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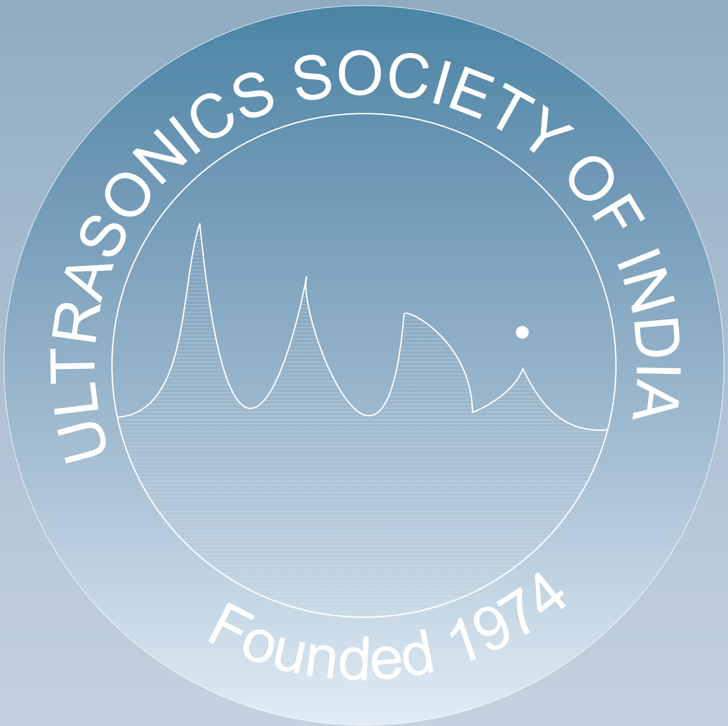

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Investigation of temperature dependent mechanical, thermophysical and ultrasonic properties of ScZrHf ternary alloy

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In this paper, we present theoretically evaluated values of temperature mechanical, thermophysical and ultrasonic properties of hexagonal close-packed structured medium entropy alloy ScZrHf in temperature range of 0-900 K. By utilizing the Lennard-Jones potential model, we have computed the second order and third order elastic constants (SOECs and TOECs) with the help of lattice parameters. While all of the SOECs have been found to be decreasing with increase in temperature, the TOECs increases with temperature. SOECs and TOECs have been used to compute the elastic moduli such as: bulk modulus, shear modulus, Young's modulus and Poisson's ratio, and ultrasonic velocities at different angle along unique axis. Further, the thermal properties such as Debye temperature, Debye heat capacity, energy density of ScZrHf in temperature range of 0-900 K and lattice thermal conductivity of ScZrHf in temperature range of 300-900K have been estimated. The lattice thermal conductivity decreases with increase in temperature. Finally, the ultrasonic attenuation due to phonon - phonon interaction in both longitudinal and shear modes and themoelastic relaxation mechanism have been computed for ScZrHf ternary alloy in the temperature range of 300-900 K and it has been found that the attenuation due to phonon-phonon interaction is much higher than that due to thermoelastic relaxation mechanism.

Keywords: Refractory medium-entropy alloys, hexagonal closed-packed, ultrasonic behaviour, rare-earth, transition metal.

Introduction

The high entropy alloys (HEAs) have attracted remarkable attention in recent decades due to being a new design concept of alloy materials[1]. HEAs are defined as a solid solution of five or more metal elements in single phase mixed in equal proportion. These alloys are assumed to be stabilized by a high configurational entropy of mixing[2]. Alloys with three or four elements are considered as medium entropy alloys[3]. The refractory high-entropy alloys (RHEAs) are composed of refractory metals such as Ti, Zr, Hf, Nb, W, Mo and Ta[4]. These RHEAs have been extensively studied due to their excellent mechanical and thermal properties and widespread applications[5,6].

Zr and Hf are two of the excellent refractory metals which shows extraordinary mechanical and thermal properties such as high tensile strength and resistance to wear and tear and high temperature, Sc is the lightest transition metal with good thermal conductivity and tensile strength. Considering the individual properties of these metals, ScZrHf becomes a potential candidate for widespread applications where high tensile strength and excellent resistant to high temperature is required. Hexagonal close-packed Ti, Zr, Hf based RHEAs are being synthesized for industrial applications. Therefore, ScZrHf is a promising alloy for high pressure and high temperature applications.

Huang *et al.*[6] have studied effect of Sc and Y addition on properties of HCP structured TiZrHf alloy and found improved the strength and ductility in TiZrHfSc alloy as compared to the TiZrHf alloy. Another study by Huang *et al.*[7] have examined the thermoelastic behaviour of ScTiZr, ScTiHf, ScZrHf, and ScTiZrHf and evaluated a number of properties of these alloys.

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There have been a few other experimental and theoretical studies focused on HCP structured high entropy alloys for their mechanical and thermal properties[8-11].

Despite these studies, temperature dependent second order elastic constants (SOECs, C^{IJ}) third order elastic constants (TOECs, C^{IJK}), thermal properties such as Debye temperature, heat capacity, thermal energy density, thermal conductivity and ultrasonic properties such as ultrasonic velocities and attenuation for HCP structured ScZrHf alloy are yet to be investigated.

Therefore, this paper presents the determination of temperature dependent SOECs, TOECs, elastic moduli, Poisson's ratio, ultrasonic velocities in different modes of vibration along different directions in the alloy crystal, Debye average velocity, Debye temperature, thermal energy density, heat capacity and ultrasonic attenuation for HCP structured ScZrHf alloy utilizing a theoretical approach.

Theory

Lennard-Jones potential method has been utilized to compute the SOECs and TOECs at different temperature for ScZrHf. The lattice parameters for computing SOECs and TOECs were found in the literature[7]. The formulation for calculating the six independent SOECs and ten independent TOECs have been taken from literature[12-14]. The bulk modulus *(B)*, shear modulus *(G)*, Young's modulus *(Y)* and Poisson's ratio have been computed by Voigt-Reuss-Hill method[15,16] for hexagonal crystals.

The ultrasonic velocities are important parameters while estimating the mechanical properties of materials, and for computing the ultrasonic velocities in longitudinal (V_L), quasi-shear (V_{S1}) and shear (V_{S2}) modes along different angles with unique axis (c-axis) of HCP crystal, Debye average velocity (V_D) and Debye temperature (θ_D) we have use formulation from literature[8,12-14].

As the medium-entropy alloys are potential materials for application in high pressure and temperature conditions, it becomes crucial to study thermal properties such as heat capacity, thermal energy density and thermal conductivity at different temperature. The heat capacity (C_V) and thermal energy density (E_0) have been evaluated using the Debye model for heat capacity[12,17,18].

Morelli and Slack[19] have described theoretical formulation for computing the lattice thermal conductivity κ, which is given by equation (1).

$$\kappa = A\frac{M_a {\theta_D}^s \delta}{\gamma^2 T_n^{2/s}} \tag{1}$$

Where *A* is a proportionality constant (with very slightly dependence on γ), δ (in Å) is the cube root of volume per atom, *n* is the number of atoms per unit cell, M_a (in amu) is average atomic mass, *T* is the temperature (in *K*), γ is Grüneisen constant which can be calculated by $\gamma = \frac{\alpha B}{C_V \rho}$ where α is the volume thermal expansion coefficient, *B* is bulk modulus, C_V is heat capacity and ρ is density of the material.

The mechanical properties such as elastic moduli, mechanical stability, thermal conductivity, heat capacity of solid and liquids is directly correlated to the ultrasonic attenuation. The major causes for the ultrasonic attenuation in materials at high temperature are phonon-phonon interaction (Akhiezer loss)[14,20] and loss due to thermo-elastic relaxation mechanism[8,21]. The formulation for evaluating the ultrasonic absorption coefficient (α) over frequency (f) squared (α/f^2) due to phonon-phonon interaction for longitudinal and shear modes in terms of the acoustic coupling constant *(D)* was developed by Mason and Bateman[22] and is given as follows:

$$(\alpha/f^2) = \frac{4\pi^2 \tau_{th} E_0 D}{2\rho V^3} \tag{2}$$

$$D = 9\left\langle (\gamma_i^j)^2 \right\rangle - \frac{3\left\langle \gamma_i^j \right\rangle 2 C_V T}{E_0} \# \tag{3}$$

Where *V* is ultrasonic velocity in longitudinal and shear modes, $\left\langle (\gamma_i^j)^2 \right\rangle$ and $\left\langle \gamma_i^j \right\rangle^2$ are square average and average square Grüneisen numbers, respectively for longitudinal and shear modes and $\tau_{th} = \frac{3\kappa}{C_V V_D^2}$ is thermal relaxation time.

The ultrasonic attenuation due to thermoelastic relaxation mechanism have been computed by:

$$\left(\frac{\alpha}{f^2}\right)_{th} = \frac{4\pi^2 \left\langle \gamma_i^j \right\rangle^2 \kappa T}{2\rho V_L^S} \tag{4}$$

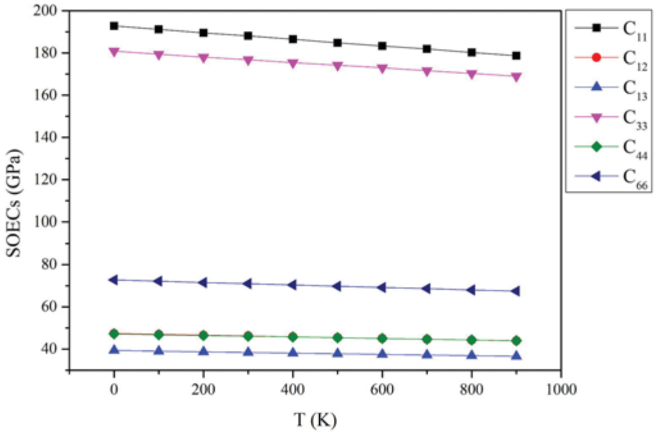

Fig. 1. Temperature (T) dependence of SOECs of ScZrHf

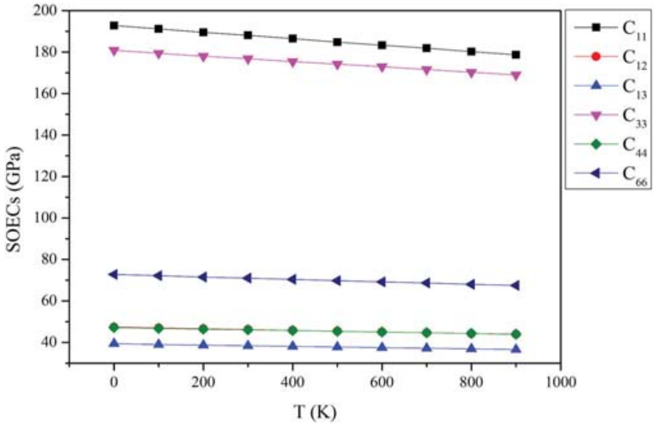

Fig. 2. Temperature (T) dependence of TOECs of ScZrHf

Results and Discussion

The computed values of SOECs and TOECs for ScZrHf at different temperatures in range of 0-900 K are presented in Fig. 1 and Fig. 2.

It is clear from Figs. 1 and 2 that the SOECs show a decrease with increase in temperature while TOECs exhibit opposite behaviour than that of SOECs *i.e.*, an increase with increase in temperature. The SOECs also follow the mechanical stability criteria[23] which is given as $C_{44}>0$, $C_{11}>|C_{12}|$, $(C_{11}+2C_{12})>2C_{13}$ for hexagonal structured crystals. This confirms that the ScZrHf alloy maintains a high mechanical stability over the temperature range 0-900 K.

The temperature dependence of bulk modulus *(B)*, shear modulus *(G)* and Young's modulus *(Y)* are presented in Fig. 3. It is evident from the Fig. 3 that all of the elastic moduli *i.e., B, G* and *Y* decrease with increase in temperature. The values of *B*, *G* and *Y* at 300 K are found to be 80.27 GPa, 51.35 GPa and 126.99 GPa respectively which are in good agreement with the value for similar materials available in literature[7,11]. The Poisson's ratio is found to be varying from 0.2364 to 0.2362 which is comparable to the Poisson's ratio of ScZrHf and similar alloys available in literature[7,8,11,24].

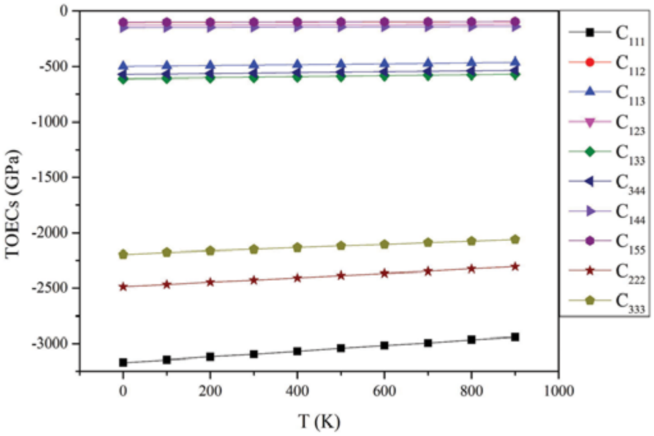

Fig. 3. Temperature (T) dependence of elastic moduli: bulk (B), shear (G) and Young's (Y) modulus of ScZrHf

The longitudinal *(V_L)* quasi-shear *(V_{S1})*, shear *(V_{S2})*, and Debye average *(V_D)* velocities at different angles with the unique axis in the temperature range 0-900 K have been evaluated using the SOECs and density of the alloy and are plotted in Fig. 4.

The longitudinal wave velocity V_L decreases with increase in temperature but decreases with increase in angle θ up to 45° with the unique axis and start increasing again from 45-90°. The maximum value of V_L is 5.10×10^3 ms^{-1} at angle 90° with the unique axis at temperature 0 K. The quasi-shear wave velocity V_{S1} also decreases with temperature but have a maximum value of 3.15×10^3 ms^{-1} at angle 45°. The shear velocity V_{S2} monotonically decreases with temperature while increases with angle θ. The Debye average velocity V_D shows similar nature to quasi-shear wave velocity with maximum value of 3.30×10^3 m/s at 0 K temperature and angle θ = 55°. We could not find the values of ultrasonic velocities of ScZrHf HCP alloy but on comparison to similar materials[8,25], a good agreement in dependence with temperature and angle has been found.

The Debye temperature θ_D decreases from 303.2 K to 292.8 K in temperature range of 0-900 K. This shows that the Debye temperature does depend on temperature but the dependence is not very significant.

The heat capacity C_V and thermal energy density E_0 have been evaluated in temperature range of 0-900 K

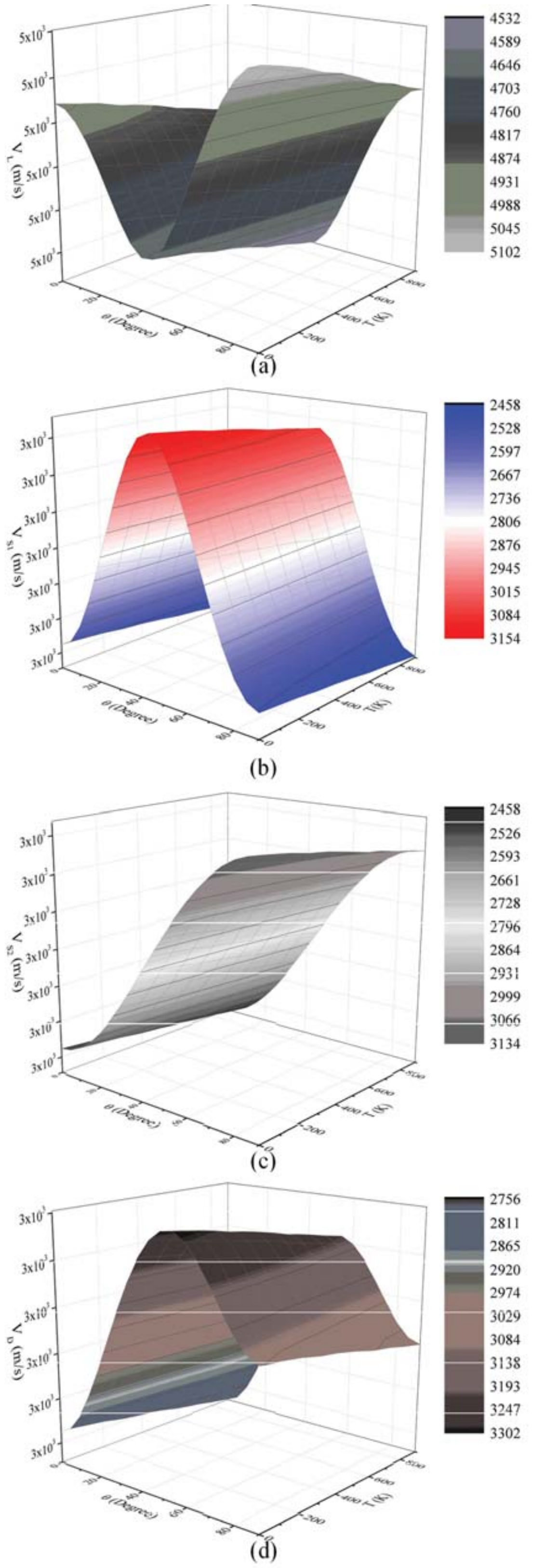

Fig. 4. Ultrasonic (a) longitudinal (V_L), (b) quasi-shear (V_{S1}) and (c) shear (V_{S2}) and (d) Debye average (V_D) velocities of ScZrHf at different temperatures.

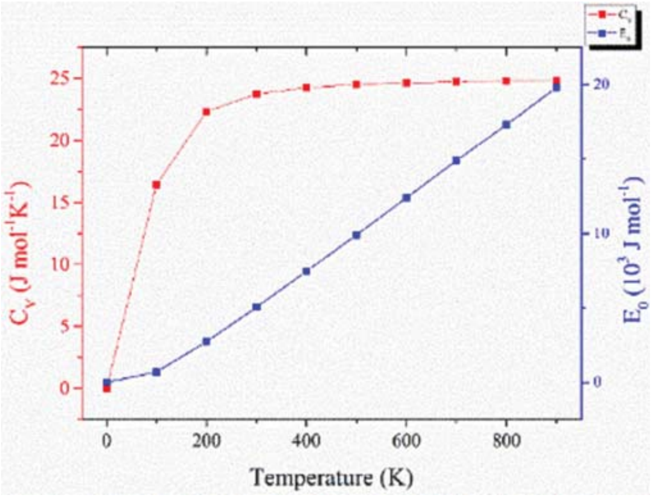

Fig. 5. Temperature dependence of heat capacity (C_V) and thermal energy density (E_0)

by employing the Debye model and have been plotted in Fig. 5.

Figure 5 depicts that C_V increases with temperature but the dependence becomes less and less significant as the temperature increases making the plot with temperature plateau at higher temperature. The thermal energy density (E_0) shows an almost linear increment with increases in temperature.

The thermal conductivity is a key factor in material characterization and to evaluate it for the alloy, we used Eq. 1. The Grüneisen numbers required for the computation of thermal conductivity have been calculated with the help of the thermal expansion coefficient from the literature[7]. The temperature

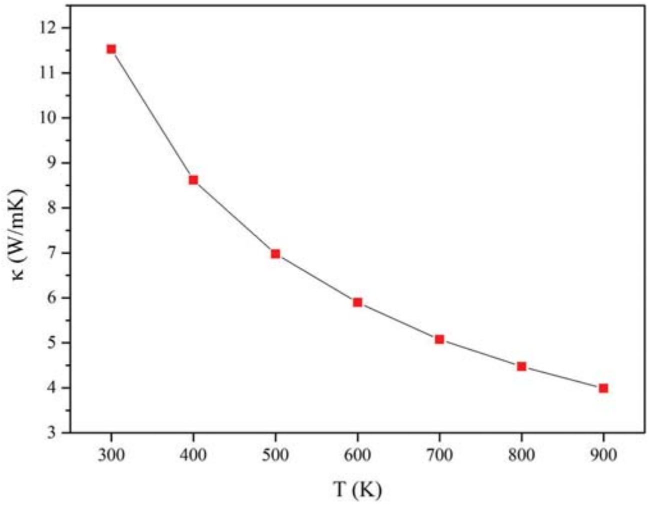

Fig. 6. Temperature dependence of thermal conductivity of ScZrHf

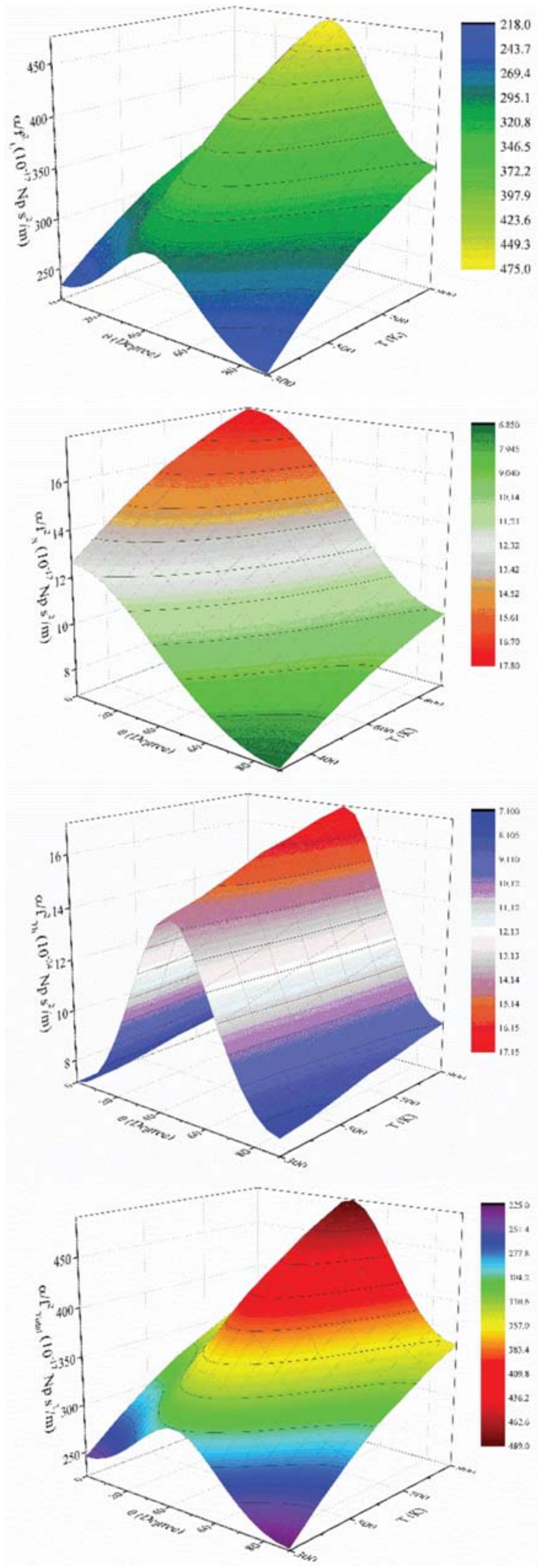

Fig. 7. Direction and temperature dependence of (a) longitudinal, (b) shear, (c) due to thermos-relaxation mechanism and (d) total ultrasonic attenuation of ScZrHf

dependence of thermal conductivity is plotted in the Fig. 6.

The thermal conductivity is 11.52 W/mK at 300 K and decreases with increase in temperature from 300 K-900 K.

The major goal of the present investigation is to examine the ultrasonic behaviour the metal alloy ScZrHf due to the fact that the ultrasonic attenuation is directly correlated to thermoelastic properties of the material. As the ultrasonic attenuation due to phonon-phonon interaction known as Akhiezer loss and due to thermoelastic relaxation are of great interest due to their significance over other losses in a perfect crystal at high (>100 K) temperature, these attenuation have been evaluated using Eqs. (2)-(4) in the temperature range of 300-900K and are presented in Fig. 7.

The ultrasonic attenuation in longitudinal mode $(\alpha/f^2)_L$ shows highest value of 474.41×10^{-17} Np s^2/m at temperature 900 K and angle (θ) 45° and lowest value of 218.97×10^{-17} Np s^2/m at 300 K of temperature and 90° angle (θ) with unique axis. Akhiezer[20] already suggested that the attenuation due to phonon-phonon interaction dominates over other forms of attenuations including thermo-elastic relaxation mechanism and is plotted in Fig. 7 clearly exhibit this. The ultrasonic attenuation due to thermo-relaxation mechanism $(\alpha/f^2)_{th}$ is in range of $6.87\times10^{-24} - 17.12\times10^{-24}$ Np s^2/m in the temperature range of 300-900 K.

The total attenuation $(\alpha/f^2)_{Total}$ is sum of ultrasonic attenuation due to phonon-phonon interactions (longitudinal and shear modes) and due to thermo-relaxation mechanism. As attenuation in longitudinal mode dominates over shear mode and attenuation due to thermo-relaxation mechanism, the total attenuation shows similar nature with temperature and angle with unique axis.

Conclusion

Based on the obtained results and discussion, the following conclusions have been drawn:

(i) The results obtained for SOECs and TOECs of ScZrHf are in good agreements with other studies available in literature. This confirms the significance and successful application of the Lennard-Jones potential approach.

(ii) The alloy shows strong mechanical stability.

(iii) The heat capacity followsDulong-Petit law and the plot with temperature becomes plateau at high temperature.

(iv) The lattice thermal conductivity decreases with temperature. It suggests that the Electronic thermal conductivity dominates at high temperature in the alloy.

(v) The ultrasonic attenuation in longitudinal mode is predominant over shear mode and thermoelastic attenuation.

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