



Impact of Sb Addition on Physical Properties of Se-Te Glassy Semiconductors

¹Parikshit Sharma, ²Shallu Jamwal, ³Deepakshi Jamwal

¹Department of Physics, Abhilashi University, Chailchowk, Mandi (175028) India

²Department of Physics, Maharaja Agrasen University, Baddi, Solan (174103) India

Abstract: The effect of Antimony (Sb) addition in $\text{Se}_{88-x}\text{Te}_{12-x}\text{Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) glassy alloy on various physical properties via coordination number, effect of lone pairs, mean bond energy, heat of atomization, energy band gap, glass transition temperature and degree of stoichiometry of is investigated. It is concluded that on increasing Sb content, average coordination number, mean bond energy, average heat of atomization and glass transition temperature increases whereas number of lone pairs, energy band gap and degree of heat of stoichiometry decreases.

Index Terms - Chalcogenide glasses, mean bond energy, glass transition temperature, energy band gap.

I. INTRODUCTION

Chalcogenide glassy semiconductors (ChGS) are one of the best materials for various scientific investigations from many past years till date. They have shown tremendous potential in the field of optoelectronics and optics. These materials show a flexible structure, huge property variations and almost limitless doping and alloy strength for chemical sensors in research into industrial aqueous solutions to control pollutant gases [1]. These glasses reveal low phonon energy and show good precision for light in the visible and mid-infrared spectrum region. These materials certify a constant change in the physical properties with change of chemical composition [2]. The glass-formation region of tin-based binary and ternary chalcogenide glasses is very constrict and equates to a small amount of tin in the alloy. Hence, no adequate studies are accounted in literature on tin-based chalcogenide glasses. Georgieva and the fellow researchers [3] have investigated the influence of crystallisation on the electronic and optical characteristics of Se-Te-Sn thin films. Based on their properties such as large band gaps, significant absorption and binding energy coefficients, high chemical stability and ecological applications, chalcogenide alloys and compounds and their thin films are of tremendous importance in the field of scientific research. As a result, these compounds were considered to be worthy for various optical and optoelectronic applications [4]. Se-based chalcogenide glasses are assuring materials due to their several applications like optical fibres, memory devices, switching devices and solar cells. Se in its pure state has disadvantages such as short lifetime, low sensitivity and low crystallization temperature. In order to overpower these faults, the addition of Te improves the corrosion resistance and optical sensitivity [5]. Because of their excellent laser writer sensitivity, xerography, and electrographic applications such as photoreceptors in photocopying and laser printing, the glassy alloys of the Se-Te method based on Se are commonly used for numerous applications in many fields as optical recording media [6]. The high photo sensitivity, greater hardness and higher crystallisation temperature of Se-Te alloys is particularly interesting [7].

Impurity results in chalcogenide glasses are important in the production of glassy semiconductors. When these impurity atoms join the glassy network, they are expected to fulfil all valence demands and hence are not expected to assume the roles of acceptors or donors. The result of impurity atoms in chalcogenide glasses is determined by the glass structure, the chemical quality of the impurity, and the value of impurity concentration [8].

According to a recent analysis, the addition of metallic impurities has an important impact on the properties of chalcogenide glasses. The ordered bond network model, also known as the chemically ordered network model, is often used to describe the impact of compositional variance on the physical properties of chalcogenide glasses [9]. The electrical properties of chalcogenide glasses have been extensively researched in light of their uses in solid-state electronic devices. Se-Te based glasses have fascinated significant interest due to their extensive applications in semiconductor devices. They are used as optical recording medium due to their good laser write sensitivity. It has been reported that properties of these glasses are appreciably affected by the presence of third impurity in the Se-Te matrix. The addition of antimony (Sb) also increases the glass forming ability of Se-Te system. The addition of Sb impurity brings out configurationally and structural changes in the host system that not only increases the stability of the system but also enlarges their sphere of the application in various devices [10]. This stimulated to us to study the Se-Te-Sb system which varies from

floppy mode to rigid mode [11]. In Se-Te system, Se atoms act as bond modifiers, which increase average bond strength by cross linking with Te. This stiffens the device, increasing the glass transition temperature and resistivity. The addition of a third impurity changes the local atomic composition, which influences the physical properties of the device [12].

The aim of the present work is to examine the system $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) theoretically for various physical parameters. The physical parameters viz. coordination number, number of constraints, number of lone pair electrons, bond energies of the different bonds formed in the system, heat of atomization, average single bond energy (which is a measure of cohesive energy [11]), mean bond energy, degree of stoichiometry and glass transition temperature. The glass transition temperature has been theoretically investigated using the model proposed by Tichy and Ticha [11].

II. EXPERIMENTAL DETAILS

Bulk samples $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) were prepared by conventional melt quenching technique. High purity (99.999%) elements, Se, Te and Sb in the appropriate weight proportion, were vacuum sealed (10^{-4} Pa) in quartz ampoules and heated up to 800°C in a rocking furnace at a heating rate of $3\text{-}4^\circ\text{C}/\text{min}$, the ampoules were frequently rocked at 800°C for 8 hrs. Since removing the ampoules from the furnace, they were quickly quenched in ice-cold water [13]. The bulk samples amorphous existence was confirmed by X-ray diffraction, which revealed no sharp peak in the spectra.

III. RESULTS AND DISCUSSION

3.1 Calculation of Coordination Number (M) and Number of Constraints in Glassy Network

Nearest neighbour coordination number (m) in the ternary system $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) is suitable for proving the correctness of topological ideas [14, 15] because of its large glass forming domain. The average coordination number in our system has been calculated using the relation

$$m = \frac{[\alpha N_{\text{Se}} + \beta N_{\text{Te}} + \gamma N_{\text{Sb}}]}{100} \quad (3.1)$$

where α , β and γ are the at % of Se, Te and Sb respectively and N_{Se} , N_{Te} and N_{Sb} are their respective coordination numbers. The calculated coordination numbers (m) lie in the range $2 \leq m \leq 2.10$ and are given in Table 1.

Table 1: Values of the average co-ordination number (m), number of constraints arising from bond stretching (N_a), number of Constraints arising from bond bending (N_b), average number of Constraints (N_c), effective co-ordination number $\langle m_{\text{eff}} \rangle$ for $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) glassy alloys.

| Composition | M | N_a | N_b | N_c | $\langle m_{\text{eff}} \rangle$ |
|---|------|-------|-------|-------|----------------------------------|
| $\text{Se}_{88}\text{Te}_{12}$ | 2 | 1 | 1 | 2 | 2 |
| $\text{Se}_{88}\text{Te}_{10}\text{Sb}_2$ | 2.02 | 1.01 | 1.04 | 2.05 | 2.02 |
| $\text{Se}_{88}\text{Te}_8\text{Sb}_4$ | 2.04 | 1.02 | 1.08 | 2.1 | 2.04 |
| $\text{Se}_{88}\text{Te}_6\text{Sb}_6$ | 2.06 | 1.03 | 1.12 | 2.15 | 2.06 |
| $\text{Se}_{88}\text{Te}_4\text{Sb}_8$ | 2.08 | 1.04 | 1.16 | 2.2 | 2.08 |
| $\text{Se}_{88}\text{Te}_2\text{Sb}_{10}$ | 2.10 | 1.05 | 1.20 | 2.25 | 2.10 |

The covalent bonded glassy networks are affected by mechanical constraints (N_c) i.e. bond stretching (N_a) and bond bending (N_b) which are associated with atomic bonding and effective coordination number $\langle m_{\text{eff}} \rangle$. The number of constraints per atom arising from bond bending can be calculated by $N_b = 2m - 3$ and from bond stretching by $N_a = m/2$ for the atomic species having coordination number (m). For different compositions of the glassy system $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$), knowing the average number of constraints i.e. $N_c = N_a + N_b$ and the average coordination number (m), the effective coordination number $\langle m_{\text{eff}} \rangle$ can be calculated

$$\langle m_{\text{eff}} \rangle = \frac{2}{5}(N_c + 3) \quad (3.2)$$

The calculated values of N_a , N_b , N_c and $\langle m_{\text{eff}} \rangle$ for the glassy system $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) are listed in Table 1. According to Thorpe [16], in the range of the glass forming compositions, the system should contain floppy and rigid regions.

According to Zachariassen [17] heteropolar bonds have supremacy over the formation of homopolar bonds. This condition is equivalent to assuming the maximum amount of chemical ordering possible. This means that the bonding between atoms will happen only when excess of a certain atom class is found, such that its valence requirements cannot be satisfied by binding them alone to atoms of different types, such that bonds are formed in the series of decreasing bond energy before all valences available of the atoms are saturated. Ovshinsky et al [18] will use chemically ordered network (CON) model to demonstrate potential bond propagation in different formulations.

This model is based on a prominent assumption that atoms combine more favorably with atoms of different kinds than with atoms of the same kind. Also bonds form in a decreasing bond energy series until all of the atoms available valences are saturated. The E_{A-B} bond energies have been determined using the relationship with heteronuclear bonds [19].

$$E_{AB} = [E_{AA} + E_{BB}/2] + 23 [\chi_A - \chi_B]^2 \quad (3.3)$$

where E_{A-A} and E_{B-B} are the bond energies of the homonuclear bonds and χ_A and χ_B are the electronegativities of the atoms involved. The values of the electronegativities of Se, Te and Sb are 2.55, 2.10 and 2.05 respectively. The calculated values for different bonds are given in Table 2.

Table 2: Values of the number of Lone Pair electrons and bond energies of different bonds possible in $Se_{88}Te_{12-x}Sb_x$ ($x = 0, 2, 4, 6, 8, 10$) glassy alloys.

| Composition | M | V | L = V-m | Bonds | Bond energy (eV/bond) |
|----------------------|------|------|---------|-------|-----------------------|
| $Se_{88}Te_{12}$ | 2 | 6 | 4 | Se-Sb | 3.339 |
| $Se_{88}Te_{10}Sb_2$ | 2.02 | 5.98 | 3.96 | Se-Se | 3.429 |
| $Se_{88}Te_8Sb_4$ | 2.04 | 5.96 | 3.92 | Te-Sb | 2.902 |
| $Se_{88}Te_6Sb_6$ | 2.06 | 5.94 | 3.88 | Te-Te | 2.673 |
| $Se_{88}Te_4Sb_8$ | 2.08 | 5.92 | 3.84 | Sb-Sb | 3.130 |
| $Se_{88}Te_2Sb_{10}$ | 2.10 | 5.90 | 3.80 | Te-Se | 3.099 |

3.2 Role of Lone Pair Electrons in the Glass Forming Ability

The number of lone pairs in a chalcogenide glass structure can be calculated using the average coordination number suggested by Phillips [14]. The number of lone pair electrons is equal to the difference of all the valence electrons of the system and the shared electrons i.e.

$$L = V - m \quad (3.4)$$

where L and V are lone pair electrons and valence electrons respectively. For the glassy system $Se_{88}Te_{12-x}Sb_x$ the number of lone pair electrons is obtained by using equation [4] and are listed in Table 2. It is inferred from the above table and from Fig.1 that with the increase in content of Sb the number of lone pair of electrons decreases continuously for $Se_{88}Te_{12-x}Sb_x$ ($x = 0, 2, 4, 6, 8, 10$) glassy system.

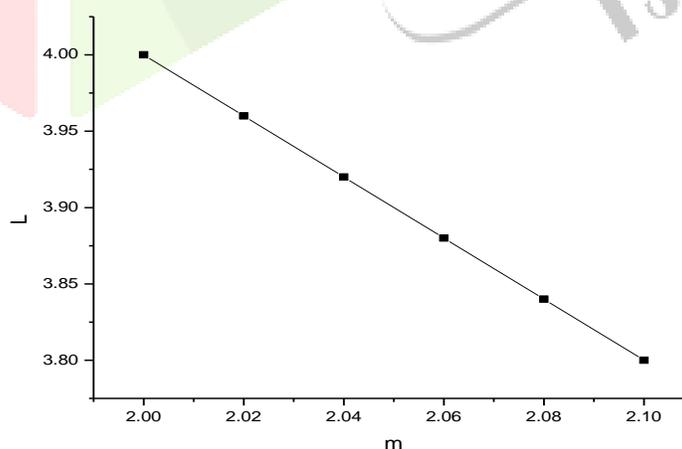


Figure 1 shows the variation of lone pair of electrons (L) with average coordination number $\langle m \rangle$

The association between the Sb ion and the lone pair electrons of a bridging Se atom causes this effect. The lone-pair electrons' position in glass forming is reduced as a result of the interaction. Zhenhua [20] proposed a basic metric for calculating a chalcogenide system's ability to maintain its vitreous state; the criterion includes the amount of lone pair electrons needed to achieve the system's vitreous state. The number of lone-pair electrons in a binary system must be greater than 2.6, and in a ternary system, it must be greater than 1. The values of lone pair electrons in our case are in the range $3.80 \leq L \leq 4$. The fact improvise that the system can be obtained in a glassy state.

3.3 Average Heat of Atomization

The lone-pair forms the top of the valence band in chalcogenide glasses containing a high concentration of group VI element, whereas the antibonding band forms the bottom conduction band [21]. The energy differential between the top of the valence band and the bottom of the conduction band closely correlates to the optical band. Thus due to the presence of a high-energy lone pair on the latter, metal atoms will form a dative bond with group VI atoms (lone pair with empty orbital) at no cost of energy. Dative bonds conform to (empty) antibonding levels in the distance [22], resulting in localised acceptor states.

Table 3: Values of Average heat of atomization, average single bond energy, mean bond energy, glass transition temperature and deviation from Stoichiometry for $\text{Se}_{88}\text{-Te}_{12-x}\text{-Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) glassy alloys.

| Composition | \bar{H}_s (eV/bond) | \bar{H}_s/m (eV/bond) | $\langle E \rangle$ | T_g (K) | R | E_g (eV) |
|---|--------------------------|----------------------------|---------------------|-----------|------|---------------|
| $\text{Se}_{88}\text{Te}_{12}$ | 2.31 | 1.155 | 3.34 | 758.84 | 7.33 | 1.7158 |
| $\text{Se}_{88}\text{Te}_{10}\text{Sb}_2$ | 2.32 | 1.1485 | 3.36 | 765.06 | 6.76 | 1.7136 |
| $\text{Se}_{88}\text{Te}_8\text{Sb}_4$ | 2.34 | 1.1470 | 3.38 | 771.28 | 6.28 | 1.7133 |
| $\text{Se}_{88}\text{Te}_6\text{Sb}_6$ | 2.35 | 1.1407 | 3.40 | 777.50 | 5.86 | 1.7117 |
| $\text{Se}_{88}\text{Te}_4\text{Sb}_8$ | 2.36 | 1.1346 | 3.42 | 783.72 | 5.50 | 1.7105 |
| $\text{Se}_{88}\text{Te}_2\text{Sb}_{10}$ | 2.38 | 1.1333 | 3.44 | 789.94 | 5.17 | 1.7090 |

The relationship between the optical band gap and the chemical bond energy is fascinating. The heat of atomization H_s (A – B) of a binary semiconductor formed from atoms A and B at normal temperature and pressure, according to Pauling [23], is the measure of the heat of formation ΔH and the average of the atomization H_s^A and H_s^B , which corresponds to the average non polar bond energy of the two atoms.

$$H_s(A - B) = \Delta H + \frac{1}{2}(H_s^A + H_s^B) \quad (3.5)$$

The first term in the above equation is equal to the square of the difference between the two atoms' electronegativities χ_A and χ_B

$$\Delta H \propto (\chi_A - \chi_B)^2 \quad (3.6)$$

To extend this concept to ternary and higher order semiconducting compounds, the average heat of atomization \bar{H}_s (in kcal per gram-atom) of a compound is described. $A_\alpha B_\beta C_\gamma$ is regarded as a direct indicator of cohesive energy and therefore of average bond strength given as

$$\bar{H}_s = \frac{\alpha H_s^A + \beta H_s^B + \gamma H_s^C}{\alpha + \beta + \gamma} \quad (3.7)$$

The heat of formation is obviously not included as part of the cohesive energy; however, it is a useful parameter for correlating the physical properties of semiconducting compounds. Since the electro negativities of the constituent elements, namely Se, Te, and Sb, are very similar in chalcogenide glasses, the heat of formation adds very little to the average heat of atomization, and in most cases of chalcogenide glasses, the heat of formation is uncertain. In the few materials for which heat of formation is known, it accounts for just 10% of the heat of atomization and is thus ignored. Hence for binary chalcogenide glasses $H_s(A - B)$ is given by

$$H_s(A - B) = \frac{1}{2}(H_s^A + H_s^B) \quad (3.8)$$

whereas for ternary and higher order compounds, \bar{H}_s is given by equation [7]. The values of heat of atomization for Se, Te and Sb elements are 226.0 kJ/mol, 197.0 kJ/mol and 262.04 kJ/mol, respectively, and the calculated average heat of atomization \bar{H}_s and average single bond energy (\bar{H}_s/m) is given in Table 3, where m is the average coordination number. According to Table 3, the heat of atomization increases as the Sb content increases, while the average single bond energy (\bar{H}_s/m), which is a metric of cohesive energy, decreases as the Sb content increases. With an increase in Sb material, the average single bond energy (\bar{H}_s/m) decreases, which can result in a decrease in the optical band gap [24].

3.4 Mean Bond Energy and Glass Transition Temperature

In the case of chalcogenide glasses the Tichy and Ticha covalent bond approach [25, 26] can be called a first approximation. The glass transition temperature is thought to be proportional to the mean bond energy $\langle E \rangle$, which is affected by factors such as average coordination number, degree of cross linking, bond energy, and bond existence. Taking both of these considerations into account, they analysed 186 chalcogenide glasses with T_g varying from 320 to 760 K and found a strong association between T_g and $\langle E \rangle$ in the form

$$T_g = 311[\langle E \rangle - 0.9] \quad (3.9)$$

which satisfies the Arrhenius relation for viscosity.

The mean bond energy of the system may be calculated using the relation

$$\langle E \rangle = E_c + E_{m} \quad (3.10)$$

where E_c is the overall contribution towards bond energy arising from strong bonds and E_m is the contribution arising from weaker bonds that remain after the number of strong bonds will become maximum. For $Se_xTe_ySb_z$ system (where $x+y+z=1$),

In the selenium rich region,

$$E_c = 2xE_{Te-Se} + 3zE_{Se-Sb} \quad (3.11)$$

$$E_m = \frac{2y-2x-3z}{\langle m \rangle} * E_{Se-Se} \quad (3.12)$$

The calculated values of the mean bond energy are given in Table 3. This is clear from mean bond energy data that when Sb content increases, the mean bond energy of the system increases.

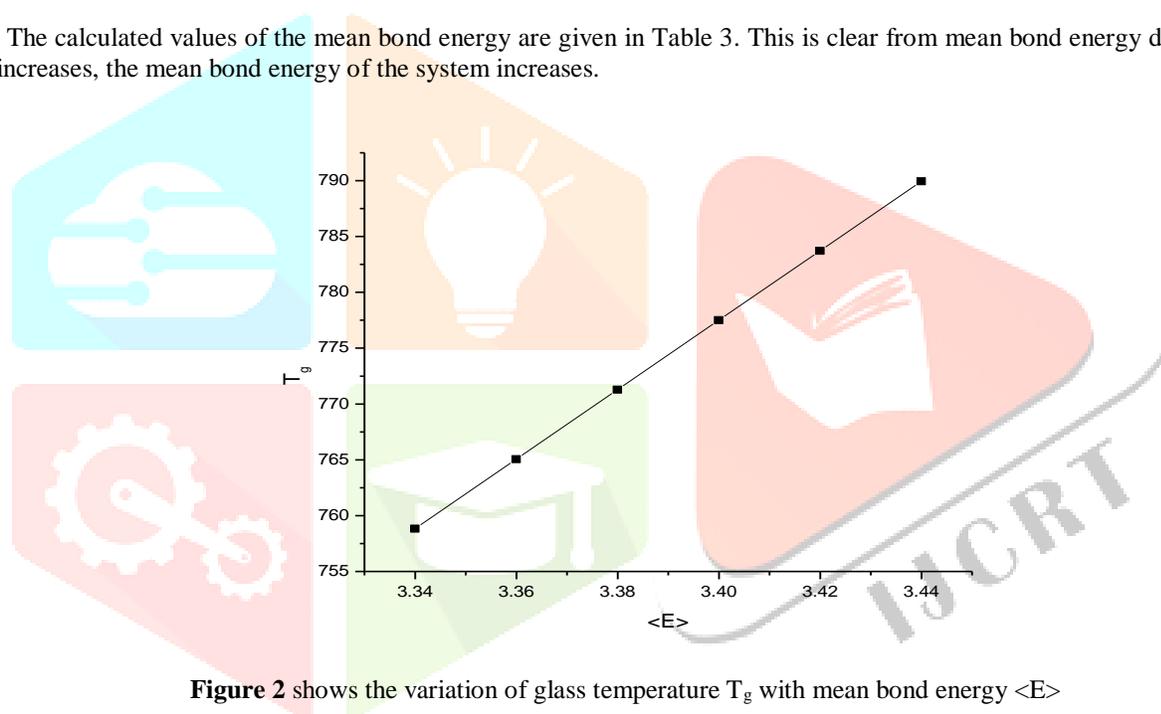


Figure 2 shows the variation of glass temperature T_g with mean bond energy $\langle E \rangle$

Figure 2 shows the variation of glass transition temperature with mean bond energy. The glass transition temperature increases with the increase of mean bond energy. The glass transition temperature for the various compositions of the system $Se_{88}Te_{12-x}Sb_x$ ($x = 0, 2, 4, 6, 8, 10$) have been calculated using equation [9] and is listed in Table 3. The glass transition temperature of the system under consideration indicates that as the Sb content increases, so does the glass transition temperature.

3.5 Theoretical Band Gap and Deviation of Stoichiometry

For binary system theoretically band gap (E_g) can be calculated by the relation given by Shimakawa [23]:

$$(E_{AB}) = V E_g(A) + (1-V) E_g(B) \quad (3.13)$$

where V is the volume fraction of element A and $(1-V)$ is the volume fraction of element B. $E_g(A)$ and $E_g(B)$ are energy gaps of elements A and B respectively. The same relation can be applied for ternary systems. It is inferred that with the addition of Sb in Te-Se alloy the band gap considerably increases. This can be associated with increase in average heat of atomization as well as decrease in electronegativity [24-25]. There is direct relationship between the Hs and E_g given by Aigrain and Balkanski [26], which leads to the important conclusion that the band gap increases with the increase in Sn content. Deviation of stoichiometry (R) is defined as the ratio of covalent bonding possibilities of chalcogen atoms. It is calculated by the relation:

$$R = YN_{Se}/(XN_{Te} + ZN_{Sb}) \quad (3.14)$$

where X, Y, Z are the atomic fractions of Te, Se and Sb respectively. This approach leads to an important conclusion that for $R > 1$, system is chalcogen rich and for $R < 1$, system is chalcogen poor and for $R = 1$, is threshold value where heteropolar bonds exists. From

Table 3 and Fig.3 it is clear that $R > 1$ for all the samples under consideration and hence system under investigation is chalcogen-rich region.

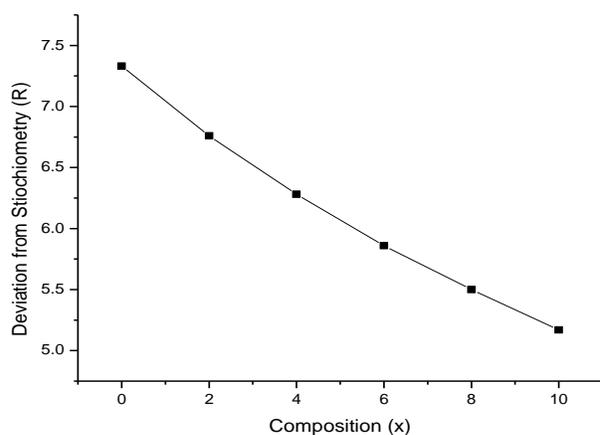


Figure 3 shows the variation of parameter R with Sb content.

IV. CONCLUSIONS

The thin films of $\text{Se}_{88-x}\text{Te}_{12-x}\text{Sb}_x$ ($x = 0, 2, 4, 6, 8, 10$) were analyzed for various physical parameters. The Average Coordination Number, Mean Bond Energy, Average Heat of Atomization, Energy Band Gap and Glass Transition Temperature increases with the increase in Sb content. On the other side Number of Lone Pair Electrons and Degree of Stoichiometry decreases with the relative increase in Sb content.

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