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THE STUDY OF EFFECT OF HIGH HYDROSTATIC PRESSURE ON THE MECHANICAL PROPERTIES OF Al-Cu BINARY ALLOYS A

ИССЛЕДОВАНИЕ ВЛИЯНИЯ ВЫСОКОГО ГИДРОСТАТИЧЕСКОГО ДАВЛЕНИЯ НА МЕХАНИЧЕСКИЕ СВОЙСТВА БИНАРНЫХ СПЛАВОВ Al-Cu

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Abstract. In this paper, computer simulations of influence of high hydrostatic pressure on the mechanical properties such as elastic constants and moduli, intrinsic hardness and acoustic velocities of *Al*, *Cu*, $CuAl_3$ and $AlCu_3$ are provided. To simulate the energy of interaction in metals and alloys, the Sutton-Chen inter-atomic potential was used. The simulation was run using the geometry optimization method with the General Utility Lattice Program (GULP) 5.1. With increment of hydrostatic pressure, the values of mechanical characteristics increased sharply. The highest percentage of increase in the in the mechanical properties was shown in the pressure step from 0 to 100 GPa. On the pressure range [0, 100], the highest percentage of increase was shown on elastic constant C_{44} while the lowest percentage of increase was on the transversal acoustic velocity for aluminuim. As the amount of aluminium in the alloys increases, the longitudinal acoustic velocity reduced, while the elastic constants and moduli, as well as intrinsic hardness, increased.

Keywords: high hydrostatic pressure, bulk modulus, shear modulus, elastic constant, intrinsic hardness, acoustic velocity.

Аннотация. В этой статье представлено компьютерное моделирование влияния высокого гидростатического давления на механические свойства, такие как константы и модули упругости, собственная твердость и акустические скорости, металлов и сплавов *Al, Cu*, $CuAl_3$ и $AlCu_3$. Для моделирования энергии взаимодействия в металлах и сплавах применялся межатомный потенциал Саттона-Чена. Моделирование проводилось с использованием метода оптимизации геометрии с помощью программы General Utility Lattice Program (GULP) 5.1. Моделирование показало, что с увеличением гидростатического давления значения

механических характеристик резко возрастают. Наибольший процент повышения механических свойств был выявлен при шаге давления от 0 до 100 ГПа. В диапазоне давлений [0, 100] ГПа самый высокий процент увеличения получен для константы упругости C_{44}, а самый низкий – для поперечной акустической скорости в алюминии. С увеличением количества алюминия в сплавах продольная скорость звука уменьшается, а постоянные и модули упругости, а также собственная твердость увеличиваются.

Ключевые слова: высокое гидростатическое давление, модуль объемного сжатия, модуль сдвига, константы упругости, собственная твердость, скорость звука.

Introduction

For generations, metallurgists and materials scientists have been working to develop new materials that are stronger, stiffer, more ductile, and lighter than existing high-temperature materials. Aluminium's application has been limited due to its low strength and poor corrosion resistance. Certain alloying elements can improve both of these qualities. Aluminium-based intermetallic alloys have had their surface hardness increased by incorporating iron, copper, and nickel [1]. It is therefore critical to investigate the effects of high hydrostatic pressure on aluminium and copper alloys.

Because measuring elastic modulus of solids at high pressure is difficult, little is known about elasticity of solids at high pressure. Ultrasonic technology and Brillouin spectroscopy are two technologies used to measure mechanical properties under the influence of pressure [2, 3]. These are used for modest pressure ranges. Ultrasonic measurements are usually confined to a few gigapascals, while Brillouin spectroscopy has been used up to a pressure of 25 GPa [2, 3].

There are recent advances in computing power and empirical potentials that reasonably model the energy of interaction among the atoms and molecules that make up the material. Due to these facts computer modelling is becoming increasingly relevant in researching material properties. Simulation techniques can provide a greater understanding of material properties when experimental procedures are difficult or impossible to carry out. Simulation approaches are now widely used to research material properties at the nano-scale, starting at the atomic level.

It's crucial to know how structural materials perform in extreme conditions like high pressure and temperature if you want to make materials that are safe and dependable. One of the key areas of research that solid state modelling is focused on is the variation of material properties under different conditions [4]. Guler and his co-authors studied the influence of high hydrostatic pressure on Gold *Au* using geometry optimization [2]. Using the first-principles method, Kimizuka et al. [3] investigated the influence of high pressure on elastic constants of alpha-quartz. Hieu and Ha [5] looked at the melting curves of silver, gold, and copper at high pressures.

The goal of this research is to see how high pressure affects the mechanical properties of materials *Al*, *Cu*, $AlCu_3$ and $CuAl_3$. The effect of high hydrostatic pressure on mechanical characteristics such as elastic constants and moduli, intrinsic hardness, and the lattice parameter will be investigated.

1. Mathematical Model and Method of Simulation

A number of inter-atomic potentials are used to represent the energy of atomic interaction in metals and alloys. Because of its simplicity and accuracy in describing the energy of atomic interactions in metals and alloys, the Sutton-Chen mathematical model was chosen for this study. The

total potential energy of interatomic interactions in the Sutton-Chen model is expressed as follows [6, 7]:

$$U_{tot} = \sum_i U_i = \sum_i \left[\frac{1}{2} \sum_{i \neq j} \varepsilon_{ij} \left(\frac{a_{ij}}{r_{ij}} \right)^{n_{ij}} - c\varepsilon_{ij} \left(\sum_{i \neq j} \left(\frac{a_{ij}}{r_{ij}} \right)^{m_{ij}} \right)^{\frac{1}{2}} \right], \quad (1)$$

where the first term in equation (1) implies a pair-wise long-range Van der Walls interaction between the atomic cores *i* and *j*. The square root term introduces the many body cohesive term in relation to the atom *i*, whereas the second term describes a many body component into the energy summation. Note that r_{ij} is the separation distance between the atoms *i* and *j*, *a* is a lattice parameter with a length dimension, $c > 0$ is a dimensionless parameter that scales the cohesive term in relation to the repulsive term, ε is an energy parameter, and *n* and *m* are integer material parameters with the property $n > m$ in equation (1).

A general utility lattice program (GULP) was used to run the simulation [8]. The simulation code is so versatile that can be used for wide ranging simulation from 0-D (molecules and clusters) to 3-D (periodic solids) for both with and without boundary conditions [8]. The parameters for the pure metals are given in table 1 [9] while for the alloys they are calculated using the mixing rules given in equation (2) [10].

Table 1 – The parameters of the Sutton-Chen potential for the metals *Al* and *Cu* [9, 11]

Metal	ν	μ	ε (eV)	C	*a* (Å)
Cu	6	9	$1.2382 \cdot 10^{-2}$	39.432	3.6100
Al	6	7	$3.3147 \cdot 10^{-2}$	16.39	4.05

$$\varepsilon_{ij} = \sqrt{\varepsilon_i \varepsilon_j}\ ;\ a_{ij} = \frac{a_i + a_j}{2}\ ;\ n_{ij} = \frac{n_i + n_j}{2}\ ;\ m_{ij} = \frac{m_i + m_j}{2}\ . \quad (2)$$

The metals *Al* [11] and *Cu* [9] have face centered cubic (fcc) crystal lattice structure while the alloys $CuAl_3$ and $AlCu_3$ have $L1_2$ structure [12]. The figure for the $L1_2$ structure is given in the article [7]. For the metals and alloys utilized in the simulation, the unit cell is employed to generate atomic coordinates for the initial configuration of crystal lattice structure. For instance, the unit cell of alloy $AlCu_3$ has the following basis vectors: *Al*: *a*(0:0; 0:0; 0:0); *Cu*: *a*(0:5; 0:5; 0:0); *Cu*: *a*(0:0; 0:5; 0:5); and *Cu*: *a*(0:5; 0:0; 0:5). The edge of an elementary cube is represented by a. The supercell was built with 108 atoms by translating the fcc unit cell in the three direction.

The simulations were run with the number of particles *N* and pressure *P* remaining constant. Periodic boundary conditions were also implemented. The simulation was run at 0 Kelvin for pressures ranging from 0 to 500 GPa with a 100 GPa incremental step size. The simulation pressure range was determined based on Dubrovinsky and his co-authors' findings [13]. Chemically and structurally similar elements to *Au*, such as *Cu*, *Ag*, and *Pt*, are not expected to phase transition up to 500 GPa, according to the researchers.

2. Mechanical properties of Metals and Alloys

The stability, stiffness, brittleness, ductility, and anisotropy of a material can all be determined using elastic constants [14]. The elastic constants must be calculated precisely in order to obtain

insight into the mechanical strength of solids, check their stability, and design material for specific applications [15]. Calculating the values of elastic moduli such as bulk and shear moduli also requires the use of elastic constants. Elastic constants are calculated using a semi-empirical potential that reflects the interaction energy among the atoms of metals and alloys, as shown below [3]:

$$C_{ij} = \frac{1}{V}\frac{\partial^2 U}{\partial \varepsilon_i \partial \varepsilon_j}, \tag{3}$$

where C_{ij} is a component of the stiffness matrix C, U is the energy expression, V is the volume of the unit cell, ε_i and ε_j are strain. The elastic constants expression given above is valid under adiabatic conditions and at zero pressure, and can be adjusted as follows to include external pressure, P [4]:

$$C_{\alpha\beta\gamma\zeta} = \frac{1}{V}\frac{\partial^2 U}{\partial \varepsilon_{\alpha\beta}\partial \varepsilon_{\gamma\tau}} + \frac{P}{2}\left(2\delta_{\alpha\beta}\delta_{\gamma\tau} - \delta_{\alpha\tau}\delta_{\beta\tau} - \delta_{\alpha\gamma}\delta_{\beta\tau}\right). \tag{4}$$

The following equation can be used to calculate the values of these independent elastic constants in terms of inter-atomic potentials [16]:

$$C_{11} = \frac{1}{V}\frac{\partial^2 U}{\partial \varepsilon_{11}^2};\ C_{12} = \frac{1}{V}\frac{\partial^2 U}{\partial \varepsilon_{11}\partial \varepsilon_{12}};\ C_{44} = \frac{1}{4V}\frac{\partial^2 U}{\partial \varepsilon_{12}^2}. \tag{5}$$

For isotropic polycrystalline materials, the bulk modulus B and shear modulus G can be estimated from elastic constants C_{ij} [15]. The formulas for estimation of bulk and shear moduli due to the Voigt V and Ruess R approximations are described in the article by Desta and his co-authors [7]. Young's modulus E is computed from the values of the bulk and shear moduli and the formula is given [7].

Hardness, which characterizes a material's resistance to deformation when an external force is applied to it, is one of the most important material properties. A material's intrinsic hardness relates to its resistance to deformation and corrosion. It's worth noting that real and intrinsic hardness are inextricably linked. To calculate the intrinsic hardness of metals and alloys, a variety of empirical formulas are utilized. The papers provide adequate information on basic formulas for estimating hardness for interested readers [16, 17]. To compute intrinsic hardness of the metals and their alloys we have used empirical relationship that represents hardness as a non-linear function of bulk and shear moduli described in the article by Desta and his co-authors [7].

Mechanical stability, phase transition, dynamic fracture, and other phenomena require an understanding of elastic wave propagation at high pressure [18]. Acoustic velocity, often known as sound speed, is the rate at which a tiny disturbance propagates across a specific material medium. Acoustic velocity measurements provide information on the properties of both artificial and natural materials. They're crucial when it comes to deciphering seismic data. Acoustic velocity measurements are the usual method for determining the structure and composition of the Earth's interior [19]. The values of the transverse V_t and the longitudinal V_l velocities of the material are computed using the formulas given [7].

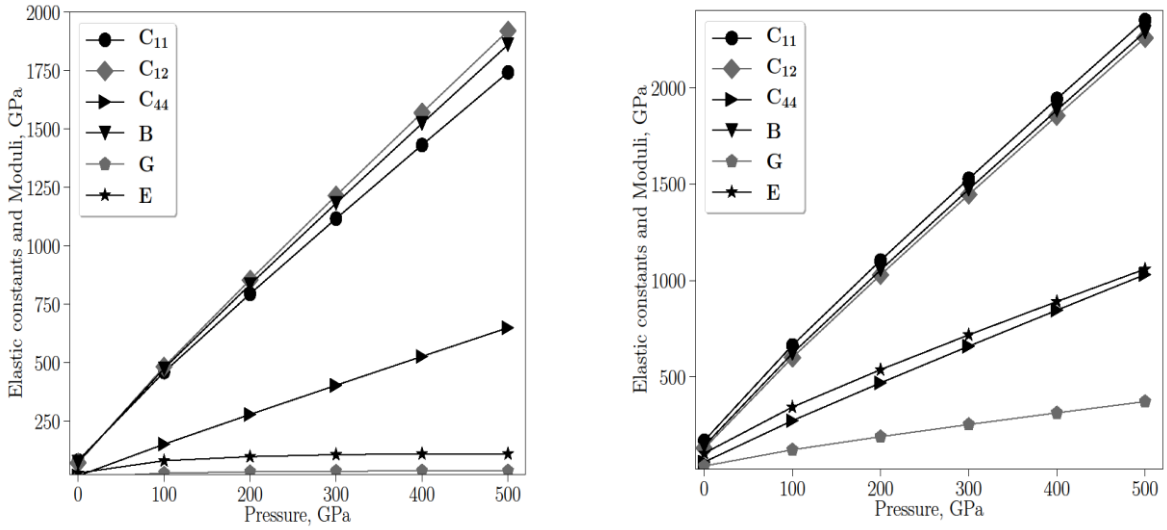
4. Results and Discussion

In the simulation, the geometry optimization method employing the Newton-Raphson technique was applied. The elastic constants were computed using equation (5) for the optimized structure. The Hill method of approximation was used to predict the bulk and shear moduli. The simulation and calculation results are shown in Table 2 and in Figures 1 through 4.

Table 2 – Effects of high pressure P on lattice parameter a (in Å), elastic constants, moduli, hardness H_{VT} (in GPa) and acoustic velocities V_t and V_l (in km/s) for the alloy system *Al-Cu*

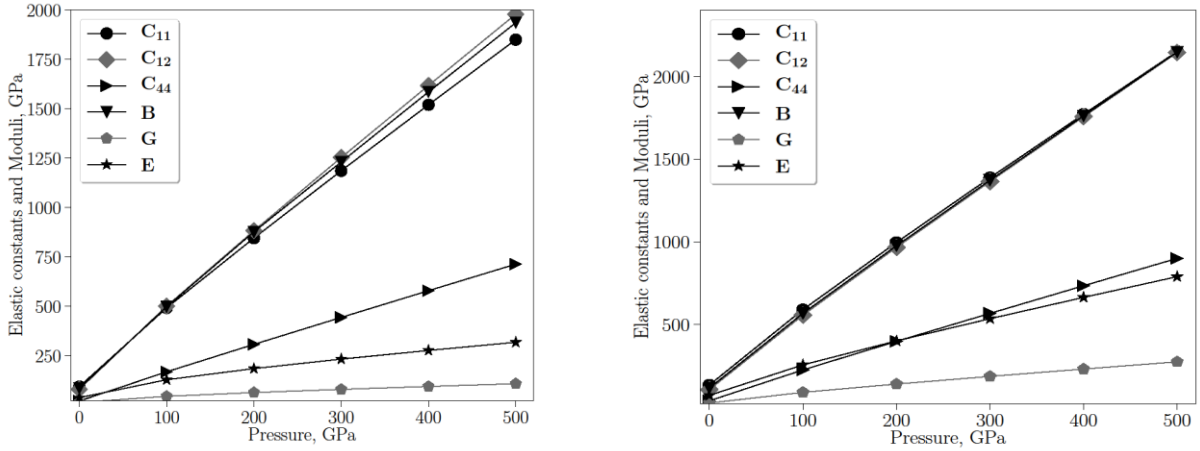
Metal	P	a	C_{11}	C_{12}	C_{44}	B	G	E	V_t	V_l	H_{VT}
Al	0	4.052	81.195	71.58	15.49	74.79	9.71	27.92	1.90	5.71	0.45
	100	3.52	460.16	482.62	151.27	475.13	27.34	80.47	2.58	11.14	0.37
	200	3.35	794.52	854.22	278.48	834.32	33.12	98.06	2.64	13.58	0.28
	300	3.25	1116.38	1214.82	403.31	1182.00	35.84	106.46	2.62	15.34	0.22
	400	3.18	1431.43	1569.35	526.82	1523.38	36.99	110.07	2.57	16.76	0.17
	500	3.12	1741.96	1919.93	649.48	1860.61	37.04	110.39	2.50	17.97	0.14
$CuAl_3$	0	3.95	93.28	79.70	19.96	84.23	12.97	37.00	1.83	5.11	0.67
	100	3.46	491.76	500.44	168.01	497.55	43.89	127.92	2.75	9.80	0.85
	200	3.30	845.03	882.04	306.88	869.71	62.93	184.34	3.07	11.97	0.87
	300	3.21	1185.80	1252.76	443.39	1230.44	79.12	232.38	3.30	13.55	0.90
	400	3.14	1519.84	1617.55	578.65	1584.98	93.89	276.23	3.48	14.83	0.92
	500	3.08	1849.56	1978.51	713.15	1935.53	107.81	317.53	3.63	15.93	0.95
$AlCu_3$	0	3.73	134.30	106.67	38.92	115.89	25.70	71.80	1.92	4.64	1.65
	100	3.34	590.20	556.68	225.64	567.85	89.90	256.17	3.04	8.42	2.73
	200	3.20	996.31	966.68	398.55	976.56	140.07	401.03	3.57	10.29	3.35
	300	3.12	1387.97	1364.96	567.55	1372.63	186.52	535.31	3.96	11.67	3.85
	400	3.06	1771.94	1756.96	734.52	1761.96	231.07	664.18	4.28	12.80	4.31
	500	3.01	2150.99	2144.96	900.24	2146.97	274.42	789.62	4.55	13.77	4.72
Cu	0	3.61	168.67	129.41	58.16	142.49	37.66	103.84	2.05	4.64	2.65
	100	3.28	663.55	600.16	272.85	621.29	121.93	343.34	3.19	8.09	4.33
	200	3.15	1103.85	1029.05	468.73	1053.99	189.85	537.29	3.76	9.86	5.38
	300	3.08	1527.40	1445.07	658.81	1472.51	252.92	717.68	4.18	11.18	6.24
	400	3.02	1941.88	1854.12	845.77	1883.38	313.40	890.78	4.52	12.25	7.01
	500	2.98	2350.36	2258.56	1030.65	2289.16	372.15	1059.07	4.82	13.18	7.71

As it can be observed from Table 2 and Figures 1 and 2, with an increase of pressure the elastic constants C_{11} and C_{12} as well as the bulk modulus B showed a very sharp linear increase for all the pure metals and their alloys. The Young modulus E and the elastic constant C_{44} showed a gradual increase in comparison with the bulk modulus and the elastic constants C_{11} and C_{12}. The change in the values of shear modulus with an increment of pressure showed very gradual increase in comparison to any other elastic constants or moduli. Especially for the pure metal aluminium, the trend of increase in the values of shear and Young moduli is almost stagnant when the pressure step reached 300 GPa and beyond.



a) b)

Figure 1 – Influence of high hydrostatic pressure on elastic constants and moduli: a) *Al*; b) *Cu*



a) b)

Figure 2 – Influence of high hydrostatic pressure on elastic constants and moduli:
a) $CuAl_3$; b) $AlCu_3$

The values of the lattice parameter *a* showed a very gradual decrease with an increase in pressure for both the pure metals and their alloys. The transversal and longitudinal acoustic velocities exhibited a gradual nonlinear increase with an increment of pressure for all the metals and their alloys, except for the transversal acoustic velocity for the pure aluminium metal. It non-linearly increased very gradually in the pressure range of [0, 200] GPa while the trend changed to non-linear very slow decrease in the pressure interval (200, 500]. It is worth noting that the trend of increase is much higher in the longitudinal acoustic velocity in comparison with the transversal sound velocity.

In general, with increasing pressure the mechanical properties of the metals increased with the exception of the intrinsic hardness of the pure aluminium metal. The biggest percentage of increase in the mechanical properties was seen in the pressure step from 0 to 100 GPa, while the

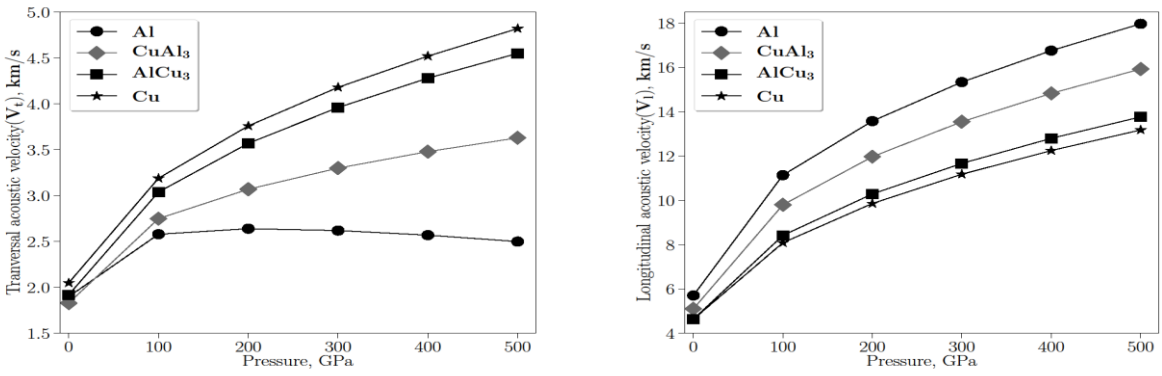
lowest percentage of increase was shown in the pressure range from 400 to 500 GPa. For instance, in the pressure range [0, 100], the elastic constant C_{44} had the biggest percentage of increase of an approximately 876 %, while the transversal acoustic V_t of metals had the lowest percentage of increase of 36 %. These values correspond to the values for pure aluminium metal. With increasing pressure, metals with a higher aluminum ratio showed a higher percentage of increase in the values of the mechanical properties, whereas metals with a lower aluminum ratio showed a decreasing trend in percentage of increase in the values of the mechanical properties.

The intrinsic hardness of the *Cu*, $CuAl_3$ and $AlCu_3$ increased non-linearly in a gradual manner. For aluminium, the intrinsic hardness showed a trend of decrease with increasing pressure. This may be attributed to the fact that the values of the shear modulus showed very gradual increase or remained almost constant with an increment of pressure.

With an increase in percentage of aluminium in the alloys, lattice parameter (a) and the longitudinal acoustic velocity kept decreasing while the elastic constants and moduli as well as intrinsic hardness kept increasing.

Table 3 – Comparing earlier data of *B*, *E*, *G*, V_t and V_l values for *Al* and *Cu* with our results at P = 0 GPa and T = 0 kelvin

Metal	Elastic parameters	Reference [21]	This study
Al	B (GPa)	79.38	29.29
	E(GPa)	78.24	27.92
	G(GPa)	29.29	9.71
	V_t (km/s)	3.274	1.90
	V_l (km/s)	6.583	5.71
Cu	B (GPa)	142.03	142.49
	E(GPa)	132.51	103.84
	G(GPa)	49.28	37.66
	V_t (km/s)	2.337	2.05
	V_l (km/s)	4.798	4.64



a) b)

Figure 3 – Influence high hydrostatic pressure on acoustic velocity:
a) transversal V_t; b) longitudinal V_l

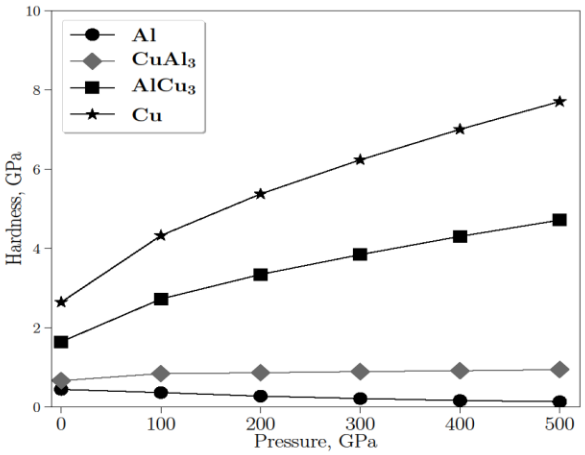


Figure 4 – Influence high hydrostatic pressure on intrinsic hardness

Conclusion

This study examines the effects of high pressure on the mechanical properties such as elastic constants and moduli, acoustic velocities and intrinsic hardness of the metals Al, Cu, $CuAl_3$ and $AlCu_3$.

The elastic constants C_{11}, C_{12}, C_{44} and the bulk modulus B are the most affected by high pressure. The highest percentage of increase in the mechanical properties of the metals was in the pressure range from 0 to 100 GPa. In the pressure range [0, 100], the highest percentage of increased was shown in the value of the elastic constant C_{44} for aluminium which is approximately 876 % while the lowest percentage of increase was show in the value of transversal acoustic velocity of aluminium which corresponds to approximately 36%. With increment of pressure, the value of an elastic constants or moduli increased sharply. With increment of pressure, the values of intrinsic hardness and acoustic velocities increased non-linearly increased with exception of the values of intrinsic hardness of the pure aluminium metal where in gradually decreased. High hydrostatic pressure has a much bigger effect on longitudinal acoustic velocity than on transversal acoustic velocity. The lattice parameter *a* and longitudinal acoustic velocity decreased as the proportion of aluminium in the alloys increased, whereas the elastic constants and moduli, as well as intrinsic hardness, increased. The study also demonstrates that computer modelling is an effective tool for studying the effects of high hydrostatic pressure in metals and alloys that goes beyond traditional methods.

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