

Acoustic wave propagation in Ni_3R ($R = \text{Mo, Nb, Ta}$) compounds

PRAMOD KUMAR YADAWA

Department of Applied Physics, AMITY School of Engineering and Technology, Bijwasan,
New Delhi 110 061, India

E-mail: pkyadawa@aset.amity.edu

MS received 30 July 2010; accepted 2 December 2010

Abstract. The ultrasonic properties of the hexagonal closed packed structured Ni_3Mo , Ni_3Nb and Ni_3Ta compounds were studied at room temperature for their characterization. For the investigations of ultrasonic properties, the second-order elastic constants using Lennard–Jones potential were computed. The 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 respect to angle with the unique axis of the crystal. The inconsistent behaviour of angle-dependent velocities is associated with the action of second-order elastic constants. Debye average sound velocities of these compounds increase with the angle and has maximum at 55° with the unique axis at room temperature. Hence, when a sound wave travels at 55° with the unique axis of these materials, the average sound velocity is found to be maximum. The results achieved are discussed and compared with the available experimental and theoretical results.

Keywords. Elastic constants; longitudinal waves; surface waves; ultrasonic properties.

PACS Nos 43.35.cg; 62.20.Dc; 63.20.Kr

1. Introduction

Ultrasonic properties offer the possibility to detect and characterize microstructural 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 in the ultrasonic properties of metals and compounds [1–4]. Wave propagation velocity is the 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 the nature of atomic bonding forces and related hardness [5,6].

The crystal structure sequence for d transition metals at ambient pressure was accurately accounted for by means of *ab initio* local-density calculations almost three decades ago [7]. From electronic band structure calculations [8], Ni was found to be stable in a

face-centred-cubic (fcc) structure. Experimentally, however, ion implantation and ion beam mixing studies demonstrated that Ni could possess the hcp structure [9,10]. The formation and thermal stability of the hcp phase in the Ni–Mo, Ni–Nb and Ni–Ta multilayered films were investigated using solid-state reaction (SSR) method by Zhang *et al* [11]. Using the linear muffin-tin orbital approach, the total energy and the formation energy of the Ni₃Mo, Ni₃Nb and Ni₃Ta alloys of different crystal structures were calculated by Zhang *et al* [12].

In the present investigation, ultrasonic properties like orientation-dependent sound velocities and second-order elastic constants for the Ni₃Mo, Ni₃Nb and Ni₃Ta compounds are evaluated and discussed.

2. Theory

2.1 Second-order elastic constants

The second-order elastic constants (C_{IJ}) of the material are defined using the following expression:

$$C_{IJ} = \frac{\partial^2 U}{\partial e_I \partial e_J}, \quad I \text{ or } J = 1, \dots, 6, \quad (1)$$

where U is the elastic energy density, $e_I = e_{ij}$ (i or $j = x, y, z, I = 1, \dots, 6$) is the component of strain tensor. Equation (1) leads six second-order elastic constants (SOEC) for the hexagonal closed packed structured materials [13,14]

$$\begin{aligned} C_{11} &= 24.1p^4C', & C_{12} &= 5.918p^4C', \\ C_{13} &= 1.925p^6C', & C_{33} &= 3.464p^8C', \\ C_{44} &= 2.309p^4C', & C_{66} &= 9.851p^4C', \end{aligned} \quad (2)$$

where $C' = \chi a/p^5$, $\chi = (1/8)[\{nb_0(n-m)\}/\{a^{n+4}\}]$, $m, n =$ integer quantity; $b_0 =$ Lennard–Jones parameter, $p = c/a$: axial ratio; a and c are the unit cell parameters.

2.2 Ultrasonic velocity

The anisotropic behaviour of the material can be understood by knowing ultrasonic velocity because velocity is related to the second-order elastic constants [14]. Depending on the mode of atomic vibration, there are three types of velocities (longitudinal, quasi-shear and shear) in acoustical region [15]. These velocities vary with the direction of propagation of wave from the unique axis of hexagonal structured crystal [16,17]. The ultrasonic velocities as a function of the angle between the direction of propagation and unique axis for hexagonal structured materials are [18]

$$V_1^2 = \{C_{33} \cos^2 \theta + C_{11} \sin^2 \theta + C_{44} + \{[C_{11} \sin^2 \theta - C_{33} \cos^2 \theta + C_{44}(\cos^2 \theta - \sin^2 \theta)]^2 + 4 \cos^2 \theta \sin^2 \theta (C_{13} + C_{44})^2\}^{1/2}\} / 2\rho, \quad (3)$$

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

$$V_3^2 = \{C_{44} \cos^2 \theta + C_{66} \sin^2 \theta\} / \rho, \quad (5)$$

where V_1 , V_2 and V_3 are longitudinal, quasi-shear and pure shear wave ultrasonic velocities. The 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)

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

Here

$$V_D = \left\{ \frac{1}{3} \left(\frac{1}{V_1^3} + \frac{1}{V_2^3} + \frac{1}{V_3^3} \right) \right\}^{-1/3}, \quad (7)$$

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

The above formulae have been used for the evaluation of ultrasonic velocity and related parameters for selected materials.

3. Results and discussion

The unit cell parameters a , for Ni_3Mo , Ni_3Nb and Ni_3Ta are 2.584 Å, 2.623 Å and 2.604 Å, and the axial ratios p are 1.63, 1.629 and 1.629, respectively [12]. The Lennard–Jones parameter b_0 is evaluated under equilibrium condition using suitable values of m , n and lattice parameter, such that calculated elastic constants approach experimental values. The evaluated b_0 value for these alloys (for $m = 6$, $n = 7$) is 3.65×10^{-65} erg cm⁷. The second-order elastic constants and bulk modulus (B) of these compounds are presented in table 1.

The second-order elastic constants (SOEC) given in table 1 are calculated using eq. (2). The elastic constants are important as they are related to hardness and are used for determining ultrasonic velocity. The bulk modulus for these compounds can be calculated using

Table 1. Second-order elastic constants (SOEC) and bulk modulus (B) in unit of 10^{10} Nm⁻² of Ni_3R ($R = Mo, Nb, Ta$) compounds at room temperature.

Compounds	C_{11}	C_{12}	C_{13}	C_{33}	C_{44}	C_{66}	B
Ni_3Mo	35.58	8.74	7.55	36.10	9.06	13.95	17.22
Ni_3Nb	32.96	8.09	6.99	33.36	8.38	12.93	15.93
Ni_3Ta	30.65	7.53	6.50	31.02	7.79	12.02	14.82

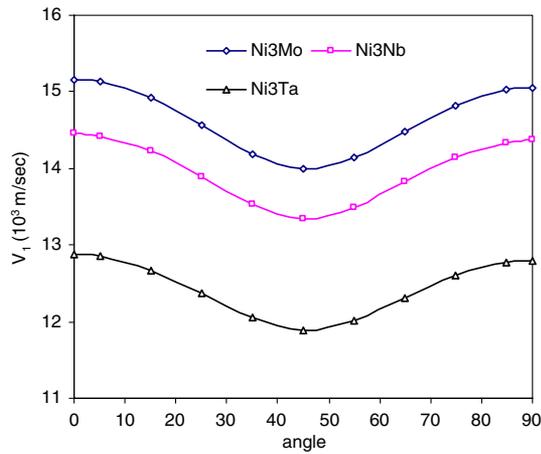


Figure 1. V_1 vs. angle with the unique axis of the crystal.

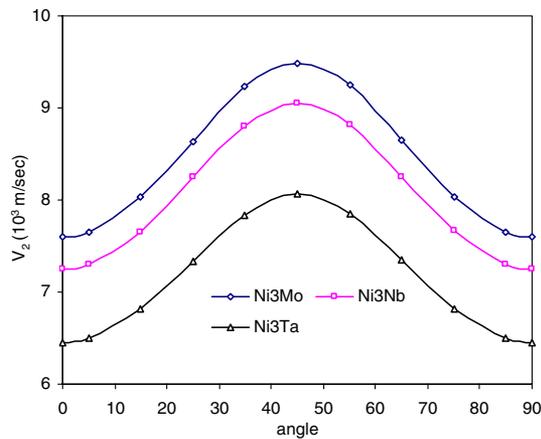


Figure 2. V_2 vs. angle with the unique axis of the crystal.

the formula $B = 2(C_{11} + C_{12} + 2C_{13} + (C_{33}/2)) / 9$ [18]. The evaluated bulk modulus (B) for Ni_3Nb using the presently calculated second-order elastic constants at 300 K is $15.93 \times 10^{10} \text{ Nm}^{-2}$. The experimental value of the bulk modulus for the same compound is $15.08 \times 10^{10} \text{ Nm}^{-2}$ [19]. Hence our theoretical approach to evaluate elastic constants seems valid for these compounds.

The computed orientation dependent ultrasonic velocities of these materials are given in figures 1–4. Figures 1–3 show 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 the angle-dependent velocities is correlated with the activity of SOEC. The tendency of the angle-dependent velocity curves (figures 1–4) reveals the same nature of angle-dependent curves for hexagonal wurtzite structured

Acoustic wave propagation in Ni_3R ($R = Mo, Nb, Ta$) compounds

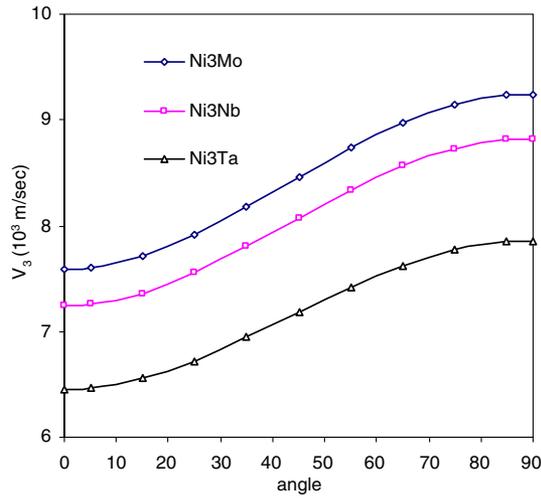


Figure 3. V_3 vs. angle with the unique axis of the crystal.

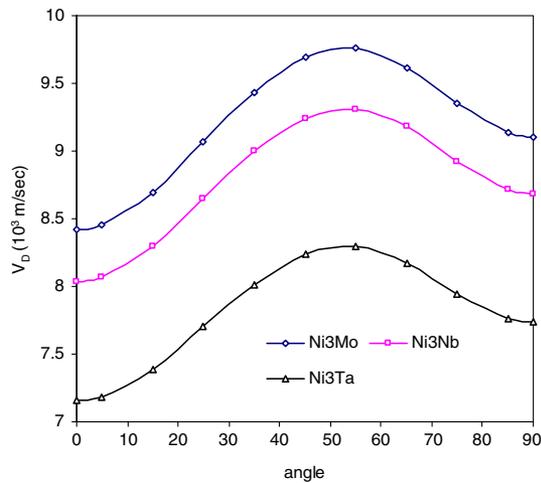


Figure 4. V_D vs. angle with the unique axis of the crystal.

materials GaN, AlN, InN, CdS, CdSe and laves-phase compounds [14,16–18,20,21]. Hence it is in conformity with the hcp structure of the chosen materials.

Figures 1–4 depict that the longitudinal ultrasonic velocity along $\langle 001 \rangle$ direction ($\theta = 0^\circ$ with unique axis) is largest for Ni_3Mo , because Ni_3Mo has the highest C_{33} value. Shear wave is also known as surface wave. Therefore, the ultrasonic velocities V_2 and V_3 are the surface wave velocities. It is clear that the values of V_2 and V_3 for ultrasonic wave propagation along $\theta = 0^\circ$ are the same, whereas these are different for other directions as visualized in figures 2 and 3. Hence $\langle 001 \rangle$ direction is the direction of symmetry for the

chosen materials. Debye average velocities (V_D) of these compounds increase with the angle and have maxima at 55° at 300 K (figure 4). Since V_D is calculated using V_1 , V_2 and V_3 values (eq. (7)), the temperature variation of V_D follows the combined effect of temperature variations 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. It can be seen from figures 1–4 that Ni_3Mo has the maximum velocity and Ni_3Ta has the least velocity for all angles of the crystals. Hence Ni_3Mo is more ductile and stable, and contains few defects in the crystal structure in comparison to other compounds.

4. Conclusions

Based on the above discussion it is worthwhile to state that

- Present method to evaluate second-order elastic constants involving many-body interaction potential for hexagonal wurtzite crystal structured materials is correct.
- Elastic constants and density are mainly the influencing factor for anomalous behaviour of ultrasonic velocity in these compounds.
- When a sound wave travels at 55° with the unique axis of these crystals, 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 , a good agreement in V_D implies that our calculated velocities are correct.
- The $\langle 001 \rangle$ direction is the direction of symmetry for these compounds as they have the same quasi-shear and pure shear wave velocities.
- Ni_3Mo compound is more suitable for industrial and other uses, as it has the highest elastic constant and wave velocity in comparison to other chosen compounds.
- The mechanical and ultrasonic properties of Ni_3Mo will be better than the other compounds due to their high SOEC and ultrasonic velocity.

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).

Acknowledgements

The author wishes to express sincere thanks to Dr Ashok K Chauhan, President, AMITY School of Engineering and Technology (ASET), New Delhi, Prof. B P Singh, Senior Director, ASET, New Delhi, Prof. S K Kor and Prof. Raja Ram Yadav, Department of Physics, Allahabad University, for encouraging and providing necessary facilities in carrying out the work.

References

- [1] K D Chaudhary, *Z. Phys.* **A155**, 290 (1959)
- [2] R R Yadav, A K Gupta and D Singh, *J. Phys. Stud.* **9**, 227 (2005)
- [3] S K Kor, U S Tandon and G Rai, *Phys. Rev.* **B6**, 2195 (1972)
- [4] S K Kor and R K Singh, *Acta Phys. Pol.* **A83**, 751 (1993)
- [5] R R Yadav and D Singh, *Intermetallics* **9**, 189 (2001)
- [6] D Singh and P K Yadawa, *Platinum Metals Rev.* **54**, 172 (2010)
- [7] H L Skriver, *Phys. Rev.* **B31**, 1909 (1985)
- [8] D G Pettifor, *CALPHAD: Comput. Coupling Phase Diagrams Thermochem.* **1**, 305 (1977)
- [9] G Was, *Prog. Surf. Sci.* **32**, 211 (1989)
- [10] Z J Jhang, H Y Bai, Q L Qiu, T Yang, K Tao and B X Liu, *J. Appl. Phys.* **73**, 1702 (1993)
- [11] Z J Zhang, X Y Huang and Z X Zhang, *Acta Mater.* **46**, 4189 (1998)
- [12] Z J Zhang and X Y Huang, *Phys. Rev.* **B58**, 1185 (1998)
- [13] P K Yadawa, D Singh, D K Pandey and R R Yadav, *The Open Acoustic J.* **2**, 61 (2009)
- [14] A K Yadav, R R Yadav, D K Pandey and D Singh, *Mat. Lett.* **62**, 3258 (2008)
- [15] C Oligschleger, R O Jones, S M Reimann and H R Schober, *Phys. Rev.* **B53**, 6165 (1996)
- [16] D K Pandey, D Singh and R R Yadav, *Appl. Acoust.* **68**, 766 (2007)
- [17] S K Verma, R R Yadav, A K Yadav and B Joshi, *Mat. Lett.* **64**, 1677 (2010)
- [18] D K Pandey, P K Yadawa and R R Yadav, *Mat. Lett.* **61**, 4747 (2007)
- [19] P Ravindran, G Subramoniam and R Asokamani, *Phys. Rev.* **B53**, 1129 (1996)
- [20] D K Pandey, D Singh and P K Yadawa, *Platinum Metals Rev.* **53**, 91 (2009)
- [21] D K Pandey, P K Yadawa and R R Yadav, *Mat. Lett.* **61**, 5194 (2007)