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Ultrasonic wave propagation in nickel aluminide based ternary alloy

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ABSTRACT

Theoretical calculations were performed to investigate the ultrasonic properties of NiAl based ternary alloy NiAl-Cu. The interaction potential model approach has been used to measure the temperature dependent single crystal higher order elastic constants. Other acoustical parameters such as ultrasonic attenuation, Gruneisen numbers, acoustic coupling constants and relaxation time have been also calculated to discuss the acoustical properties insight the ternary alloys. Attention is given to analyze the ultrasonic wave propagation behavior at different temperatures for the alloy and is correlated with respect to the microstructural phenomena during the wave propagation and thermal behavior of the ternary alloy. An ultrasonic mechanism has been developed to correlate the temperature dependent ultrasonic properties with thermophysical properties particularly with the thermal conductivity of the material. Results from the theoretical model calculations were found to be in good agreement with the experimental measurements on the alloy.

KEYWORDS

Nickel aluminide based ternary alloy;
Elastic constants;
Ultrasonic properties;
Phonon-phonon interaction.

INTRODUCTION

Studies about the various properties of binary and ternary alloys are technologically very important[1,2]. NiAl is well known shape memory alloy having high thermal conductivity[3-7]. For higher temperature applications nickel aluminide has been considered as base material to replace nickel-based super alloys[8,9]. Alloying is considered to be effective to improve the interesting properties on NiAl. There are various features of nickel aluminides based ternary alloys[10-18] which make the ternary alloy very useful for various advanced applications such as in making gas turbine and aircraft engines[19,20]. It is important to study the mechanical properties of alloys for applications at elevated temperatures. There are very few theories to describe the physical properties of the alloys such as Young's modulus, heat capacity, thermal conductivity etc. Among these physical properties our main work is focused to develop a theory for the evaluation of second and third order elastic constants (SOEC and TOEC) and ultrasonic attenuation in the alloys and to correlate it with the thermal conductivity of the alloys at high temperatures. The ultrasonic properties are well correlated to micro structural and thermophysical properties of the materials[21-24]. Measurement of ultrasonic attenuation in alloys is very useful in order to characterize the materials. Therefore we have established the theoretical approach for the determination of the temperature dependent ultrasonic attenuation, velocity and anisotropic elastic properties of the NiAl-Cu ternary alloy. Present paper is concerned with the theoretical estimations of

anharmonic higher order elastic constants and ultrasonic properties of the ternary alloys. The calculations have been done for the $Ni_{50.0}$-$Al_{48.0}$-$Cu_{2.0}$ ternary alloy. The ultrasonic attenuation, thermal relaxation time, ultrasonic velocity and non linearity parameters in the ternary single crystals NiAl-Cu is calculated in the temperature range 300-1000K along <100> direction. Our theory provides powerful computational techniques for the basic knowledge about the new materials development as it is very costly and time consuming process to improve and develop the new alloys through extensive experimental trial and error method.

Theory

The elastic constant of the n^{th} order is defined as[25]:

$$C_{ijklmn\ldots} = (\partial^n F/\partial\eta_{ij}\partial\eta_{kl}\partial\eta_{mn}\ldots) \quad (1)$$

Here F is the free energy density of undeformed material and η_{ij} is lagrangian strain components tensor. The total free energy density F expanded in terms of strain η, can be written (using Taylor's series expansion) as:

$$F = \sum_{n=0}^{\infty} F_n = \sum_{n=0}^{\infty} \frac{1}{n!}(\partial^n F/\partial\eta_{ij}\partial\eta_{kl}\partial\eta_{mn}\ldots)\eta_{ij}\eta_{kl}\eta_{mn}\cdots \quad (2)$$

Hence the free energy density up to the cubic terms of strain can be written as:

$$F_2 + F_3 = \frac{1}{2!}C_{ijkl}\eta_{ij}\eta_{kl} + \frac{1}{3!}C_{ijklmn}\eta_{ij}\eta_{kl}\eta_{mn}$$

The free energy density of a crystal at a finite temperature T is given as[26]:

$$F = U + F^{Vib} \quad (3)$$

Here U is the internal energy of unit volume of the crystal when all atoms (ions) are at rest on their lattice point. F^{Vib} is the vibrational free energy density. Thus the elastic constants can be separated in to two parts.

$$C_{IJK\ldots} = C^0_{IJK\ldots} + C^{Vib}_{IJK\ldots} \quad (4)$$

The first and the second ones are strain derivatives of U and F^{vib}, and they represent the 'static' and 'vibrational' elastic constants[27].

The elementary cell of bcc structured crystal is supposed to have a cube edge of 2 r_0, hence the volume becomes as $4(r_0)^3$. The nearest neighbour distance is $r_1 = \sqrt{3}r_0$. The summation is done up to second neighbourhood whose co-ordinates are (±1, ±1, ±1) r0, (±b2,0,0) r0, (0±b2,0) r0 and (0,0±b2)r0. The obtained expression for second and third order elastic constants at 0K using above equations are as:

$$\left.\begin{aligned}
C^0_{11} &= \frac{3}{8}\frac{e^2}{r_0^4}S_5^{(2)} + \frac{3\phi(r_1)}{br_0}\left(\frac{\sqrt{3}}{3r_0}+\frac{1}{b}\right) + \frac{2\phi(r_2)}{br_0}\left(\frac{1}{2r_0}+\frac{1}{b}\right)\\
C^0_{12} &= C^0_{44} = \frac{3}{8}\frac{e^2}{r_0^4}S_5^{(1,1)} + \frac{\phi(r_2)}{br_0}\left(\frac{1}{2r_0}+\frac{1}{b}\right)\\
C^0_{111} &= -\frac{15}{8}\frac{e^2}{r_0^4}S_7^{(3)} - \frac{\phi(r_1)}{9b}\left(\frac{\sqrt{3}}{r_0^2}+\frac{3}{br_0}+\frac{\sqrt{3}}{b^2}\right) -\\
&\frac{\phi(r_2)}{2b}\left(\frac{3}{r_0^2}+\frac{6}{br_0}+\frac{4}{b^2}\right)\\
C^0_{112} &= C^0_{166} = -\frac{15}{8}\frac{e^2}{r_0^4}S_7^{(2,1)} - \frac{\phi(r_1)}{9b}\left(\frac{\sqrt{3}}{r_0^2}+\frac{3}{br_0}+\frac{\sqrt{3}}{b^2}\right)\\
C^0_{123} &= C^0_{456} = C^0_{144} = -\frac{15}{8}\frac{e^2}{r_0^4}S_7^{(1,1,1)} -\\
&\frac{\phi(r_1)}{9b}\left(\frac{\sqrt{3}}{r_0^2}+\frac{3}{br_0}+\frac{\sqrt{3}}{b^2}\right)
\end{aligned}\right\} \quad (5)$$

where $\phi(r_1) = A\exp(-r_1/b)$, $\phi(r_2) = A\exp(-r_2/b)$, $r_1 = \sqrt{3}r$, $r_2 = 2r_0$ and S is the lattice sum[27]. The value of A can be obtained with the equilibrium condition and is given as:

$$A = (bZ_0e^2/r_0^2)\left[8\sqrt{3}\exp(-r_1/b) + 12\exp(-r_2/b)\right] \quad (6)$$

The elastic constants due to vibrational free energy component is given by[28]:

$$C^{Vib}_{IJK\ldots} = a_{IJK\ldots}T \quad (7)$$

Here

$$a_{IJK\ldots} = l_1 K\ B\left|\frac{\partial C^0_{IJK\ldots}}{\partial r}\right|_{r=r_0} + \frac{f^{Vib}_{IJK}}{TV_C},$$

$$l_1 = -r_0\begin{bmatrix}\left\{\frac{8}{3}\left(2y_1 + 2y_1^2 - y_1^3\right)\phi(r_1)\right\} + \\ \left\{\frac{3}{2}\left(2y_2 + 2y_2^2 - y_2^3\right)\phi(r_2)\right\}\end{bmatrix}Y^{-1} \quad (8)$$

$$Y = \begin{bmatrix}\left\{\frac{8}{3}\left(y_1^2 - 2y_1\right)\phi(r_1)\right\} + \\ \left\{\frac{3}{2}\left(y_2^2 - 2y_2\right)\phi(r_2)\right\}\end{bmatrix}\begin{bmatrix}\left\{\frac{8}{3}\left(y_1^2 - 2y_1\right)\phi(r_1)\right\} + \\ \left\{2\left(y_2^2 - 2y_2\right)\phi(r_2)\right\}\end{bmatrix} \quad (9)$$

where $y_1=r_1/b$, $y_2=r_2/b$, k_B is the Boltzmann constant and $f^{Vib}_{IJK....}$ is the vibrational free energy per unit cell[27]. Using above expressions, the vibrational part of elastic components have been calculated by eqn.(7), whose addition to static part of elastic constants gives the elastic constants at a particular temperature.

For the evaluation of ultrasonic attenuation coefficient, we have used the phonon-phonon interaction mechanism given by Mason and Bateman[29,30]. It is more genuine theory for studying the anharmonicity of crystals as it directly involves elastic constants through non-linearity parameters D in the evaluation of ultrasonic attenuation coefficient α. The ultrasonic attenuation over frequency square $(\alpha/f^2)_{Akh}$ (Akhieser type loss) due to phonon- phonon interaction mechanism at $\omega\tau<<1$ is expressed as[31]:

$$(\alpha/f^2)_{Akh} = \frac{E_0 (D/3) 4\pi^2 \tau}{2dV^3} \quad (10)$$

where $$D = 9<(\gamma_i^j)^2> - 3<\gamma_i^j>^2 \frac{C_v T}{E_0} \quad (11)$$

Here D and V are the non-linearity parameter and velocity of ultrasonic wave for longitudinal and shear wave, E_0 and C_v are thermal energy density and the specific heat per unit volume respectively. $<\gamma_i^j>$ is the average Grüneisen no., j is the direction of propagation and i is the mode of propagation. $<\gamma_i^j>$is determined using SOEC/TOEC[29]. τ is the thermal relaxation time which is equal to thermal relaxation time for shear wave (τ_s) and is half of thermal relaxation time for longitudinal wave $(\tau_L)$and is written as:

$$\tau = \tau_S = \tau_L/2 = \frac{3K}{C_V V_D^2} \quad (12)$$

where K is the thermal conductivity and V_D is the Debye average velocity of ultrasonic wave as:

$$\frac{3}{V_D^3} = \frac{1}{V_L^3} + \frac{2}{V_S^3} \quad (13)$$

where V_L and V_S are the velocity of longitudinal and shear wave respectively.

The propagation of longitudinal wave creates compression and rarefaction throughout the lattice. The rarefied regions are colder than that of compressed regions. Thus there is flow of heat between these two regions. Hence thermoelastic loss $(\alpha/f^2)_{Th}$ occurs and is obtained by the expression as[31]:

$$(\alpha/f^2)_{Th.} = \frac{4\pi^2 <\gamma_i^j>^2 KT}{2dV_L^5} \quad (14)$$

RESULTS

The higher order elastic constants (SOEC and TOEC) in the temperature range 300-1000K have been calculated using eqns. (5) and (7). Calculated values of SOEC and TOEC for NiAl-Cu at different temperatures are presented in TABLES 1 and 2. Thermal conductivity (K) data at different temperatures are taken from the literature[32]. The values of specific heat per

TABLE 1: SOEC (10^{10}N/m^2) of NiAl-Cu in the temperature range 300K to 1000K

SOEC → T.[K]↓	C_{11}	C_{12}	C_{44}
300	14.84	11.63	9.67
400	14.87	11.60	9.72
500	14.89	11.58	9.78
600	14.91	11.55	9.83
700	14.93	11.53	9.88
800	14.95	11.50	9.93
900	14.98	11.47	9.98
1000	15.00	11.45	10.03

TABLE 2: TOEC(10^{10}N/m^2) of NiAl-Cu in the temperature range 300K to 1000K

TOEC→ T.[K]↓	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
300	-141.27	-179.36	-123.15	-103.75	-171.50	-132.23
400	-137.52	-167.42	-115.88	-90.02	-156.95	-127.99
500	-133.77	-155.48	-108.62	-76.29	-142.39	-123.75
600	-130.03	-143.55	-101.36	-62.55	-127.84	-119.51
700	-126.28	-131.61	-94.09	-48.82	-113.29	-115.27
800	-122.53	-119.67	-86.82	-35.09	-98.74	-111.03
900	-118.79	-107.74	-79.56	-21.36	-84.18	-106.79
1000	-115.04	-95.79	-72.29	-7.62	-69.63	-102.56

TABLE 3: Thermal conductivity (K), density (ρ), specific heat per unit volume (C_V) and thermal energy density (E_0) of NiAl-Cu in the temperature range 300-1000K

T[K]	K (J/sec. m. K)	ρ (10^3Kg/m^3)	C_V (× 10^6 J/m^{3}K)	E_0 (x 10^8 J/m^3)
300	6.77	5.913	8.88	1.59
400	7.67	5.907	9.22	2.51
500	8.33	5.900	9.49	3.45
00	8.99	5.894	9.64	4.39
700	9.33	5.887	9.69	5.35
800	9.56	5.881	9.73	6.33
900	9.67	8.874	9.72	7.28
1000	9.56	5.867	9.76	8.24

TABLE 4: Ultrasonic velocities (V_L, V_S, Debye average velocity (V_D) in10^3m/s) and thermal relaxation time (τ) (in 10^{-13} Sec) in NiAl-Cu in the temperature range 300-1000K

T[K]	V_L	V_S	V_D	τ
300	5.011	4.046	4.229	128.03
400	5.018	4.058	4.241	138.71
500	5.024	4.071	4.252	145.67
600	5.031	4.084	4.264	154.12
700	5.037	4.097	4.276	158.04
800	5.044	4.109	4.288	160.27
900	5.050	4.122	4.299	161.42
1000	5.057	4.135	4.311	158.05

TABLE 5 : Average Grüneisen number $\langle\gamma_i^j\rangle_L$ for longitudinal wave, average square Grüneisen number $\langle(\gamma_i^j)^2\rangle_L$ and $\langle(\gamma_i^j)^2\rangle_S$ for longitudinal and shear wave and acoustic coupling constant for longitudinal (D_L) and shear (D_S) waves for NiAl-Cu from 300-1000K

T (K)	$\langle\gamma_i^j\rangle_L$	$\langle(\gamma_i^j)^2\rangle_L$	$\langle(\gamma_i^j)^2\rangle_S$	D_L	D_S
300	0.191	1.155	0.482	10.211	4.338
400	0.158	1.061	0.470	9.441	4.231
500	0.124	0.978	0.459	8.737	4.134
600	0.092	0.905	0.449	8.109	4.048
700	0.059	0.841	0.441	7.558	3.972
800	0.028	0.787	0.433	7.080	3.906
900	0.003	0.741	0.428	6.673	3.849
1000	0.034	0.704	0.422	6.333	3.802

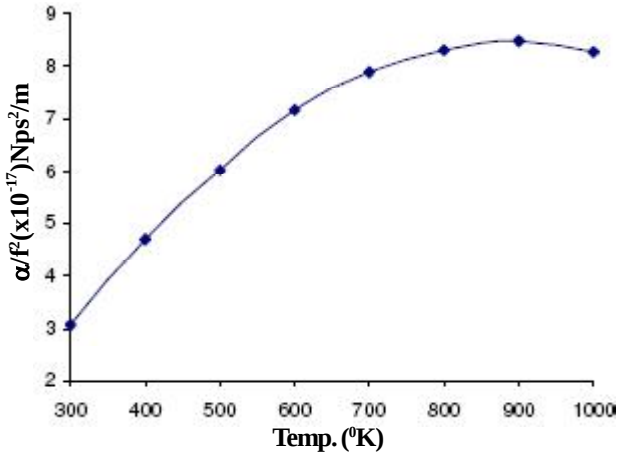

Figure1: $(\alpha/f^2)_{th.}$ vs temperature

unit volume (C_V) and thermal energy density (E_0) are evaluated using physical constant table and Debye temperature[33] and are presented in TABLE 3.

The ultrasonic longitudinal and shear wave velocities are calculated using SOEC. Thermal relaxation time (τ) is calculated by eqn. (12) using thermal conductivity data and Debye average velocity using eqn.(13) at different temperatures. The values of longitudinal, shear wave velocities, Debye average velocity (V_D) and thermal relaxation time (τ) at different temperatures are shown in TABLE 4. The Gruneisen parameters are calculated using SOEC and TOEC in the temperature range 300-1000K. The values of Gruneisen parameters, acoustic coupling constants for longitudinal (D_L) and shear wave (D_S) are presented in TABLE 5. The ultrasonic attenuation coefficient over frequency square $(\alpha/f^2)_{Akh}$ for longitudinal and shear wave are calculated using eqn.(10). Values of thermoelastic loss over frequency square $(\alpha/f^2)_{Th}$ is calculated using eqn.(14). The calculated values of attenuation for different types of losses are plotted in figures 1-2. The temperature dependent variation of anisotropy is shown in figure 3.

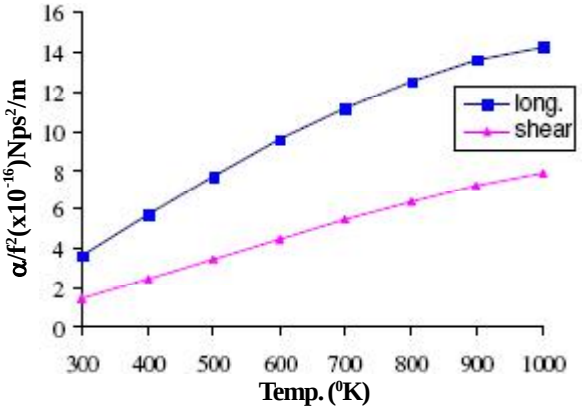

Figure 2: $(\alpha/f^2)_{Akh.long}$ and $(\alpha/f^2)_{Akh.shear}$ vs temperature

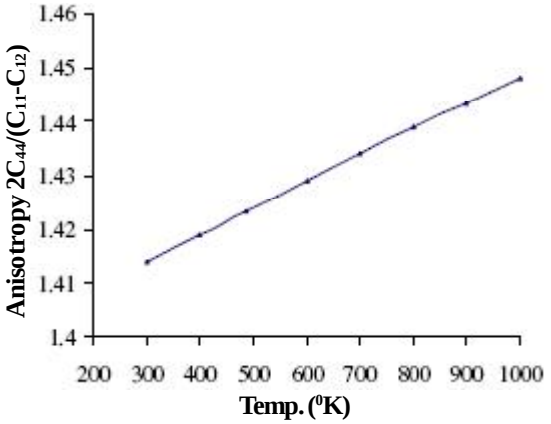

Figure 3: Anisotropy vs temperature

DISCUSSION AND CONCLUSIONS

At the room temperature the values of SOEC for the alloy NiAl-Cu ($Ni_{50.0}Al_{48.0}Cu_{2.0}$) C_{11}, C_{12} and C_{44} are 148.4 GPa, 116.3 GPa and 96.7 GPa determined by our theoretical calculations. M. Landa and cowork-

ers[7] calculated the values for the alloy $Ni_{81.6}Al_{4.1}Cu_{14.3}$, C_{11}=142.80GPa, C_{12}= 126.84 GPa and C_{44}=95.90 GPa by experimental observation. There is a good agreement between the values calculated by our interaction potential model approach and other workers. In the present calculations C_{111} is negative. The value of C_{111} for CsCl and Al is negative[28,34]. We can say that our theory for the calculation of the higher order elastic constants is justified. Bulk modulus of the alloy NiAl-Cu is 127.03 GPa and that is 158.5GPa for NiAl[35]. Thus the present alloy is more ductile at 300K due to ternary addition of Cu. This may be correlated as the ultrasonic attenuation in the alloy NiAl-Cu increases from 300K onwards (figure 1) and it decreases from 300K onwards in NiAl[36]. The ratio $A=2C_{44}/(C_{11}-C_{12})$ is the measure of elastic anisotropy in the crystal. The temperature dependent variation of A for the alloy NiAl-Cu is shown in figure 3. The values of A for the alloy NiAl-Cu is1.414 while the value of A for NiAl is ≈0.33[36]. Hence by proposing that high value of anisotropy favors stability, we may say that the alloy NiAl-Cu is more stable than NiAl at room temperature. Also the value of A increases with the temperature. Thus the present alloy is more stable at higher temperature, hence the applicability of the ternary alloys enhances at higher temperatures.

A perusal of figure 1 shows that the ultrasonic attenuation due to thermal relaxation process for NiAl-Cu increases from 300K to 900K and its value become slightly less for 1000K. This behavior can be correlated with the thermal conductivity of NiAl-Cu alloy as the thermal conductivity increases from 300K up to 900K then its value decreases slightly for higher temperature. figure 1 reveals that ultrasonic attenuation due to thermoelastic loss is mainly affected by the elastic constants through Gruneisen parameters. In similar way figure 2 reveals that the ultrasonic attenuation due to phonon-phonon interaction (Akhiezer type loss) for longitudinal wave increases almost linearly from 300-1000K for NiAl-Cu alloy. From figure 2 we can observe that the ultrasonic attenuation due to phonon-phonon interaction for shear wave increases linearly from 300-1000K. The acoustic coupling constants (D_L and D_S) contribute to the temperature variation of ultrasonic attenuation. The acoustic coupling constant depends on elastic constants through Gruneisen parameters, specific heat per unit volume and thermal energy density. In TABLE 3 one can observe that the thermal energy density for the ternary alloy NiAl-Cu is increasing with temperature. So the temperature dependent behavior of ultrasonic attenuation is also affected by the thermal energy density as from figure 2 it is clear that the attenuation due to phonon-phonon interaction is increasing with temperature. The variation of thermal conductivity with temperature also affects reasonably the ultrasonic attenuation for the ternary alloy NiAl-Cu. Such type of temperature dependency of ultrasonic attenuation due to thermal relaxation process can be correlated with the behavior of temperature dependent thermal conductivity of the material as the thermal conductivity for the alloy increases linearly with temperature[32]. Thus among various parameters affecting the ultrasonic attenuation, thermal conductivity plays important role governing the temperature dependent variation of ultrasonic attenuation.

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