. 1953-1958, 1990.
- [7] B. A. Omar, S. J. Fathi, and K. A. Jassim, "AIP Conference Proceedings," *AIP Conf. Proc.*, vol. 1968, p. 030047, 2018.
- [8] J. S. Berkes, S. W. Ingand, and W. J. Hillegas, "J. Appl. Phys.," vol. 42, p. 4908, 1971
- [9] A. K. Saadon, A. M. Mhayyal, and K. A. Jasim, "Effect of BaTiO₃ mixture on the structural, electrical properties and morphology for PET/BaTiO₃ composite," *AIP Conf. Proc.*, vol. 2307, p. 020014, 2020. [Online]. Available: <https://doi.org/10.1063/5.0035304>.
- [10] B. B. Kadhim, R. H. Risan, A. H. Shaban, and K. A. Jasim, "Electrical characteristics of nickel/epoxy - Unsaturated polyester blend nanocomposites," *AIP Conf. Proc.*, vol. 2123, pp. 020062-1-020062-5, 2019. [Online]. Available: <https://doi.org/10.1063/1.5116989>.
- [11] B. T. Kolomieto, "Phys. Status Solidi," vol. 7, p. 713, 1964.
- [12] N. F. Mott and E. A. Davis, *Electronic processes in non-crystalline materials*, Oxford: Clarendon Press, Oxford Univ. Press, 1971, pp. 437. [Online]. Available: <https://doi.org/10.1016/0040-6090>.
- [13] L. A. Mohammed and K. A. Jasim, "Synthesis and Study the Structural and Electrical and Mechanical Properties of High Temperature Superconductor Tl_{0.5}Pb_{0.5}Ba₂Ca_{n-1}Cu_xNi_xO_{2n+3-δ} Substituted with Nickel Oxide for n=3," *Ibn Al-Haitham J. Pure Appl. Sci.*, vol. 31, no. 3, pp. 26-32, 2018.
- [14] A. N. Abdulateef, A. Alsudani, R. K. Chillab, K. A. Jasim, and A. H. Shaban, "Calculating the Mechanisms of Electrical Conductivity and Energy Density of States for Se₈₅Te₁₀Sn_{5-x}In_x Glasses Materials," *J. Green Eng.*, vol. 10, no. 9, pp. 5487-5503, Sep. 2020.
- [15] R. K. Chillab, S. S. Jahil, K. M. Wadi, K. A. Jasim, and A. H. Shaban, "Fabrication of Ge₃₀Te_{70-x}Sb_x Glasses Alloys and Studying the Effect of Partial Substitution on D.C Electrical Energy Parameters," *Key Eng. Mater.*, vol. 900, pp. 163-171, 2021.
- [16] B. A. Ahmed, J. S. Mohammed, R. N. Fadhil, K. A. Jasim, A. H. Shaban, and A. H. Al Dulaimi, "The dependence of the energy density states on the substitution of chemical elements in the Se₆Te_{4-x}Sb_x thin film," *Chalcogenide Lett.*, vol. 19, no. 4, pp. 301-308, Apr. 2022.
- [17] B. A. Aljurani, G. Hermiz, and M. Alias, "Superconductivity Measurements of (Hg,Tl)-1223 Compound Prepared in Capsule," *Iraqi J. Sci.*, vol. 62, no. 9, pp. 2934-2939, 2021.
- [18] K. A. Jassim, W. H. Jassim, and S. H. Mahdi, "The effect of sunlight on medium density polyethylene water pipes," *Energy Procedia*, vol. 119, pp. 650-655, 2017.
- [19] K. A. Jasim, "The effect of neutron irradiation on the properties of Tl_{0.6}Pb_{0.3}Cd_{0.1}Ba₂Ca₂Cu₃O_{9-?} Superconductors," *Turkish J. Phys.*, vol. 37, no. 2, pp. 237-241, 2013.