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Behaviour of ultrasonic velocities and elastic constants in Ag-Zn alloys

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ABSTRACT

The ultrasonic properties of the hexagonal closed packed structured Ag-Zn alloys have been studied at room temperature for their characterization. For the investigations of ultrasonic properties, I have also computed second order elastic constants using Lennard–Jones Potential. The velocities V_1 and V_2 have minima and maxima respectively with 45^0 with unique axis of the crystal, while V_3 increases with the angle from unique axis. The inconsistent behaviour of angle dependent velocities is associated to the action of second order elastic constants. Debye average sound velocities of these alloys are increasing with the angle and has maximum at 55^0 with unique axis at room temperature. Hence when a sound wave travels at 55^0 with unique axis of these alloys, then the average sound velocity is found to be maximum. $Ag_{0.2}Zn_{0.8}$ alloy is more suitable for industrial and other uses, as it having highest elastic constants and lowest attenuation in comparison to other Ag-Zn samples. The mechanical and ultrasonic properties of these alloys will not be better than pure Ag and Zn due to their low SOEC and high ultrasonic attenuation. Achieved results have been discussed and compared with available experimental and theoretical results.

Keywords: Alloys; elastic properties; ultrasonic properties.

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Introduction

Ultrasonic offer the possibility to detect and characterize microstructral properties as well as flaws in materials, controlling materials behaviour based on physical mechanism to predict future performance of the materials. Various investigators have shown considerable interest on ultrasonic properties of metals and alloys [**1-4**]. Wave propagation velocity is key parameter in ultrasonic characterization and can provide information about crystallographic texture. The ultrasonic velocity is directly related to the elastic constants by the relationship $V=\sqrt{(C/\rho)}$, where C is the relevant elastic constants and ρ is the density of that particular material. The elastic constants of a solid provide valuable insight into nature of atomic bonding forces and also related hardness [**5, 6**].

The crystal structure gives the key to many properties of solid materials. Unusual structural properties that can be subtly tuned by chemical composition or external conditions are of great interest. Most elemental metals crystallizing in the hexagonal closed packed (hcp) lattice have an axial ratio c/a that is close to the "ideal" value 1.633 obtained in a stacking of rigid spheres [**7**].

Yoshie Matsuo [**8**] has been experimentally investigated elastic behaviour and phase stability in the hcp structure of Ag-Zn alloys using an ultrasonic pulse-echo overlapping method. An X-ray diffraction pattern obtained from the pink color film showed CsCl-type β phase (bcc) structure, and the pattern from silver color film showed ζ phase (hcp) structure of Ag-Zn alloys films [**9**].

Zener alloys (Ag-Zn alloys), which show Zener relaxation, have a great interest for scientists since last four decades [**10-12**].Low temperature internal friction and sound velocity in Zener alloys have been measured by Raychaudhuri and Pohl [**13**]. Magyari-Kope *et al*. [7] investigated lattice parameter and elastic constants in Zener alloys using *ab-initio* calculation. Interdiffusion coefficients in Ag-Zn α phase in the temperature range between 823 K and 1023 K have been determined by Suer and Freise analysis using solid diffusion couples consisting of pure silver and a 20 at % Zn-Ag alloys [**14**].

There are three types of acoustic mode of lattice vibrations: one longitudinal acoustical and two transverse

acoustical for hexagonal structured materials. Hence, there are three types of ultrasonic wave velocities for each direction of propagation of wave, which are well related to second order elastic constants. But all the three types of ultrasonic velocities and elastic constants of these alloys are not reported in the literature. Therefore in this paper, I have calculated the three types of ultrasonic sound velocities for the alloys $Ag_{1.0}Zn_{0.0}$: alloy-1; $Ag_{0.9}Zn_{0.1}$: alloy-2; $Ag_{0.8}Zn_{0.2}$: alloy-3; $Ag_{0.7}Zn_{0.3}$: alloy-4; $Ag_{0.6}Zn_{0.4}$: alloy-5; $Ag_{0.5}Zn_{0.5}$: alloy-6; $Ag_{0.4}Zn_{0.6}$: alloy-7; $Ag_{0.3}Zn_{0.7}$: alloy-8; $Ag_{0.2}Zn_{0.8}$: alloy-9 for each direction of propagation of wave using second order elastic constants that are important for surface and structural study of these alloys. The six second order elastic constants are calculated using Lenard-Jones Potential that is a many body interaction potential. The results obtained are interesting for the characterization of these alloys.

Theory

Second order elastic constants

The elastic energy density (U) is function of the strain components.

$U = F(e_{xx}; e_{yy}; e_{zz}; e_{yz}; e_{zx}; e_{xy}) = F(e_1; e_2; e_3; e_4; e_5; e_6)$

---------- (1)

where e_{ij}(i or j = x, y, z) is component of strain tensor. The second (C_{IJ}) order elastic constants of material are defined by following expressions.

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

---------- (2)

The elastic energy density is well related to interaction potential φ(r) between atoms. Let the interaction potential be the Lennard–Jones Potential or many body interaction potential, which is formulated as:

$$\Phi(r) = -\frac{a_0}{r^m} + \frac{b_0}{r^n};$$

---------- (3)

where a_0, b_0 are constants and m, n are integers. The definition of higher order elastic constants (Eqn.2) with this potential (Eqn.3) under equilibrium and symmetric condition leads six second order elastic constants (SOEC) for the hexagonal closed packed structured materials [15, 16]

$$\left.\begin{aligned} C_{11} &= 24.1\, p^4 C' & C_{12} &= 5.918\, p^4 C' \\ C_{13} &= 1.925\, p^6 C' & C_{33} &= 3.464\, p^8 C' \\ C_{44} &= 2.309 p^4 C' & C_{66} &= 9.851\, p^4 C' \end{aligned}\right\}$$

---------- (4)

where p = c/a: axial ratio; $C' = \chi\, a / p^5$; $B = \psi\, a^3 / p^3$;

The rest second order elastic constants have zero value because under 180° rotation they have equal and opposite value for the same stress. The harmonic and anharmonic parameters (χ and ψ) can be calculated using one experimental SOEC [15, 16]. In the present study, I have expanded the theory for theoretical evaluation of parameters χ and ψ. The potential energy can be expanded in the powers of changes in the squares of distances. The expansion up to cubic term can be written as:

$$\Phi = \Phi_0 + \chi \sum_{i=1}^{2} \left[\Delta r_i^2\right]^2 + \psi \sum_{i=1}^{2} \left[\Delta r_i^2\right]^3$$

---------- (5)

According to the Eqn. (5), χ and ψ can be written as:

$$\chi = \frac{1}{2!}\left[\frac{d^2\Phi(r)}{d(r^2)^2}\right]$$

---------- (6)

$$\psi = \frac{1}{3!}\left[\frac{d^3\Phi(r)}{d(r^2)^3}\right]$$

---------- (7)

In solving Eqns. (6) and (7) for hexagonal closed packed structured materials:

$$\chi = (1/8)[\{nb_0\,(n-m)\}/\{a^{n+4}\}]$$

---------- (8)

$$\psi = -\chi/\{6\,a^2(m+n+6)\}$$

---------- (9)

The parameters χ and ψ can be calculated using Eqs. (8)- (9) with appropriate values of m, n and b_0 so that the calculated values of elastic constants justify the experimental data.

Ultrasonic velocity

The anisotropic behaviour of the material can be understood with the knowledge of ultrasonic velocity because the velocity is related to the second order elastic constants [**16**]. On the basis of mode of atomic vibration, there are three types of velocities (longitudinal, quasi shear and shear) in acoustical region [**17**]. These velocities vary with the direction of propagation of wave from the unique axis of hexagonal structured crystal [**18, 19**]. The ultrasonic velocities as a function of angle between direction of propagation and unique axis for hexagonal structured materials are [**20**]:

$$V_1^2 = \{C_{33}\cos^2\theta + C_{11}\sin^2\theta + C_{44} + \{[C_{11}\sin^2\theta - C_{33}\cos^2\theta + C_{44}(\cos^2\theta - \sin^2\theta)]^2 + 4\cos^2\theta \sin^2\theta\,(C_{13} + C_{44})^2\}^{1/2}\}/2\rho$$

---------- (10)

$$V_2^2 = \{C_{33}\cos^2\theta + C_{11}\sin^2\theta + C_{44} - \{[C_{11}\sin^2\theta - C_{33}\cos^2\theta + C_{44}(\cos^2\theta - \sin^2\theta)]^2 + 4\cos^2\theta\sin^2\theta(C_{13}+C_{44})^2\}^{1/2}\}/2\rho$$

---------- (11)

$$V_3^2 = \{C_{44}\cos^2\theta + C_{66}\sin^2\theta\}/\rho$$

---------- (12)

where V_1, V_2 and V_3 are longitudinal, quasi shear and pure shear wave ultrasonic velocities. Variables ρ and θ represent the density of the material and angle with the unique axis of the crystal respectively. The Debye temperature (T_D) is an important physical parameter for the characterization of materials, which is well related to the Debye average velocity (V_D).

$$T_D = \frac{\hbar\, V_D\, (6\pi^2\, n_a)^{1/3}}{k_B}$$

---------- (13)

here $$V_D = \left\{\frac{1}{3}\left(\frac{1}{V_1^3} + \frac{1}{V_2^3} + \frac{1}{V_3^3}\right)\right\}^{-1/3}$$

---------- (14)

where $\hbar$ is quantum of action and is equal to Planck's constant divided by 2π ; k_B is Boltzmann Constant; n_a is atom concentration.

The above formulae have been used for the evaluation of ultrasonic velocity and related parameters for the selected materials. All the relevant mathematical formulations are given as: **Appendix-1**.

Results and discussion

The unit cell parameters 'a', axial ratio 'p' and density 'ρ' of the chosen alloys are presented in **Table 1**[**21**]. The value of m and n for these alloys are 6 and 7. The value of b_0 is 1.0×10^{-67} erg cm^7 for all alloys. The second order elastic constants and bulk modulus of these alloys are presented in **Table 2**.

Table1. Lattice parameter 'a' (in Å), 'p' and density 'ρ' (in 10^2 gm/cm^3) of Ag-Zn alloys at room temperature.

Alloys	a	p	ρ
$Ag_{1.0}Zn_{0.0}$	1.656	1.645	2.366
$Ag_{0.9}Zn_{0.1}$	1.64	1.64	2.315
$Ag_{0.8}Zn_{0.2}$	1.641	1.646	2.219
$Ag_{0.7}Zn_{0.3}$	1.63	1.645	2.188
$Ag_{0.6}Zn_{0.4}$	1.619	1.63	2.133
$Ag_{0.5}Zn_{0.5}$	1.609	1.613	2.072
$Ag_{0.4}Zn_{0.6}$	1.598	1.598	2.011
$Ag_{0.3}Zn_{0.7}$	1.582	1.579	1.966
$Ag_{0.2}Zn_{0.8}$	1.572	1.574	1.894

Table 2. Second order elastic constants (SOEC) and Bulk modulus (B) (in $10^{10}Nm^{-2}$) of Ag-Zn alloys at room temperature.

Alloys	C_{11}	C_{12}	C_{13}	C_{33}	C_{44}	C_{66}	B
$Ag_{1.0}Zn_{0.0}$	8.265	2.030	1.786	8.699	2.143	3.241	4.048
$Ag_{0.9}Zn_{0.1}$	8.808	2.163	1.892	9.158	2.270	3.454	4.297
$Ag_{0.8}Zn_{0.2}$	9.047	2.222	1.958	9.545	2.348	3.548	4.434
$Ag_{0.7}Zn_{0.3}$	9.682	2.378	2.093	10.19	2.510	3.797	4.742
$Ag_{0.6}Zn_{0.4}$	10.456	2.568	2.219	10.61	2.662	4.100	5.059
$Ag_{0.5}Zn_{0.5}$	11.242	2.761	2.336	10.94	2.802	4.409	5.361
$Ag_{0.4}Zn_{0.6}$	12.153	2.984	2.479	11.39	2.973	4.766	5.718
$Ag_{0.3}Zn_{0.7}$	13.601	3.340	2.709	12.15	3.249	5.334	6.287
$Ag_{0.2}Zn_{0.8}$	14.538	3.570	2.877	12.83	3.451	5.701	6.688
$Ag_{0.3}Zn_{0.7}$ [21Theo.]	11.0	5.6	6.3	12.9	2.7		
$Ag_{0.3}Zn_{0.7}$ [8Exp.]	13.0	6.5	6.4	15.8	4.1		

The elastic constants of the material are important, since they are related to hardness parameter. Also, the second order elastic constants are used for the determination of the ultrasonic parameters. The calculated values of C_{12}, C_{13} are few different than some other experimental [**8**] and theoretical [**21**] results for alloys: 8. Actually Magyari-Kope *et al*. [**21**] have based *ab-initio* exact muffin-tin orbitals total energy theoretical approached to evaluate elastic constants, which is quite different from present approach. Also Y. Matsuo [**8**] was experimentally investigated elastic behaviour and phase stability in the hcp structure of Ag-Zn alloys using an ultrasonic pulse-echo overlapping method. Although obtained order of SOEC are of the same as others hexagonal structured materials [**5, 6, 12**]. Relative magnitude of C_{11}, C_{33}, C_{44} are well presented by our theoretical approach. The bulk modulus (B) for these alloys can be calculated with the formula $B= 2(C_{11}+ C_{12}+ 2C_{13} + C_{33}/2)/9$. The evaluated B for these alloys is presented in **Table 2**. It is obvious from **Table 2** that, there is good agreement between the present and reported theoretical/experimental second order elastic constants of these alloys [**8, 21**]. Thus our theoretical approach for the calculation of second order elastic constants for hexagonal structured alloys at room temperature is well justified. Hence applied theory for the evaluation of higher order elastic constants at room temperature is justified.

It can be also seen from **Table 2**, that the SOEC are found to be increasing from alloys: 1-9. Also the SOEC of these alloys are less than pure Zn metals [22] and pure Ag metals [**23**]. This increasing nature of SOEC shows that the alloys are more stable, ductile with more Zn amount.

The compute oriented dependent ultrasonic velocities of these alloys are visualized in **Figs. 1-4**. **Figs. 1–3** clear that velocities V_1 and V_2 have minima and maxima respectively at 45° with the unique axis of the crystal, while V_3 increases with the angle from the unique axis. Indistinct activity of angle dependent velocities is correlated with activity of SOEC. The tendency of angle dependent velocity curves (**Figs. 1-4**) reveals the same nature of angle dependent curves for hexagonal wurtzite

structured materials GaN, AlN, InN, CdS, CdSe and laves-phase compounds [**16, 18-20, 24, 25**]. Hence it is conformity to the hcp structured of chosen alloys.

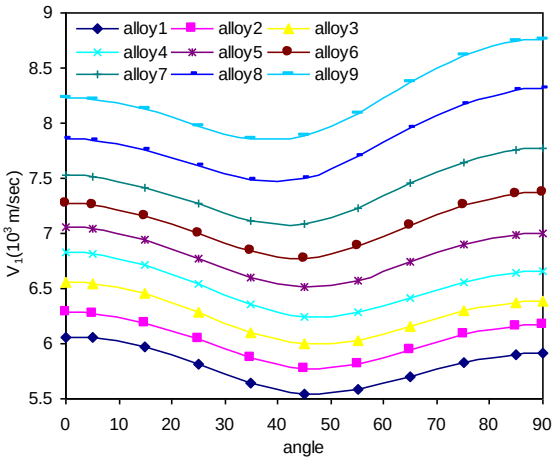

Fig. 1. V_1 vs angle with unique axis of crystal.

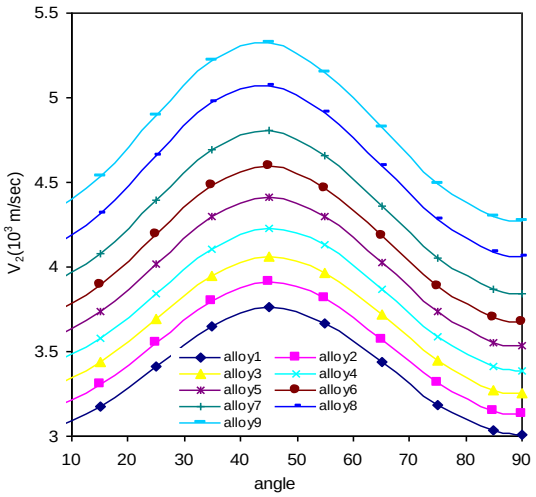

Fig. 2. V_2 vs angle with unique axis of crystal.

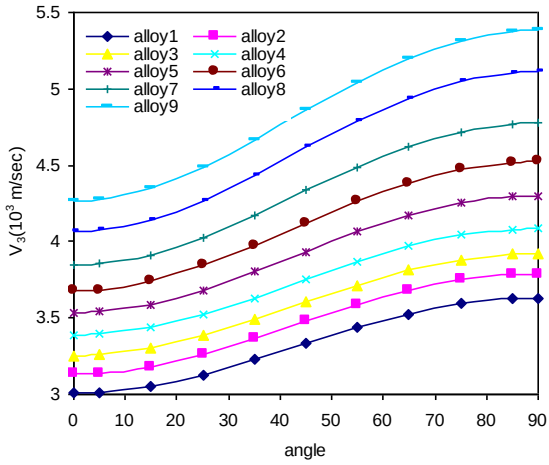

Fig. 3. V_3 vs angle with unique axis of crystal.

Figs. 1–4 depict that the magnitudes of ultrasonic velocities increase from alloy: 1 to alloy: 9 as boost up of zinc amount. Hence as we increase amount of zinc, the quality of the material is also increased. The longitudinal ultrasonic velocity along <001> direction (θ=0° with unique axis) is largest for alloy: 9, because alloy: 9 have highest zinc amount as well as highest C_{33} value. As we know that shear wave is also known surface wave. Therefore the ultrasonic velocities V_2 and V_3 are the surface wave velocity. It is clear from the values of V_2 and V_3 for ultrasonic wave propagation along θ=0° are the same, while these are different for other directions as visualized in **Figs. 2-3**. Hence <001> direction is the direction of symmetry for chosen alloys. Debye average velocities (V_D) of these alloys are increasing with the angle and have maxima at 55° at 300 K (**Fig. 4**). Since V_D is calculated using V_1, V_2 and V_3 with Eqn. (14), therefore the temperature variation of V_D follows the combined effect of temperature variation of V_1, V_2 and V_3. The maximum in V_D at 55° is due to a significant increase in longitudinal and pure shear wave velocities and a decrease in quasi-shear wave velocity.

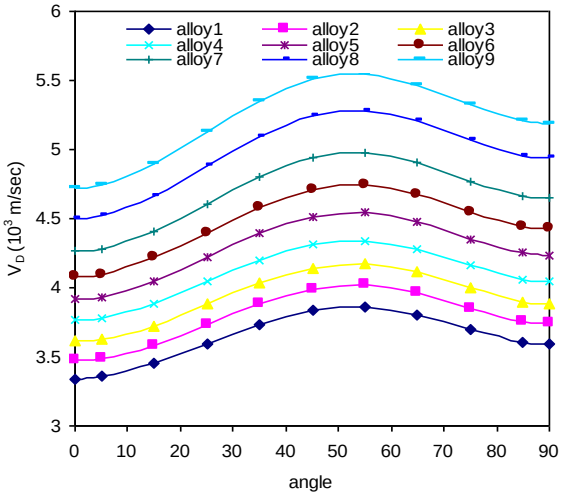

Fig. 4. V_D vs. angle with unique axis of crystal.

It can be seen that from **Figs. 1-4** alloy: 9 has maximum velocity and pure Ag has least velocity for all angles of the crystals. Since ultrasonic attenuation A $\propto V^{-3}$ and velocity is the largest for alloy: 9 among others thus the attenuation A should be smallest and material should be most ductile. The minimum ultrasonic attenuation for alloy: 9 justify its quite stable hcp structure state. Also alloy: 9 has maximum elastic constants and bulk modulus among others. Hence alloy: 9 ($Ag_{0.2}Zn_{0.8}$) is more ductile, stable and contain few defects in the crystal structure in comparison to other alloys.

In the present investigation the expressions in above Table is used for calculating the ultrasonic velocity for longitudinal and shear wave in hexagonal structured materials. An important application of velocity measurements for both longitudinal and shear waves is the determination of the elastic constants along the various axes of single crystals over very wide ranges of temperature. The determination of anisotropy of ultrasonic velocity and, hence elastic constants can yield useful information about other physical properties of the crystal. The degree of anisotropy of the ultrasonic

velocities in a single crystal of such a material should agree with that for the lattice vibrations components of thermal conductivity.

Appendix 1. Ultrasonic velocities for hexagonal structured materials.

Direction of propagation	Direction of polarization	Type of wave	Velocity expression	Velocity notation
001	001	Long.	$(C_{33}/\rho)^{1/2}$	$V_1=V_L$
(Along unique axis or z-axis)	Any direction in 001 plane	Shear	$(C_{44}/\rho)^{1/2}$	$V_2=V_3$ $V_{S1}=V_{S2}$
100 (or any other direction perpendicular to 001)	100	Long.	$(C_{11}/2\rho)^{1/2}$	$V_1=V_L$
	001	Shear	$(C_{44}/\rho)^{1/2}$	$V_2=V_{S1}$
	010	Shear	$((C_{11}-C_{12})/2\rho)^{1/2}$	$V_3=V_{S2}$
At angle θ with the unique axis of the crystal		Long.	$[\{C_{33}Cos^2\theta+C_{11}Sin^2\theta+C_{44}+\{[C_{11}Sin^2\theta-C_{33}Cos^2\theta+C_{44}(Cos^2\theta-Sin^2\theta)]^2+4Cos^2\theta Sin^2\theta(C_{13}+C_{44})^2\}^{1/2}\}/2\rho]^{1/2}$	$V_1=V_L$
		Shear	$[\{C_{33}Cos^2\theta+C_{11}Sin^2\theta+C_{44}-\{[C_{11}Sin^2\theta-C_{33}Cos^2\theta+C_{44}(Cos^2\theta-Sin^2\theta)]^2+4Cos^2\theta Sin^2\theta(C_{13}+C_{44})^2\}^{1/2}\}/2\rho]^{1/2}$	$V_2=V_{S1}$
		Shear	$[\{C_{44}Cos^2\theta+C_{66}Sin^2\theta\}/\rho]^{1/2}$	$V_3=V_{S2}$

Conclusion

Based on the above discussion is worthwhile to state that:

i. Present method to evaluate second order elastic constants involving many body interaction potential for hexagonal wurtzite crystal structured materials (alloys) is correct.
ii. All elastic constants and density are mainly the affecting factor for anomalous behaviour of ultrasonic velocity in these alloys.
iii. When a sound wave travels at 55° with the unique axis of these crystals then the average sound wave velocity is maximum. Since the Debye average velocity is calculated using the constituent velocities V_1, V_2 and V_3, hence a good resemblance in V_D implies that our calculated velocities are correct.
iv. The <001> direction is the direction of symmetry for these alloys as they have the same quasi-shear and pure shear wave velocities.
v. $Ag_{0.2}Zn_{0.8}$ alloy is more suitable for industrial and other uses, as it having highest elastic constants as well as wave velocity and lowest attenuation in comparison to other chosen alloys.
vi. The mechanical and ultrasonic properties of these alloys will not be better than pure Ag and Zn due to their low SOEC and high ultrasonic attenuation.
vii. The preliminary results obtained in this work can be used for further experimental investigation with pulse echo overlap (PEO) technique for ultrasonic measurements and with conventional analytic techniques such as polarizing microscopy, X-ray diffraction (XRD), surface tension, solid state nuclear magnetic resonance (NMR), scanning electron microscopy (SEM) and transmission electron microscopy (TEM).

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