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Pressure Dependent Elastic and Thermo-Physical Properties of U Ternaries with ZrNiAl Structure Compounds

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Abstract: The Thermophysical and ultrasonic aspects of U- ternaries are examined theoretically under high pressure. In order to determine the second- and third-order elastic constants of U- ternaries (URhAl and ThRhAl) compounds under pressure conditions (0-45GPa), the potential model technique has been used. The assessed second-order elastic constants are used along with the pressure-dependent ultrasonic velocities, thermal relaxation time, and other significant and important thermophysical data to determine the orientation. It is observed that thermal relaxation time and mechanical & thermal properties exhibit an increasing trend with increasing pressure for U-ternaries URhAl and ThRhAl compounds. The effect of increasing pressure on the URhAl and ThRhAl compounds shows up as an increase in elastic constants, ultrasonic velocities, ultrasonic attenuation, and Debye temperature. The ratio B/G increases with pressure which indicates that the materials are ductile in nature. We have used the approximate values of SOECs, density, and lattice properties in calculating the melting point, ultrasonic velocity, and thermal conductivity.

Keywords: U-ternaries compound; Elastic properties; Thermal conductivity; Ultrasonic properties

I. INTRODUCTION

Uranium compounds have a wide range of characteristics Because of the complicated behaviour of their 5f electrons. UTX (T, transition metal; X, non-transition metal) is a large class of uranium ternary equiatomic compounds. The ZrNiAl type, which is an ordered ternary derivative of the Fe_2 P type [1], is one of the most common structural types in which such compounds crystallized. The hexagonal structure is layered, with one form of basal-plane sheet holding all U atoms and some T atoms, and the other containing the remaining T and X atoms. The 5f-band states, which appear near the Fermi energy E_F, dominate the electronic structure of the UTX ternaries, as they do in most other uranium intermetallics. The transition metal (T) in d states has greater binding energies and contributes less to the electronic states at E_F [2]. In light actinides, the 5 f states have often been close to the boundary between localised and itinerant behaviour. Their sensitivity to external variables and their composition present themselves in a broad range of magnetic properties, which are often extremely strange. [3]. A hard-magnetization axis is equivalent to the chain direction whenever U atoms are coordinated in linear chains (TiNiSi or GaGeLi structure types from UTX compounds), with in-plane anisotropy perpendicular to the chain direction being weaker [3]. In this approach, magnetic anisotropy in U-based substances reflects bonding anisotropy. A large elastic anisotropy was also found in UTX compounds with the ZrNiAl structural type [4]. Many of these compounds are ferromagnetic, with a wide range of Curie temperatures and ordered moment magnitudes [5], and can thus be used to investigate discontinuous phase transitions with changeable magnetic order parameters but a constant crystal structure. The occurrence of discontinuous phase transitions in several of these compounds has recently been established (e.g. UCoAl [6, 7], URhAl, UCoG,a.) On either hand, two members of this group, UPtAl [6] and URhGa [8], showed an initial increase in TC with increasing pressure (in the range of several GPa). It is thus desirable to be able to anticipate how TC behaves under pressure in such compounds.

This study uses Lennard-Jones potential techniques to compute elastic, mechanical and ultrasonic characteristics. Ultrasonic attenuation (UA), which alludes to the specific association between the anisotropic behavior of the proximal hematinic planes and structural motion affinity, is the most important physical parameter used to explain the characteristics of the materials. There is a significant correlation between numerous physical properties, such as thermal energy density, specific heat, thermal conductivity and higher-order elastic coefficients with varying pressures. The information in this work will be helpful for future scientific investigations into the mechanical characteristics of URhAl and ThRhAl compounds as well as for comparing the elastic and ultrasonic properties of the URhAl and ThRhAl) compounds. Although ultrasonic is a non-destructive and practical method with a wide diversity of applications in every field. Ultrasonic characterization of these U- ternaries compounds has been a significant topic of focus for engineers and researchers

II. THEORY

One of the most popular theories for determining the higher order elastic coefficients of hexagonal wurtzite and HCP shaped compounds is the interaction potential model technique [9, 10]. The formulations of higher-order elastic constants were determined using the second- or third-order strain gradient of elastic energy density.

A common definition of elastic constants of certain value is the partial derivatives of the substance's thermodynamic potential applied to finite compression. The responsiveness of a material to external stress or the amount of stress required to maintain deformation is described by its elastic constant. The following is the description of the nth order elastic constant:

$$C_{ijklmn\ldots} = \frac{\partial^n F}{\partial\eta_{ij}\partial\eta_{kl}\partial\eta_{mn\ldots.}} \qquad [1]$$

The subsequent formula shows the substance's second (C_{IJ}) and third (C_{IJK}) order elastic values. Elastic coefficients (C_{IJ}) reflect the strain that a compound experiences when tension is applied in a certain direction or the stress required under a unique case.

$$C_{IJ} = \frac{\partial^2 U}{\partial e_I \partial e_J}; \qquad \text{I or J} = 1,\ldots\ldots 6 \qquad [2]$$

$$C_{IJK} = \frac{\partial^3 U}{\partial e_I \partial e_J \partial e_K}; \ \text{I , J and K} = 1,\ldots\ldots 6 \qquad [3]$$

Six second order coefficients (SOEC) and ten third order elastic coefficients (TOEC) are determined from, respectively, [9, 10] and Eqs. (2) and (3) apply to hexagonally close-packed structural materials. The T_m is associated with C_{11} and C_{33} in hexagonally shaped materials [11]. The melting point (T_m) has been calculated using the following formula

$$:T_m = 354 + 4.5(2C_{11} + C_{33})/3 \qquad [4]$$

The formula below has been used to determine the thermal conductivity (k_{min}) [12].

$$k_{min} = K_B V_D \left(\frac{N\,n\rho}{M}\right)^{\frac{2}{3}} \qquad [5]$$

Also a compound's macro hardness is denoted by H_V are described as [13]:

$$H_V = 2.0(\lambda^2 G)^{0.585} - 3 \qquad [6]$$

In a hexagonal compound, present are 3- different types of ultrasonic velocities: one longitudinal (V_L) and two shear (V_{S1}, V_{S2}) wave velocities, $V_L = \sqrt{\frac{C_{33}}{\rho}}$ and $V_S = \sqrt{\frac{C_{44}}{\rho}}$ where V_L is the longitudinal and V_{S1} and V_{S2} are shear and quasi-shear wave speeds, respectively [14,15] .

The equation gives the Debye average velocity (V_D) as follows [14, 15]:

$$V_D = \left[\frac{1}{3}\left(\frac{1}{V_L^3} + \frac{1}{V_{S1}^3} + \frac{1}{V_{S2}^3}\right)\right]^{-1/3} \qquad [7]$$

The V_m (average wave velocity) of the expression is as follows:

$$V_m = \left[\frac{1}{3}\left(\frac{2}{{V_s}^3 + {v_l}^3}\right)\right]^{-1/3} \qquad [8]$$

The mathematical formulation of UA (ultrasonic attenuation) is described as[16]:

$$\alpha_{long} = \frac{2\pi^2 f^2}{\rho V_l^3}\left(\frac{4}{3}\eta_e + \chi\right) \qquad [11]$$

$$\alpha_{shear} = \frac{2\pi^2 f^2}{\rho V_S^3}\eta_e \qquad [12]$$

Through a variety of causes, ultrasonic attenuation offers crucial micro-structural properties of the compounds by evaluating the ultrasonic wave's absorption in the medium. The following equation gives the ultrasonic attenuation coefficient $(\alpha)_{Akh}$ resultant from the phonon-phonon interaction process [17, 18].

$$(\alpha/f^2)_{Akh} = \frac{4\pi^2 \tau E_0\left(\frac{D}{3}\right)}{2\rho V^3} \qquad [13]$$

The thermal relaxation (τ) time, is as follows:

$$\tau = \frac{3k}{C_V V_D^2} = \tau_S = \tau_L/2 \qquad [14]$$

Here τ_L and τ_S are the thermal relaxation time for the longitudinal wave (V_L) and shear wave (V_S) respectively, the thermal conductivity is represented as ‘k’. The $(\alpha/f^2)_{Th}$ (thermoelastic loss) is evaluated through the subsequent equation [19]:

$$(\alpha/f^2)_{Th} = 4\pi^2 < \gamma_i^j >^2 \frac{kT}{2\rho V_L^5} \qquad [15]$$

The total ultrasonic attenuation $(\alpha/f^2)_{Total}$ is indicated through the following equation as [20]:

$$(\alpha/f^2)_{Total} = (\alpha/f^2)_{Th} + (\alpha/f^2)_L + (\alpha/f^2)_S \qquad [16]$$

Wherever $(\alpha/f^2)_{Th}$ is the thermoelastic loss, $(\alpha/f^2)_L$ and $(\alpha/f^2)_S$ are respectively the ultrasonic attenuation coefficients for the longitudinal wave (V_L) and shear wave (V_S).

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III. RESULTS AND DISCUSSION

A. *Higher Order Elastic Constants*

Six SOECs and ten TOECs, the elastic coefficients, were calculated in this work using the interaction potential approach. The lattice parameters 'a' (basal plane parameter), 'p' (axial ratio), and Density (: x 103 kg m-3) for the URhAl and ThRhAl compounds are 7.00, 0.554, and 4.904, respectively [21]. The advised values of m and n for compounds containing URhAl and ThRhAl are 6 and 7, respectively. b_o= 1.13x10^{-62} erg cm^7 for the substances URhAl and ThRhAl.

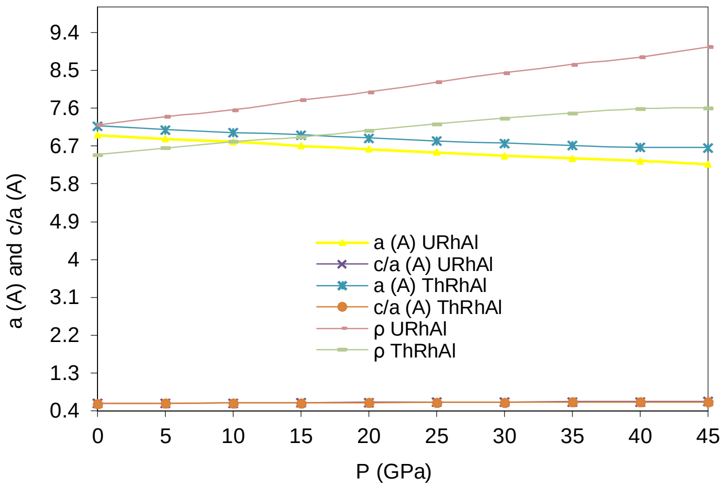

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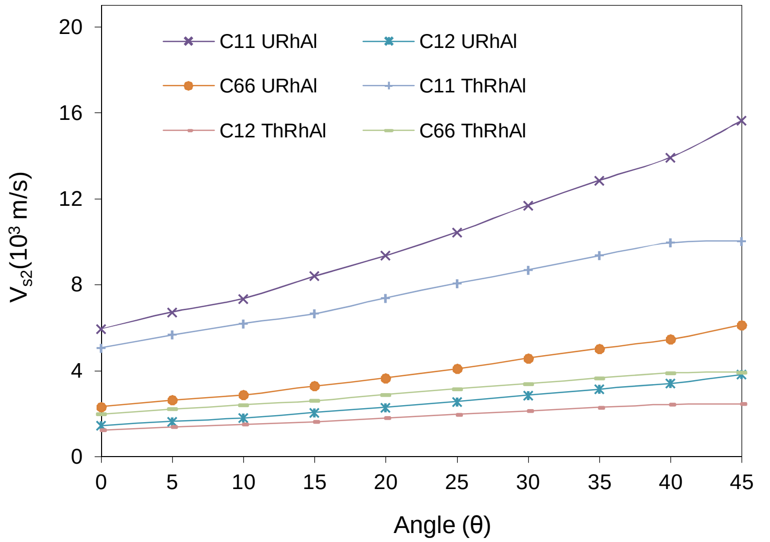
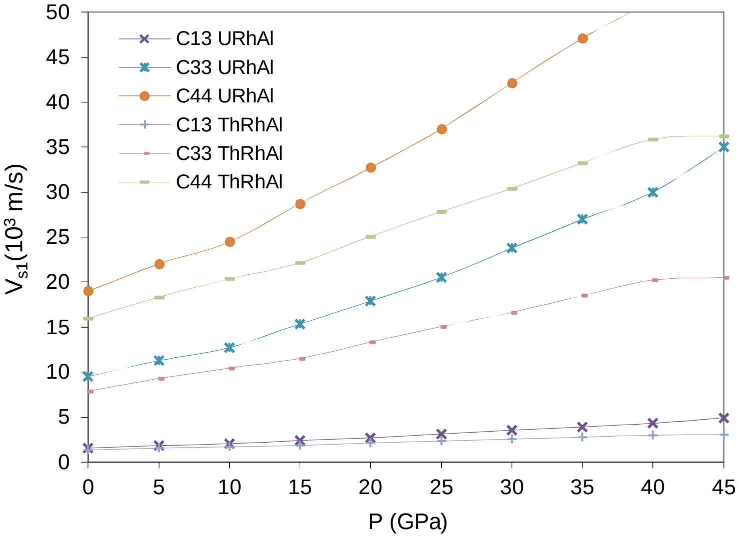
Fig.1. Pressure dependent Lattice parameter (a and c/a Å) and Density (ρ)



(a)



(b)

Fig:2(a,b) Pressure dependent second order elastic cofficents

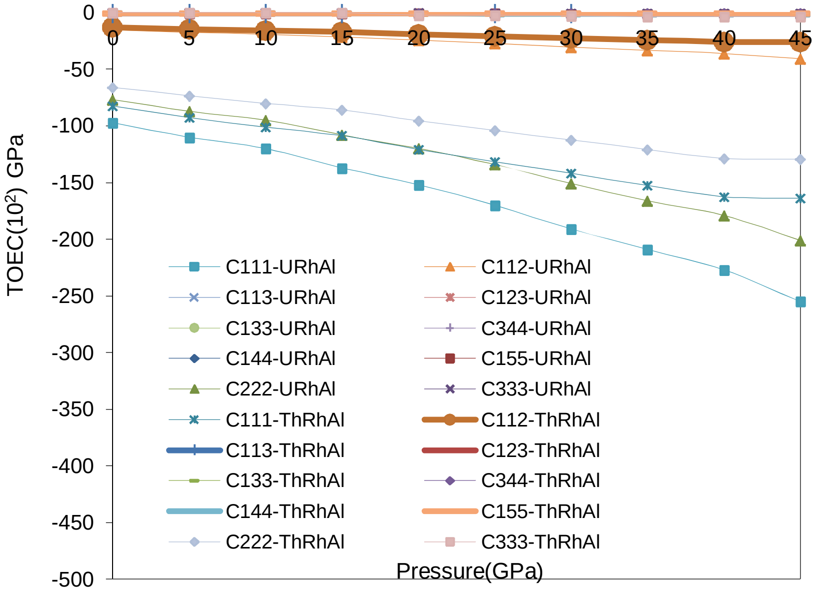


Figure: 2. Pressure dependent third order elastic coefficients

Its elastic constant describes how the compound behaves to strain or the amount of tension required maintaining a deformation in place. The elastic constant is an important variable that contributes crucial elements to mechanical and dynamic behaviors. Under pressure, we examine the elastic coefficients of URhAl and ThRhAl compounds (0–45GPa).

Figures 1 and 2 illustrate the pressure-dependent SOECs and TOECs that were determined for this chemical (a,b). In fact, the stiffness matrix's various hexagonal phases' independent elastic coefficients were C_{11}, C_{33}, C_{44}, C_{66}, C_{12}, and C_{13}. Figure 1 depicts the correlation between pressure and the second order elastic coefficients of the compounds URhAl and ThRhAl. It has been found that as pressure increases, the elastic constants of the URhAl and ThRhAl compounds increase monotonically. C_{11} and C_{33} change suddenly as pressure rises, while C_{13}, C_{12}, and C_{44} behave reasonably. Furthermore, we have determined the C_{33} of URhAl and ThRhAl compounds at 347 GPa, which is the same as the 347.5GPa estimated at 0-GPa by M. Hamici et al. [22].

According to table 1, the third order constants increase as pressure increases. The computed TOEC values are shown to be negative, indicating that the solid is under strain. As a result, all higher order elastic properties are acceptable [23].

The pressure-dependent URhAl and ThRhAl compounds [24] satisfy the following Born elastic stability criteria for hcp compounds:

$$\begin{bmatrix} C_{11} > |C_{12}| \\ C_{11}\,C_{33} + C_{12}\,C_{33} > 2C_{13}{}^{2} \\ C_{11} > 0 \\ C_{33} > 0 \\ C_{44} > 0 \end{bmatrix} \qquad [20]$$

As their evaluated elastic coefficients, C_{jj}, fulfil the criteria, it is expected that URhAl and ThRhAl compounds are mechanically stable at such dependent pressures. Bulk, shear, and Young's modulus (B, G, and Y) can all have an influence on a compound's physical characteristics [25, 26].Because of its obvious connection to the cohesive energy or bond formation energy of atoms in crystals, bulk modulus can be used to explain the average atomic bond strength. Compound hardness is significantly influenced by the G. The stiffness of a solid, Y, is computed as a function of the strength of its atomic bonds. From [27, 28], it is possible to calculate the B and the G for the compounds with the hexagonal URhAl and ThRhAl.

$$\left.\begin{array}{l} M = C_{11} + C_{12} + 2C_{33} - 4C_{13},\mathrm{C}^2 = (C_{11} + C_{12})C_{33} - 4C_{13} + C^2{}_{13}; \\ B_R = \frac{C^2}{M}; \mathrm{B}_V = \frac{2(C_{11}+C_{12})+4C_{13}+C_{33}}{9}; \\ G_V = \frac{M+12(C_{44}+C_{66})}{30}; \mathrm{G}_R = \frac{5C^2C_{44}C_{66}}{2[3B_VC_{44}C_{66}+C^2(C_{44}+C_{66})]}; \\ Y = \frac{9GB}{G+3B}; \quad \mathrm{B} = \frac{B_V+B_R}{2}; \quad \mathrm{G} = \frac{G_V+G_R}{2}; \sigma = \frac{3\mathrm{B}-2\mathrm{G}}{2(3\mathrm{B}+G)} \end{array}\right\} \qquad [21]$$

The evaluated results are shown in fig. 2.

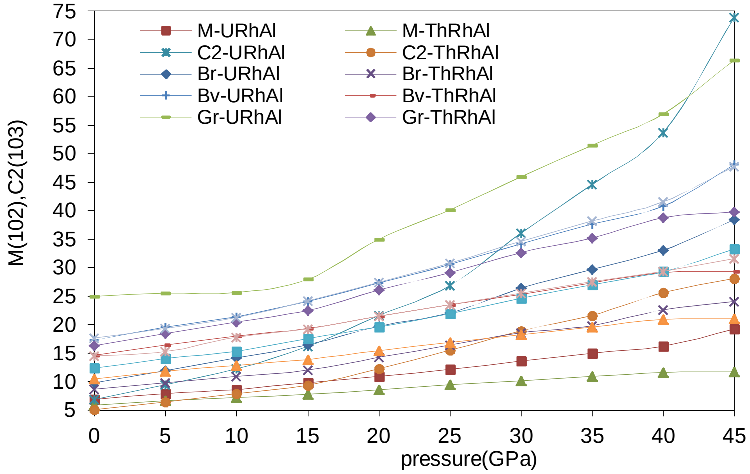


Fig: 3 Pressure dependent Voigt–Reus' constants (M and C^2), B (x 10^{10}Nm^{-2}), G (x 10^{10}Nm^{-2}), Y (x 10^{10}Nm^{-2}), σ, B/G for URhAl and ThRhAl.

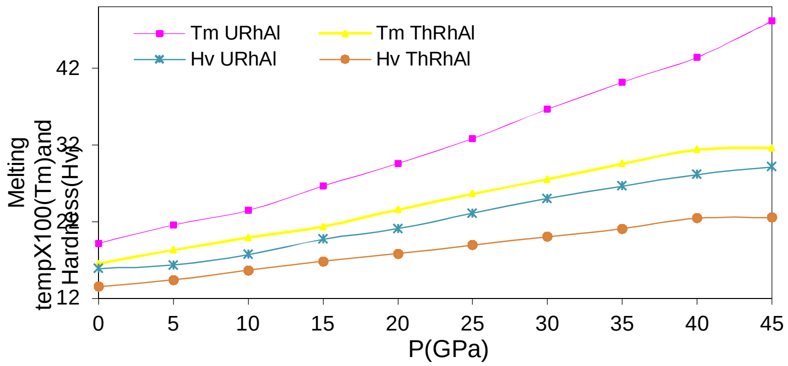


Fig:4 Pressure dependent Hardness(H_v),and Melting temperature (T_m) for URhAl and ThRhAl.

Table 2 shows that in the pressure range of 0 to 50 GPa, the B is much bigger than the G, showing that the resistance of URhAl and ThRhAl compounds to volume change is greater than that of shape change. This is supported by the hardness in Fig. 2(b). The material is brittle if the B/G ratio is higher than 1.75; otherwise, it is ductile. Indicating that the URhAl and ThRhAl compounds are ductile under high pressure, the value of B/G rises from 1.552 to 1.554 as the pressure is increased from 0 to 50 GPa. The stiffness of the URhAl and ThRhAl compounds improves as Young's modulus increases, as illustrated in Fig 2(a) [29]. Table 2 shows that the B increases significantly with increasing pressure, implying that increasing pressure can better the hardness of the compounds [30]. The hexagonal crystal's melting temperature Tm can be calculated using equation 7 [31]. Figure 3 illustrates the estimated melting points for URhAl and ThRhAl compounds at pressure conditions (c). The pressure increase is evidently caused by the gradual augmentation of the URhAl and ThRhAl compounds of Tm, as seen in Fig. 3(c).

B. Ultrasonic Velocity And Allied Parameters

In this analysis, the mechanical and isotropic properties of the compounds URhAl and ThRhAl have been associated to ultrasonic velocity. V_L, V_S, V_D, and were estimated for the URhAl and ThRhAl. Table.3 depicts the values of the pressure-dependent acoustic coupling constants as well as the thermal energy density (E0), which were calculated using the physical constant and Debye temperatures (D_L and D_S).

Table.3 Thermal conductivity ($K_{(min)}$Erg/cm-sec-K) and acoustic coupling constant ($D_{L,}$ D_S) of U-ternaries compounds (URhAl and ThRhAl) in table.3 at different pressure.

Pressure(GPa)	D_L(URhAl)	D_S(URhAl)	D_L(ThRhAl)	D_S(ThRhAl)	K_{min}(URhAl)	K_{min}(ThRhAl)
0	39.937	210.8	36.612	179.60	0.3206	0.2905
15	33.593	144.45	35.17	166.56	0.4074	0.3507
30	30.301	111.74	30.89	128.56	0.506	0.4210
45	45.41	229.59	28.48	110.089	0.6137	0.4661

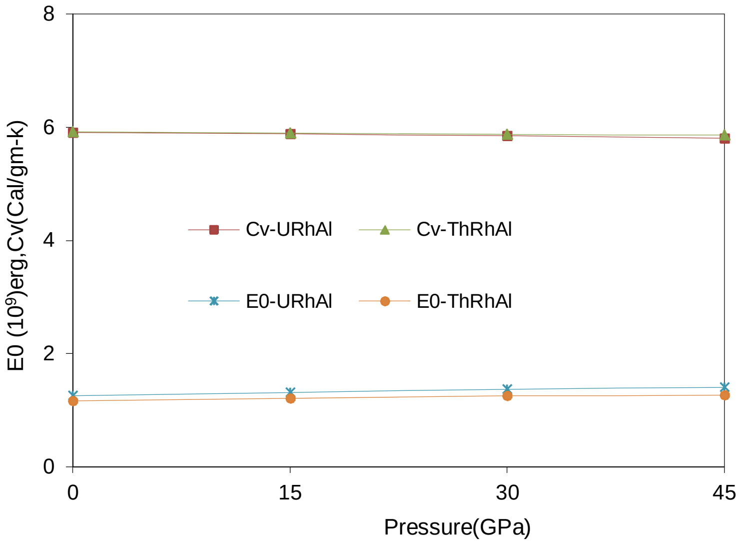


Fig: 4 E_o, C_v vs Pressure

Debye temperature (Θ) is an interesting and promising characteristic parameter that is essential to take into consideration while examining concepts in the field of solid-state physics. The melting point, thermal conductivity, and specific heat are corresponding. The correlation between the Debye temperature and a variety of materials' physical properties, such as melting temperature and specific heat, is considerable. Using elastic constants, the Debye temperature can be calculated [32].

The "k_{min}" values that were estimated using Eq. 12 at various pressures (0–50GPa) are shown in Fig. 3. (a). It is evident from Fig. 3(a) that as pressure increases, k_{min} of the compounds URhAl and ThRhAl gradually increase.

Table.4 demonstrates conclusively that the values of DL for URhAl and ThRhAl compounds at various pressures are even much higher than DS. This illustrates that shear ultrasonic waves exchange ultrasonic energy into heat energy more slowly than longitudinal ultrasonic waves. One of the most crucial physical properties for condensed substances to be considered when analysing the physical mechanisms underlying heat conduction is their thermal conductivity, which further offers comprehensive data on lattice vibrations in solids [33].

Figures 4–7 show the ultrasonic velocities along of the crystal's z-axis at (0-50 GP) pressures, with the URhAl and ThRhAl compounds' velocity VL increasing with angle. (Fig.4), and in Figure 5, the velocity VS1 is at its minimum at an angle of 300 and its greatest at 450. Figure 6 shows how VS2 velocities increase with increasing angle. The peculiar behaviour of direction dependent velocity is also caused by the presence of SOECs and density. The alignment dependent velocity curves obtained in this study are comparable to the direction dependent velocity curves seen in other U-ternary compounds [34, 35]. Therefore, the URhAl and ThRhAl compounds including an angle-dependent hexagonal structure are completely justified. Figure.9 depicts the fluctuation of Debye average velocity (VD) as a function of angle () with the crystal's z-axis. VD increases with angle () and reaches its maximum at 550 because to the inclusion of the velocities VL, VS1, and VS2 in the estimation of VD for the URhAl & ThRhAl compounds [36, 37]. It appears to be assume that changes in debye average velocity are effected by necessary ultrasonic velocities. A significant increase in all wave velocities results in a maximum value of VD at 55°. The z-axis of this crystal rotates at an angle of 55^0 degrees, as well as the average sound wave velocity confirms that this is the maximum

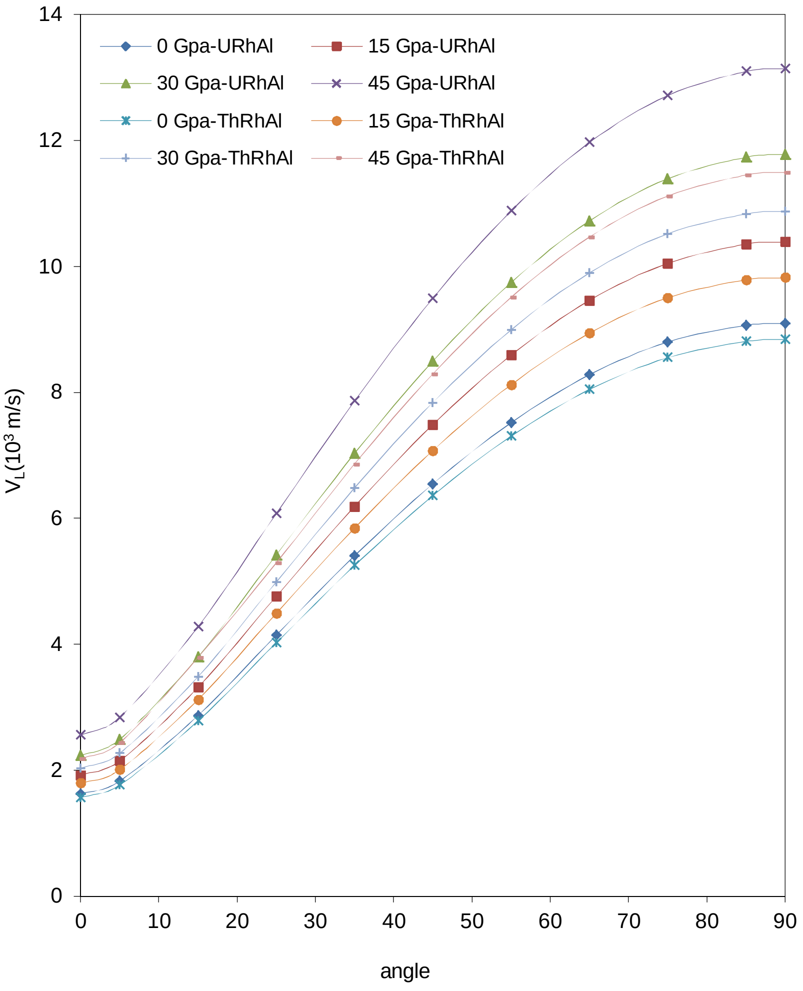


Fig.5. Angle dependent V_L

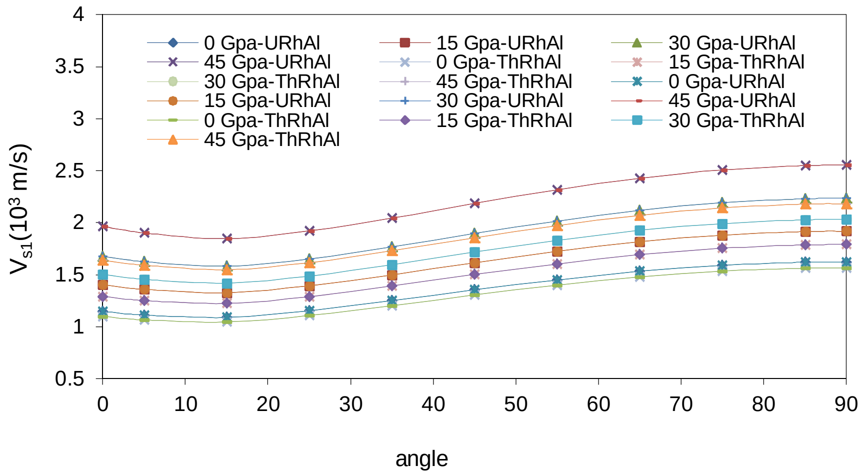


Fig 6. Angle dependentVs$_1$

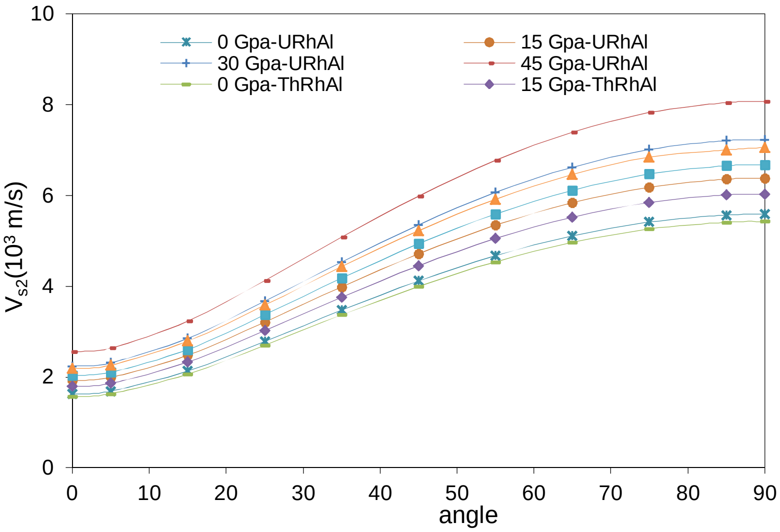


Fig.7. Angle dependent Vs$_2$

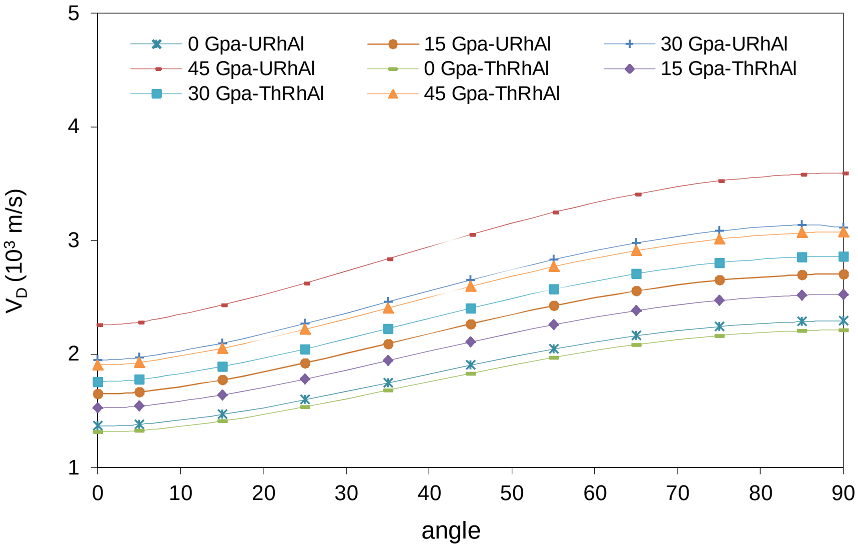


Fig.8. Angle dependent V$_D$

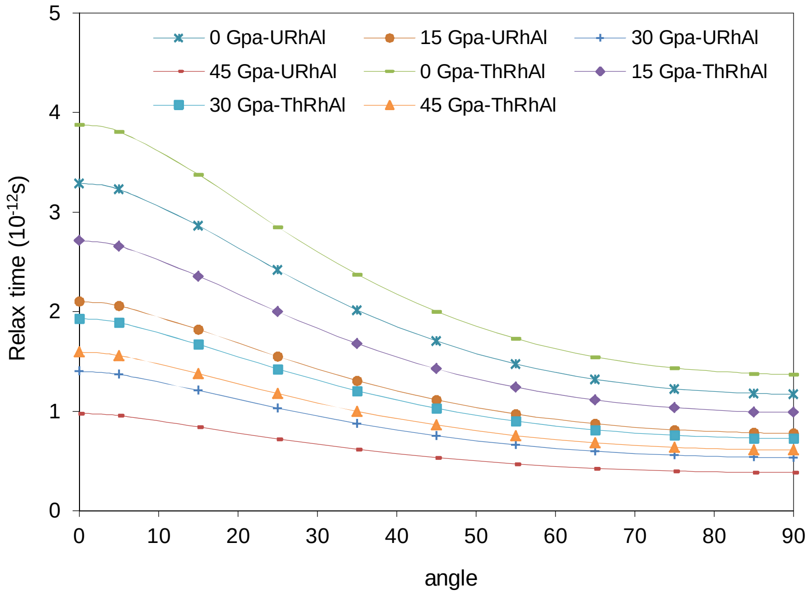


Fig.9. Angle dependent Relaxation time

An alignment-dependent plot of the determined thermal relaxation time is shown in Figure 10. The evaluation of V_D as having a $\tau \propto 3k/C_V V_D^2$reciprocal nature is accomplished using angle dependant curves. The thermal relaxation times of the compounds URhAl and ThRhAl were in the picosecond range [38, 39]. "k_{min}" obviously had an impact on the thermal relaxation times of the compounds URhAl and ThRhAl. Ultrasonic attenuation is caused by p-p interaction and thermal relaxation, as shown by the shortest s actually time used to calculate the thermal phonon equilibrium distribution for wave propagation along = 55^0.

C. Ultrasonic Attenuation Due To Phonon-Phonon Interaction And Thermal Relaxation Phenomena

Evaluating the UA is based on the assumption that the wave is propagating along the z-axis of the URhAl and ThRhAl compounds. Equation 15 is utilized to determine, at different pressures, the attenuation coefficient divided by frequency squared $(\alpha/f^2)_{Akh}$for the longitudinal wave $(\alpha/f^2)_L$ and the shear wave $(\alpha/f^2)_s$ under ωτ<<1 condition. The thermoelastic loss is divided by frequency squared$(\alpha/f^2)_{Th}$ using Equation.17 [40]. Figure 8(a,b) depicts the results for the pressure-dependent longitudinal, shear, and total attenuations of URhAl and ThRhAl compounds.

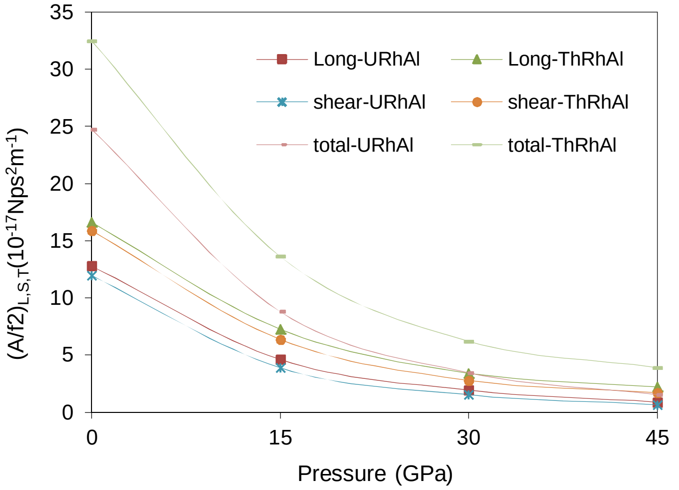


Fig.10. Pressure dependent Long.& Shear attenuation of URhAl and ThRhAl

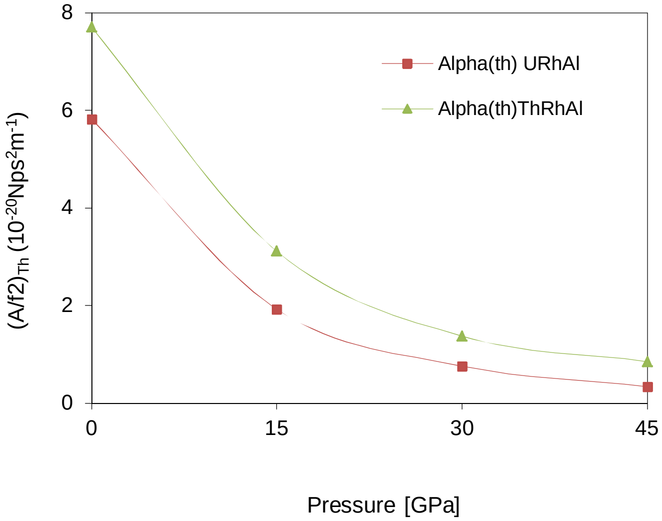


Fig.11. Th. attenuation vs temperature of URhAl and ThRhAl

The z-axis of the crystal is assumed to be the direction travelled by the ultrasonic wave in this analysis. It is evident that the energy losses are proportional to D, E_0, V^{-3} and $(\alpha/f^2)_{Akh}$ (Equations 17 and 18). Figure 11 demonstrates that "E_0" and "V" increase with pressure (0-50GPa).Therefore, in the URhAl and ThRhAl compounds, E_0 and k significantly affect Akhieser losses. As a consequence, the decrease in thermal conductivity is causes the increase in UA. Due to this, the p-p interaction is what affects ultrasonic attenuation. As a result, correlations of UA could not be performed out because there were no theoretical or experimental data that existed in the literature [38, 39].

Figures.8(a,b) indicate that for URhAl and ThRhAl compounds, thermo-elastic loss is much lower than Akhieser loss, with UA related with p-p interactions for longitudinal and shear waves being the major factor. The two most significant factors affecting total attenuation are thermal conductivity and thermal energy density. Although being the least brittle, URhAl and ThRhAl compounds behave in its purest form at greater pressures and they are more ductile, as shown by the minimal attenuation. The URhAl and ThRhAl compounds will therefore have the minimum impurities at ambient pressures.

IV. CONCLUSIONS

In the current investigation, the Lennard-Jones potential approach was used to evaluate the elastic, mechanical, and thermophysical aspects of the URhAl and ThRhAl compounds at pressures ranging from 0 to 50 GPa. The analysis yields the following finding:

1) The elastic constant, which plays a vital role in establishing essential points of mechanical and dynamic behaviour. The higher order elastic constants in the URhAl and ThRhAl compounds verify our theory. The URhAl and ThRhAl compounds' assessed elastic coefficients fulfil the criteria, indicating that they are mechanically stable under the pressures applied.
2) Furthermore, reports have been made on the B, G, Y (Young's modulus), Poisson's ratio (σ), and Hardness of URhAl and ThRhAl compounds under various pressures (Hv). In particularly, the estimated B/G values show that URhAl and ThRhAl compounds are brittle in the pressure range of 0–50 GPa. Applying increasing pressure can made it more ductile since it becomes more brittle the more pressure is applied. URhAl exhibit better mechanical response with increasing pressure in comparison to ThRhAl.
3) Moreover, data has been provided regarding the melting point, thermal conductivity, and specific heat capacity of URhAl and ThRhAl compounds under various pressures. As pressure is increased, the thermal conductivity (kmin) of the compounds of URhAl and ThRhAl gradually increases. The pressure which causes the melting point of the compounds of URhAl and ThRhAl to gradually increase.
4) The "τ" value for URhAl and ThRhAl compounds is found to be in the order of picoseconds, supporting their hexagonal shape. The period for the re-establishment of symmetry propagating of phonons will be the shortest for wave propagation in this direction since " has the least value along = 55^0 at all pressures.

5) The most important element in determining thermal conductivity is UA, which governs total attenuation and is carried on by the p-p interactions. URhAl and ThRhAl compounds exhibit their best performance with higher pressure, and their increased ductility is supported by the minimum UA under higher pressure.
6) URhAl exhibit better response with increasing pressure in comparison to ThRhAl.

The research could help with laves phase URhAl and ThRhAl compounds processing and non-destructive characterization. The current study is expected to contribute significantly to a comprehensive understanding of U-ternary compounds and provide guidance for such applications.

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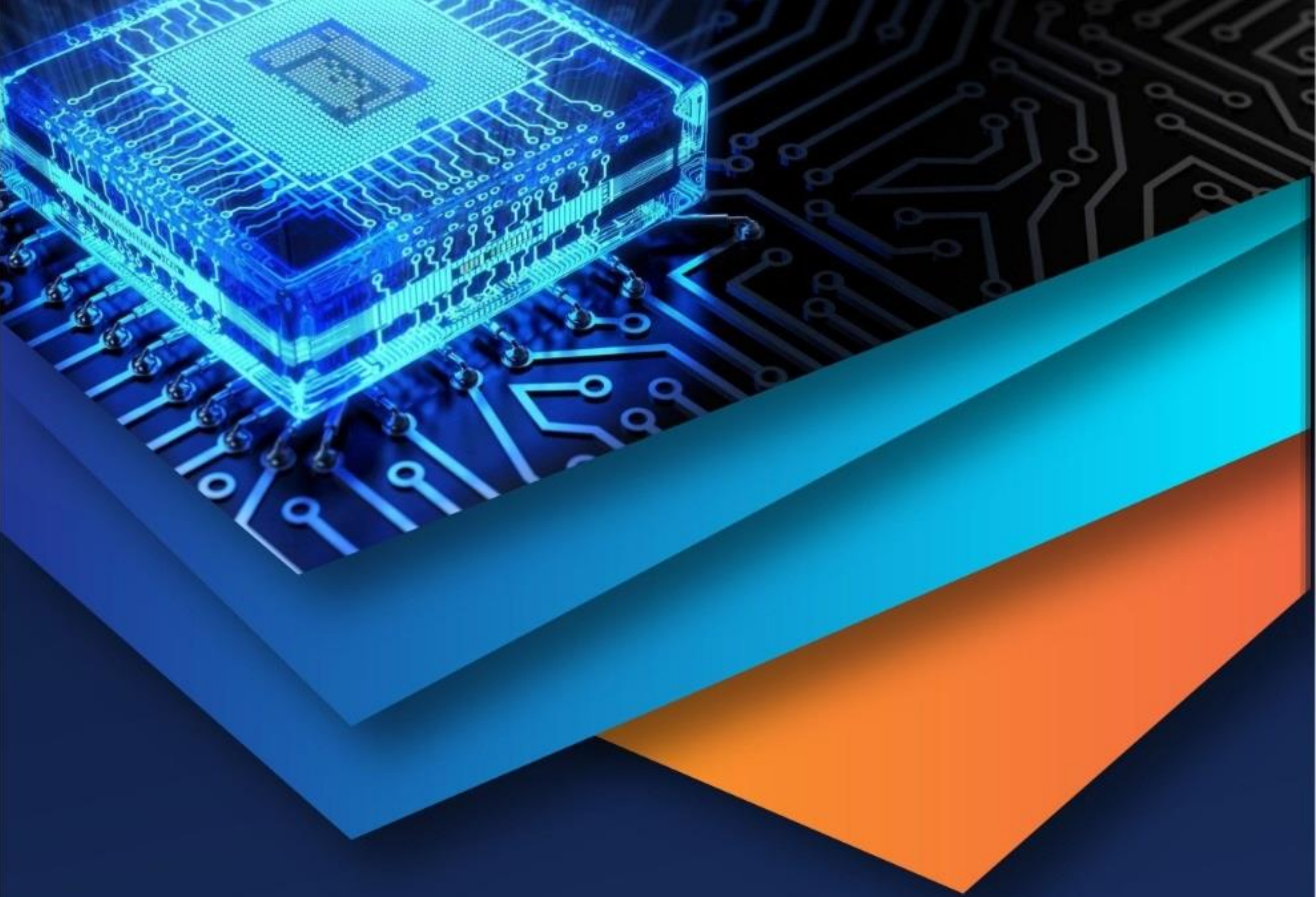

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