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Ultrasonic Characterization of $Bi_2(Te_{1-x}Se_x)_3$ System

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Longitudinal and shear ultrasonic wave velocities were measured in the ternary compounds $Bi_2(Te_{1-x}Se_x)_3$ $(0 \leq x \leq 1)$, using pulse echo technique. Measurements were carried out at 4 MHz frequency and at room temperature. Elastic moduli and acoustic impedance have been calculated. The results indicate that these parameters are influenced by the increased addition of selenium from $x = 0$ to 1.

Keywords: polycrystalline materials, Bi-Te-Se systems, ultrasonic wave velocity, elastic properties.

1. Introduction

There is an increasing interest in thermoelectric materials for refrigeration applications in industry and other areas like space, defense, biology and photonics as well as sensor applications; an increasing interest is expected to develop with advances and discoveries in nanotechnologies and microsystems. Ternary alloys made of semiconductor materials can be grouped as a class of semiconductors in which lattice parameter, energy band-gap and other operational parameters could be continuously varied per requirements by selecting the constituents and their relative concentrations suitably. The use of thin film polycrystalline semiconductors has attracted much interest in an expanding variety of applications in various electronic and optoelectronic devices [1–5]. Group V_2VI_3 compounds are known as good thermoelectric materials. These thermoelectric materials have been widely applied to cool electronic devices such as integrated circuit packages, laser diodes and infrared detectors, because quick and precise control of temperature is possible with almost no noise during operation [6–8], in particular the binary semiconductors Bi_2Te_2 and Bi_2Se_3 and their solid solutions, which have attracted much attention because of their potential application in the microfabrication of integrated thermoelectric devices [9, 10], and they have very high value of figure of merit [11, 12].

Solid solutions formed by melting of Bi, Se and Te are well-known to be the best materials for thermoelectric refrigeration at room temperature. A more interesting solid solution is that of the system $Bi_2(Te_{1-x}Se_x)_3$, where $(0 \leq x \leq 1)$. Structural parameters directly affect the optical and electrical properties of thin films of this system [13]. In particular, the preferential orientation plays a major role in determination of the physical properties of these films.

In many cases, neither the composition nor the crystal structure of the films of the system $Bi_2(Te_{1-x}Se_x)_3$ with different compositions, $(x = 0, 0.2, 0.4, 0.6, 0.8$ and $1)$, have been studied in sufficient detail to allow the knowledge of a correct avenue to follow, in order to improve the properties. Even though, many works have been done on this system with compositions x (where $x = 0, 1$ and < 0.1) [14–16]; there is no literature available on the structural, optical, electrical and mechanical properties of the system $Bi_2(Te_{1-x}Se_x)_3$, where $(x = 0.2, 0.4, 0.6, 0.8)$.

Longitudinal and shear ultrasonic wave velocities were measured by Gaafar *et al.* [17] in the quaternary compounds $Cu_xAg_{1-x}InTe_2$ $(0 \leq x \leq 1)$, using the pulse echo technique. They have reported that these compounds becomes close-packed and more rigid with the substitution of Cu instead of Ag, through the decrease in molar volume, increase in covalence bonds, the increase in ultrasonic velocities (longitudinal and shear) and consequently, the increase in elastic moduli.

The elastic constant C_{11} of $Al_xGa_{1-x}As$ was determined by Krieger *et al.* [18] using near-infrared Brillouin scattering. It showed a linear decrease with increasing Al concentration. This linear decrease in C_{11} was proposed to be dominated by the change in ionicity or the valence bonds in $Al_xGa_{1-x}As$.

Ultrasonic velocities in molten $(Ga_xIn_{1-x})As$ pseudo-binary alloys have been measured by Tsuchiya and Hisakabe [19] in the temperature range from liquidus temperature to 1280°C, under the saturated vapour pressure. The isotherm of the ultrasonic velocities showed a smooth increase from InAs to GaAs. They interpreted this increase in ultrasonic velocities since the fraction of covalent bonds in liquid GaAs is considerably larger, so that changes to a structure with more isotropic local arrangement with increasing temperature would be plausible to account for the observed increase in temperature dependence of the sound velocity.

To our knowledge, no ultrasonic study has been done before on the ternary compounds $Bi_2(Te_{1-x}Se_x)_3$ $(x = 0.0, 0.2, 0.4, 0.6, 0.8$ and $1.0)$. Therefore, this paper is aimed at investigation of the structure of these compounds, using ultrasonic pulse-echo technique.

2. Experimental

2.1. Preparation of samples

Ingots of $\mathrm{Bi_2(Te_{1-x}Se_x)_3}$ solid solutions ($x = 0, 0.2, 0.4, 0.6, 0.8, 1.0$) were prepared by the Bridgman Technique (described elsewhere in [20]) of a mixture of the constituent elements in stoichiometric proportions. Elements of 99.999% pure bismuth, selenium and tellurium (Aldrich Mark) were weighed in stoichiometric ratio using electric balance which was characterized with sensitivity up to 10^{-4} gm (Sartorius Mark). The mixture of each composition was taken in a silica tube, sealed under vacuum of 10^{-6} torr.

2.2. Density measurements

Density ρ of all samples was measured employing the Archimedes principle at 298 K, using toluene as a buoyant and applying the relation:

$$\rho = \rho_b \left(\frac{W_a}{W_a - W_b} \right), \tag{1}$$

where ρ_b is the density of the buoyant, W_a and W_b are the sample weights in air and the buoyant respectively. The experiment was repeated three times, and the estimated error in density measurement for all samples was found to be ± 0.001 g/cm^3.

2.3. Ultrasonic velocity measurements

The ultrasonic wave velocities were obtained applying the pulse-echo technique by measuring the elapsed time between the initiation and the receipt of the pulse, appearing on the screen of a flaw detector (USIP20 – Kraütkramer) via the standard electronic circuit (Hewlett Packard 54615 B). The velocity was therefore calculated by dividing the round-trip distance by the elapsed time according to the relation:

$$U = \frac{2X}{\Delta t}, \tag{2}$$

where X is the sample thickness and Δt is the time interval.

All ultrasonic wave velocity measurements were carried out at room temperature 298 K using the Karl Deutsch transducer S12 HB4 with fundamental frequency of 4 MHz to measure the longitudinal ultrasonic wave velocity, and the Kraütkramer transducer K4KY with fundamental frequency of 4 MHz for measuring the ultrasonic shear wave velocity. The estimated error in velocity measurements was $\pm 3 \times 10^2$ cm/s for longitudinal wave velocity U_ℓ and $\pm\, 2 \times 10^2$ cm/s for shear wave velocity (U_s).

3. Determination of elastic moduli

Elastic moduli (longitudinal L, shear G, bulk K, and Young's E) as well as Debye temperature θ_D, softening temperature T_s, Poisson's ratio σ and microhardness H of the ternary compounds $Bi_2(Te_{1-x}Se_x)_3$ (x = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0), have been determined from the measured ultrasonic wave velocities and density, using the relations [21]:

$$\begin{aligned} L &= \rho U_\ell^2, \\ G &= \rho U_s^2, \\ K &= L - \left(\frac{4G}{3}\right), \\ E &= (1+\sigma)^2 G, \\ \sigma &= \left(\frac{(L-2G)}{2(L-G)}\right), \\ H &= \frac{(1-2\sigma)E}{6(1+\sigma)}. \end{aligned} \tag{3}$$

The Debye temperature was obtained from the relation given as

$$\theta_D = \left(\frac{h}{k_B}\right)\left(\frac{3\psi N_A}{4\pi V_m}\right)^{1/3} U_m, \tag{4}$$

where h is Planck's constant, k_B is Boltzmann's constant, N_A is Avogadro's number, V_m is the molar volume calculated from the effective molecular weight and density (i.e. M/ρ), Ψ is the number of atoms in the chemical formula, and U_m is the mean ultrasonic velocity defined by the relation

$$U_m = \left(\frac{1}{3}\left(\frac{1}{U_\ell^3} + \frac{2}{U_s^3}\right)\right)^{-1/3}. \tag{5}$$

The transmission and reflection of sound energy in the sample was determined using the acoustic impedance Z_i given by the relation

$$Z_i = U_m \rho. \tag{6}$$

4. Results and discussions

As reported earlier [20], the composition analysis of the synthesized $Bi_2(Te_{1-x}Se_x)_3$ compounds with indicated x, confirms the presence of selenium, tellurium and bismuth in the prepared samples with the average atomic percentages which are illustrated in Table 1. The calculated contents of bismuth, tellurium and selenium wt % were comparable with wt % of the starting mixed materials, indicating that the synthesized materials are nearly stoichiometric for all the samples (Table 1).

Table 1. Chemical analysis data for $Bi_2(Te_{1-x}Se_x)_3$ ternary compounds.

X Mol fraction	Bi, at. %		Te, at. %		Se, at. %	
	Cal.	Exp.	Cal.	Exp.	Cal.	Exp.
0.00	40	40.77	60	59.23	0	0
0.2	40	40.93	48	49.23	12	9.84
0.4	40	38.22	36	36.64	24	25.14
0.6	40	37.52	24	28.34	36	34.14
0.8	40	36.76	12	13.87	48	49.37
1.0	40	39.97	0	0	60	60.03

Cal. – Calculated, Exp. – Experimental.

The density and molar volume of solid materials depend upon many factors such as structure, coordination number, cross-link density, and dimensionality of interstitial spaces [22].

Experimental values of density and molar volume are tabulated in Table 2 and Fig. 1. The density values were found to decrease from 10.441 g/cm^3 to 9.710 g/cm^3. In fact, this is due to the direct substitution of Se with atomic weight of 78.96 g/mol, on the expense of Te with atomic weight of 127.6 g/mol. On the other hand, the values of molar volume of the studied materials are reduced from 76.69×10^6 (cm^3/mol) to 67.44×10^6 (cm^3/mol). This decrease in the molar volume of the present materials may be attributed to the direct substitution of selenium with atomic radius of 1.15 Å on the expense of tellurium, with atomic radius of 1.4 Å [23]. However, the increase in packing density (V_t) [24, 25] from 0.037×10^6 to 0.339×10^6 (cm^3/mol) correlates with the decrease in molar volume, indicating the increased packing of atoms. Moreover, SOLIMAN *et al.* [20] reported that addition of Se on the expense of Te in the ternary compounds $Bi_2(Te_{1-x}Se_x)_3$, caused the decrease in the lattice parameters a and c

Table 2. Variation of density (ρ), molar volume (V_m), packing density (V_t), longitudinal wave velocity (U_l), shear wave velocity (U_s), with Se mol fraction.

X Mol fraction	ρ [g/cm^3]	$V_m \times 10^6$ [cm^3/mol]	$V_t \times 10^6$ [cm^3/mol]	$U_l \times 10^2$ [cm/s]	$U_s \times 10^2$ [cm/s]
0.00	10.441 ± 0.001	76.69 ± 0.15	0.037	366 ± 3	223 ± 2
0.2	10.183 ± 0.001	75.77 ± 0.15	0.090	405 ± 3	246 ± 2
0.4	10.050 ± 0.001	73.87 ± 0.15	0.147	420 ± 3	262 ± 2
0.6	10.119 ± 0.001	70.48 ± 0.15	0.211	558 ± 3	368 ± 2
0.8	9.753 ± 0.001	70.13 ± 0.15	0.269	658 ± 3	452 ± 2
1.0	9.710 ± 0.001	67.44 ± 0.15	0.339	777 ± 3	541 ± 2

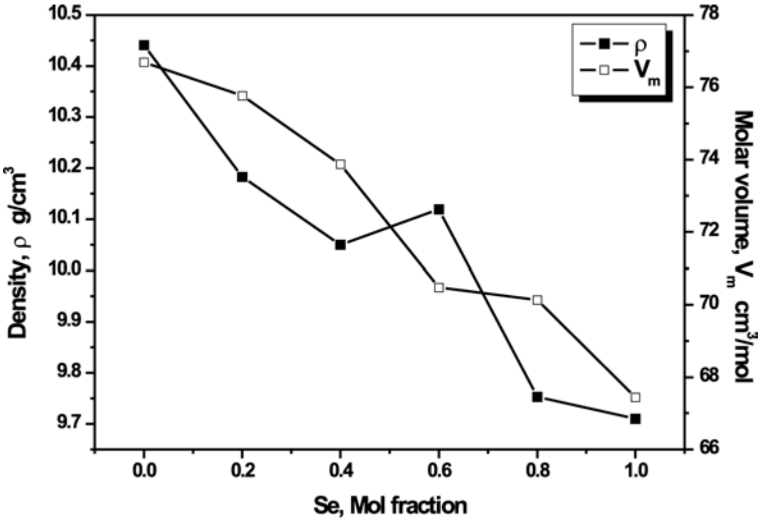

Fig. 1. Plot of boththe density and molar volume with x, Se mol fraction in $Bi_2(Te_{1-x}Se_x)_3$ compounds.

and unit cell volume, revealing that Se is incorporated into the lattice onto Te sites which agrees with molar volume in our results.

Values of ultrasonic wave velocities (longitudinal and shear), elastic moduli, and Poisson's ratio of the investigated glasses are also listed in Table 2. Both the longitudinal U_l, and shear U_s wave velocities increased from 366×10^2 and 223×10^2 (cm/s), respectively, to 777×10^2 and 541×10^2 (cm/s) with the increase of selenium on the expense of silver mol % content as shown in Table 2 and Fig. 2.

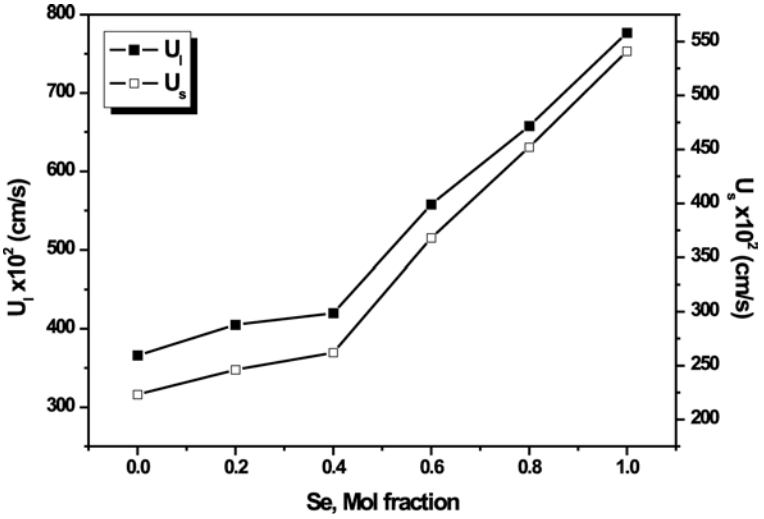

Fig. 2. Plot of ultrasonic wave velocities (longitudinal (U_l) and shear (U_s)) with x, Se mol fraction in $Bi_2(Te_{1-x}Se_x)_3$ compounds.

In general, the increase of ultrasonic wave velocity is related to the decrease in inter-atomic spacing of the material. This increase in ultrasonic wave velocities can be explained by the decrease in molar volume (i.e. the decrease in the inter-atomic spacing), which means that the incorporation of selenium instead of tellurium leads to the decrease in the average inter-atomic spacing (bond lengths and bond angles), supporting our results of molar volume and the results reported earlier [17, 20, 26–29] for $AgInTe_2$, $CuInTe_2$, $Bi_2(Te_{1-x}Se_x)_3$ and $Cu_xAg_{1-x}InTe_2$.

Table 3 gives the experimentally estimated values of the elastic moduli; Longitudinal modulus L, shear modulus G and Young's modulus E. As seen from Table 3, longitudinal, shear and Young's elastic moduli values showed an increase from 1.40, 0.52 and 1.250 GPa, respectively to 5.856, 2.84 and 5.846 GPa showing the same trend as ultrasonic wave velocities.

Table 3. Variation of longitudinal modulus (L), shear modulus (G), Young's modulus (E), Bulk modulus (K), Poisson's ratio (σ) and dissociation energy per unit volume (G_t), with Se mol fraction.

X Mol fraction	L [GPa]	G [GPa]	E [GPa]	K [GPa]	σ	G_t [kJ/cm^3]
0.00	1.401 ± 0.016	0.52 ± 0.01	1.250 ± 0.001	0.71 ± 0.02	0.207 ± 0.016	11.37
0.2	1.671 ± 0.016	0.62 ± 0.01	1.492 ± 0.001	0.85 ± 0.02	0.206 ± 0.016	12.29
0.4	1.772 ± 0.016	0.69 ± 0.01	1.632 ± 0.001	0.85 ± 0.02	0.179 ± 0.016	13.20
0.6	3.156 ± 0.016	1.37 ± 0.01	3.059 ± 0.001	1.33 ± 0.02	0.117 ± 0.016	14.11
0.8	4.221 ± 0.016	1.99 ± 0.01	4.195 ± 0.001	1.57 ± 0.02	0.054 ± 0.016	15.02
1.0	5.856 ± 0.016	2.84 ± 0.01	5.846 ± 0.001	2.07 ± 0.02	0.029 ± 0.016	15.93

Figure 3, shows the variation of bulk K modulus with molar volume of $Bi_2(Te_{1-x}Se_x)_3$ specimens under investigation. From this figure, it is noted that a decrease in volume should lead to an increase in bulk modulus with the increase in selenium content. It is clear from the above results that the type of bonding in the network structure plays a dominant role in deciding on the rigidity of these structures, and the increase in elastic moduli is associated with the increase in covalence of bonds in these structures [30], as the electro-negativity of selenium atoms (2.55) is higher than that of tellurium atoms (2.1). SOLIMAN *et al.* [20], reported that the structure of $Bi_2(Te_{1-x}Se_x)_3$ $(0 \leq x \leq 1)$ is a single phase polycrystalline material corresponding to the rhombohedral type structure, and the refined unit cell parameters and bond distances in $Bi_2(Te_{1-x}Se_x)_3$ unit cell were found to decrease with the increase in selenium content. This is maybe the reason for the increase in the rigidity of these structures. Moreover, the increased dissociation energy of bonds per unit volume G_t [24, 25] from 11.37 to 15.93 (kJ/mol) may contribute to the increased rigidity of these compounds (see Table 3). Furthermore, the increased values of micro-hardness from 0.101 to 0.891 GPa confirm the increased rigidity of these compound structures.

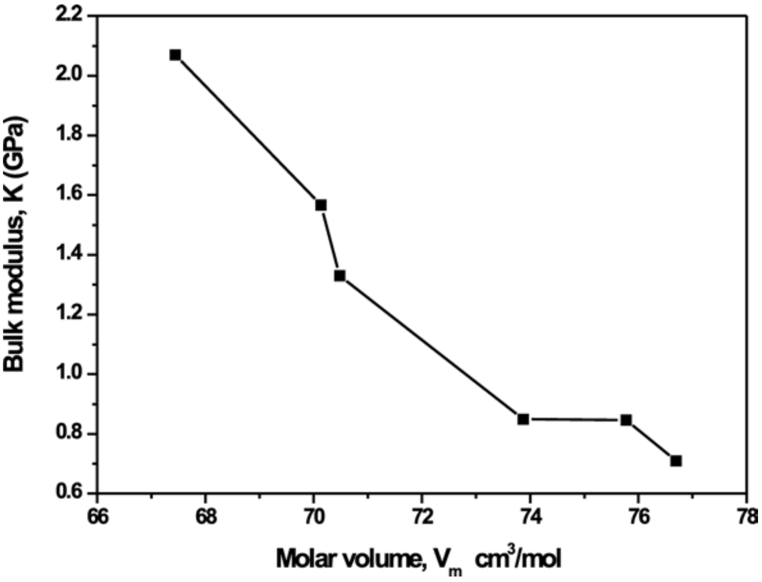

Fig. 3. Plot of bulk modulus (K) with x, Se mol fraction in $Bi_2(Te_{1-x}Se_x)_3$ compounds.

Table 3 gives the values of Poisson's ratio σ. Poisson's ratio is formally defined for any structure as the ratio of lateral to longitudinal strain, produced when tensile forces are applied. In solid materials, the tensile strain produced in the network is unaffected by the cross-links, while the lateral strain is greatly decreased with increasing covalence of bonds [31, 32]. The values of σ have decreased from 0.207 to 0.029 with increasing selenium content from 0 to 1. Therefore, the decrease in Poisson's ratio can be explained in terms of covalence bonds that have increased with selenium content [32].

Values of E/G ratio showed the same trend as Poisson's ratio σ, they have decreased from 2.4132 to 2.0589 when selenium mol fraction has increased from 0 to 1, confirming the decreased Poisson's ratio.

Debye temperature θ_D is an important parameter of solids, which represents the temperature at which nearly all the vibrational modes are excited. Calculated values of the mean ultrasonic velocity U_m, Debye temperature θ_D and acoustic impedance Z_i of these compounds under study, are collected in Table 4.

It can be observed from Table 4 that Debye temperatures increased from 24.9 to 62.1 K with the increase in selenium mol fraction from 0 to 1. The increase in the mean ultrasonic velocity and consequently, the increase in Debye temperatures [33], are mainly due to the decrease in molar volume and the increase in covalence of bonds.

The values of the acoustic impedance Z_i were found to increase from 25.69 to 57.06 (kg·m^{-1}sec^{-1}) when the selenium mol fraction increased from 0 to 1. The variation of the acoustic impedance resulting in higher impedance to the propagation of ultrasonic waves in these compounds [34].

Table 4. Variation of mean ultrasonic velocity (U_m), Debye temperatures (θ_D) and acoustic impedance (Z_i) with Se mol fraction.

X Mol fraction	$U_m \times 10^2$ [cm/s]	θ_D [K]	$Z_i \times 10^{-6}$ [kg·m^2/sec^{-1}]
0.00	246 ± 4	24.9 ± 0.8	25.69 ± 0.02
0.2	272 ± 4	27.7 ± 0.8	27.72 ± 0.02
0.4	289 ± 4	29.6 ± 0.8	29.05 ± 0.02
0.6	403 ± 4	41.9 ± 0.8	40.76 ± 0.02
0.8	492 ± 4	51.3 ± 0.8	47.98 ± 0.02
1.0	588 ± 4	62.1 ± 0.8	57.06 ± 0.02

5. Conclusions

Detailed measurements have clarified that the structure of $\mathrm{Bi_2(Te_{1-x}Se_x)_3}$ compounds becomes close-packed and more rigid with the substitution of Se instead of Te through the decrease in molar volume, increase in covalence bonds, the increase in ultrasonic velocities (longitudinal and shear) and consequently, the increase in elastic moduli.

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