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THE GEOMETRY OPTIMIZATION CALCULATIONS ON MECHANICAL PROPERTIES OF $L1_2$ STRUCTURE Al_3X AND AlX_3-TYPE (X = Au, Ag, Cu) INTERMETALLIC COMPOUNDS

ГЕОМЕТРИЧЕСКИЕ ОПТИМИЗАЦИОННЫЕ РАСЧЕТЫ МЕХАНИЧЕСКИХ СВОЙСТВ ИНТЕРМЕТАЛЛИЧЕСКИХ СОЕДИНЕНИЙ ТИПА Al_3X И AlX_3 (X = Au, Ag, Cu) С $L1_2$ СТРУКТУРОЙ КРИСТАЛЛИЧЕСКОЙ РЕШЕТКИ

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Abstract. In this work, computer simulation of mechanical properties such as elastic constants and moduli as well as intrinsic hardness of *Al*, Al_3X and AlX_3 having crystal lattice structure of the type $L1_2$ is presented. To describe the energy of interaction in metals and alloys, the Sutton-Chen semi-empirical inter-atomic potential was utilized. The simulation was run using the geometry optimization method with the General Utility Lattice Program (GULP) 5.1. From the six different alloys studied, the alloy with highest intrinsic hardness is $AlAg_3$ while with the lowest value for $CuAl_3$. The findings show that *Al*-based alloys have values of mechanical characteristics that are higher than the pure aluminium metal. The values of mechanical characteristics of the alloys are indirectly proportional to the percentage of aluminium in a given alloy system. The work further confirms that the percentage of aluminium in the alloy systems have significant impact on the mechanical properties.

Keywords: bulk modulus, shear modulus, elastic constant, intrinsic hardness.

Аннотация. В данной работе представлено компьютерное моделирование механических свойств, таких как константы и модули упругости, а также собственная твердость *Al*, Al_3X и AlX_3, имеющих структуру кристаллической решетки типа $L1_2$. Для описания энергии взаимодействия в металлах и сплавах использовался полуэмпирический межатомный потенциал Саттона-Чена. Моделирование проводилось с использованием метода оптимизации геометрии с помощью программы General Utility Lattice Program (GULP) 5.1. Из шести различных исследованных сплавов самую высокую внутреннюю твердость имеет $AlAg_3$, а наименьшую – $CuAl_3$. Полученные данные показывают, что сплавы на основе алюминия имеют более высокие значения механических характеристик, чем чистый металлический алюминий. Значения механических характеристик сплавов косвенно пропорциональны

процентному содержанию алюминия в данной системе сплавов. Работа также подтверждает, что процентное содержание алюминия в системах сплавов оказывает значительное влияние на механические свойства.

Ключевые слова: объемный модуль, модуль сдвига, постоянная упругости, собственная твердость.

Introduction

Aluminium's application has been limited due to its low strength and poor corrosion resistance. Certain alloying elements can improve both of these qualities. The high melting temperature, comparatively low density, superior oxidation resistance, increase in yield strength with increasing temperature, and extreme hardness of aluminium-based inter-metallic systems are all key qualities for possible technological applications [1]. It is imperative to find ways of improving the mechanical properties of aluminium by different means.

Aluminium (*Al*) is a light metal whose strength can be increased through alloying, mechanical, and thermal treatment, resulting in better mechanical properties [2]. Aluminium alloys are a significant class of materials due to their high technological value and wide range of applications, particularly in aerospace, microelectronics, motorized vehicles, and domestic industries.

The mechanical behavior of alloys has been a subject of different researches due to the importance of alloys in engineering and basic research [3]. *Al*-based alloys have been the focus of many researchers. Guan and his co-authors investigated alloying stability, electrical structure, and mechanical characteristics of *Al*-based inter-metallic using first-principles methods [3]. Tian and his co-authors studied elastic properties of *Al*-included with $L1_2$ crystal lattice structure using Ab initio calculations [4].

The purpose of this study is to calculate the mechanical properties of the *Al*-based inter-metallic compounds having $L1_2$ crystal lattice structure. The study will use the geometry optimization method to conduct a systematic assessment of the mechanical properties of the alloys. It will look at how different percentages of aluminum in alloys affect mechanical characteristics including elastic constants and moduli, as well as intrinsic hardness.

1. Computational Method

Atomistic simulation techniques such as molecular dynamics (MD) have become a powerful tool in the field of nanotechnology because they provide physical insight into various phenomena at the atomic scale and allow one to predict physical properties such as structure and thermodynamic properties of the nano-materials [5]. To elucidate the mechanical properties of aluminum alloys having crystal structures of the type $L1_2$, we performed geometry optimization simulations while keeping the number of particles *N*, temperature *T*, and pressure *P* constant.

In atomistic modelling simulations, the interactions between atoms or molecules must be defined accurately in order to predict crystal formations and physical properties of materials. The Sutton-Chen semi empirical atomic potential is one of the methodologies that has been successfully used to model the many-body interactions in metallic systems. The total potential energy is expressed as [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 is the pair-wise repulse potential between atoms *i* and *j*, whereas the second term describes a many body component into the energy summation. The separation distance between atoms *i* and *j* is r_{ij}. Note in equation (1), a is a length-dimensioned lattice parameter, $c > 0$ is a dimensionless parameter that scales the cohesive term with respect to the repulsive term, ε is an energy parameter, and *n* and *m* are integer material parameters with the property $n > m$.

The simulation was done using a general utility lattice program (GULP) of code 5.1 [8]. The simulation algorithm is so adaptable that it can be used for everything from 0-D (molecules and clusters) to 3-D (periodic solids) simulations, both with and without boundary conditions [8]. Table 1 [9] contains the parameters for pure metals, whereas equation (2) [10] can be used to compute the parameters for alloys.

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

The pure metals used in the simulation have face centered cubic (fcc) crystal lattice structure [9, 11] while the alloys of the type AlX_3 and XAl_3 have $L1_2$ structure (see Figure 1) [3]. The unit cell is used to create atomic coordinates for the initial configuration of the crystal lattice structure for the metals and alloys used in the simulation. For instance, the unit cell of alloy was constructed using the basis vectors: *Al*: *a*(0:0; 0:0; 0:0); *Au*: *a*(0:5; 0:5; 0:0); *Au*: *a*(0:0; 0:5; 0:5); and *Au*: *a*(0:5; 0:0; 0:5). The edge of a unit cell is represented by *a*. The supercell was constructed with 108 atoms by three-dimensionally translating the fcc unit cell. The simulation was carried out at $T = 0$ Kelvin and $P = 0$ GPa.

Table 1 – The parameters of the Sutton-Chen potential for the metals *Cu*, *Al*, *Au* and Ag [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
Au	8	10	$1.2793 \cdot 10^{-2}$	34.408	4.0800
Ag	6	12	$2.5415 \cdot 10^{-3}$	144.41	4.0900

2. Mechanical properties of Metals and Alloys

Elastic constants can be used to determine a material's stability, stiffness, brittleness, ductility, and anisotropy. Elastic constants (C_{ij}) are key parameters for predicting material physical characteristics and mechanical stability [12]. Elastic constants are also required when calculating the values of elastic moduli like bulk and shear moduli. As demonstrated below, elastic constants are determined using a second derivative of the semi-empirical potential with respect to strain [13]:

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

where U is the energy expression, C_{ij} is a component of the stiffness matrix C, V is the volume of the unit cell, ε_i and ε_j are strain.

For cubic systems, there are only three constants. The values of these independent elastic constants in terms of inter-atomic potentials can be calculated using the equation below [14]:

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

The bulk modulus B and shear modulus G of isotropic polycrystalline materials can be calculated using elastic constants C_{ij}. The formulas for calculating bulk and shear moduli using the Voigt V and Ruess R approximations are described in the articles [7, 15]. For fcc crystal structures, the value of the bulk modulus remains the same in the three methods of approximations. The Young modulus E is calculated from the values of bulk and shear moduli using the formula described in [7].

One of the most essential material qualities is hardness, which describes a material's resistance to deformation when an external force is applied to it. The intrinsic hardness of a material is related to its resistance to deformation and corrosion. Real and intrinsic hardness are intricately linked. A variety of empirical formulas are used to calculate the intrinsic hardness of metals and alloys. For interested readers, the papers [14, 16] contain sufficient information on basic formulas for determining hardness. The empirical relationship used in this study to depict hardness as a non-linear function of bulk and shear moduli is given the article by Desta and his co-authors [7].

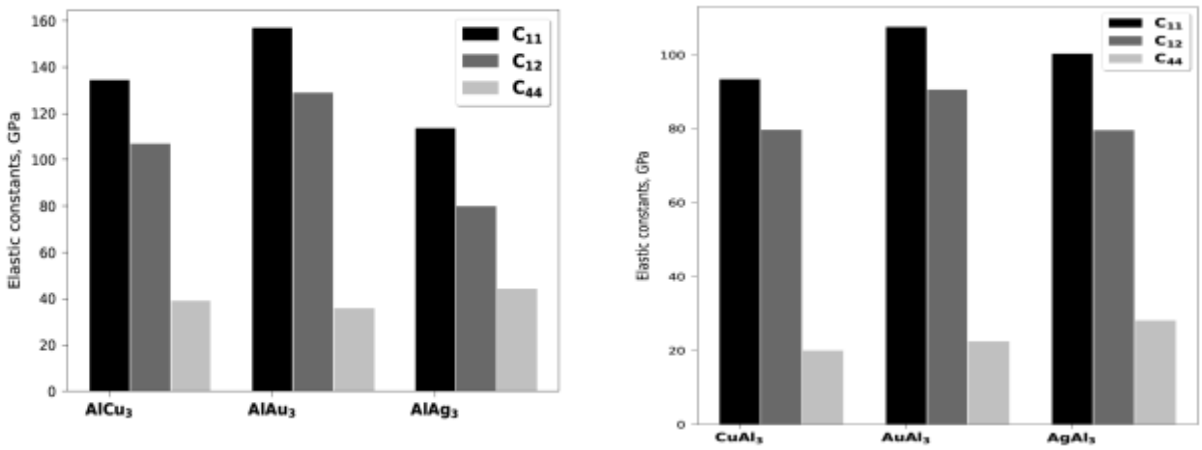
3. Results and Discussion

The geometry optimization method using the Newton-Raphson technique was used in the simulations. It's necessary to find an optimized geometric structure as a precondition for computing mechanical properties. After picking the optimal structure, the mechanical properties were determined using different relationships.

Table 2 – A comparison of geometry optimization calculations mechanical properties of *Al*-based alloys with $L1_2$ crystal lattice structure with earlier reported data. Lattice parameter a (in Å), elastic constants, moduli and hardness H_{VT} (in GPa)

Metal	a	C_{11}	C_{12}	C_{44}	B	G	E	H_{VT}	Ref.
Al	4.052	81.195	71.582	15.492	74.786	9.709	27.920	0.451	this study
$CuAl_3$	3.951	93.2846	79.6959	19.955	84.225	12.967	37.003	0.673	this study
$AuAl_3$	4.058	107.523	90.538	22.339	96.200	15.160	43.211	0.771	this study
$AgAl_3$	4.058	100.207	79.574	28.0148	86.452	18.775	52.522	1.292	this study
$AlCu_3$	3.728	134.295	106.686	38.924	115.889	25.7015	71.797	1.653	this study
		155.02	120.57	82.10	132.05	44.45			[3]
$AlAu_3$	4.073	156.801	128.743	35.675	138.095	24.538	69.4985	1.244	this study
		145.48	121.58	25.95	129.55	19.01			[3]
$AlAg_3$	4.189	113.431	79.715	44.121	90.953	30.003	81.092	2.897	this study
		106.38	87.88	44.65	94.05	24.07			[3]

For the optimized structure, the elastic constants were calculated using equation (4). The bulk and shear moduli were calculated using the Hill method of approximation. Given the values of the shear and bulk moduli, the Young's modulus was calculated using the equation in [7]. The intrinsic hardness of metals and alloys was approximated using the empirical expression presented in [7]. Table 2 and Figures 1 through 3 present the simulation and calculation findings.

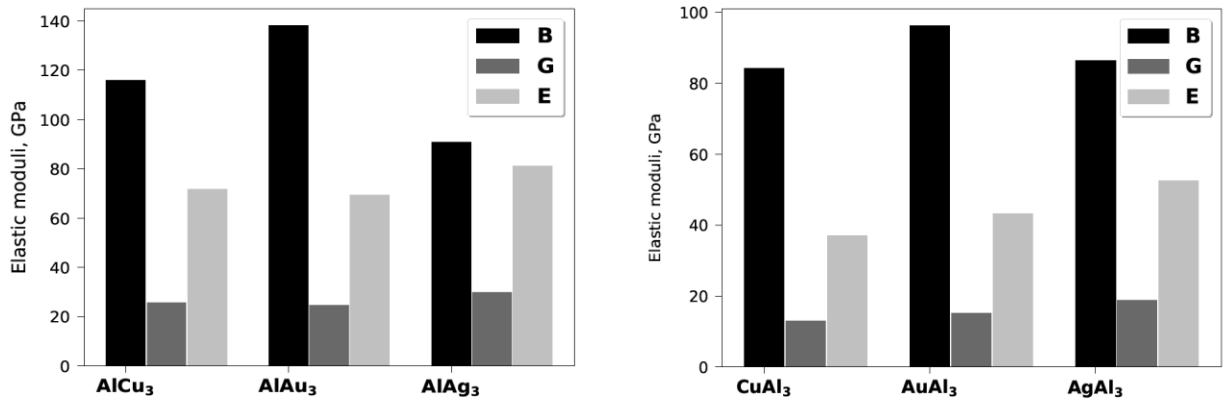


a) b)

Figure 1 – Influence of aluminium ratio on the elastic constants of *Al*-based alloys: a) AlX_3; b) XAl_3

The requirements of mechanical stability of materials having cubic crystal lattice are given: $C_{11}+2C_{12} > 0$; $C_{11} - C_{12} > 0$; $C_{44} > 0$. In our case all the alloys investigated in this work satisfied the criteria of mechanical stability.

As shown in Table 2 and Figures 1 through 3, except for the lattice parameter, all of the mechanical values computed here for aluminium based alloys are higher than that of *Al*. It's worth noting that we used two sets of alloys, one with 25% aluminium and the other with 75% aluminium. For 75% aluminium ratio, the values of the elastic constants and moduli, as well as intrinsic hardness, are given in increasing order as $CuAl_3 < AuAl_3 < AgAl_3$.



a) b)

Figure 2. Influence of aluminium ratio on the elastic constants of Al-based alloys:
a) AlX_3; b) XAl_3

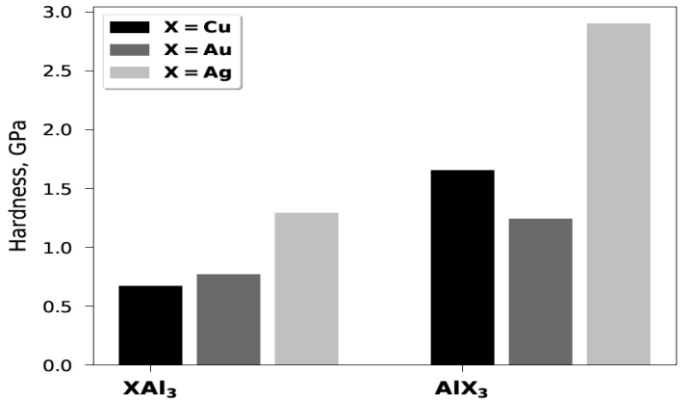


Figure 3 – Influence of aluminium ratio on the intrinsic hardness of *Al*-based alloys

On the other hand, for the alloy system with 25% aluminium, the trend is not similar to the above discussion. The elastic constants C_{11}, C_{12} and the bulk modulus (B) are arranged as $AlAg_3 < AlCu_3 < AlAu_3$ while for the elastic constant C_{44} as well as the shear and Young moduli is arranged as $AlAu_3 < AlCu_3 < AlAg_3$. The values of intrinsic hardness for the alloys system with 25% aluminium composition is given as $AlAu_3 < AlCu_3 < AlAg_3$ while for the alloy system with 75% aluminium composition the in equality for intrinsic hardness is given as $CuAl_3 < AuAl_3 < AgAl_3$.

For each alloy system investigated, with decrease of the percentage of aluminium in the material the mechanical properties increased. For instance, this is to say that the mechanical properties of $AlCu_3$ are lower than that of $CuAl_3$.

Conclusion

The work uses method of geometry optimization to compute mechanical properties of $AlCu_3$ and $CuAl_3$ alloys such as elastic constants and moduli, as well as intrinsic hardness. The mechanical properties of six distinct aluminium alloys were calculated, three of which had the form AlX_3 while the others had the formula XAl_3 where *X* stands for *Cu*, *Au* or *Ag*.

The computed findings demonstrate that all of the alloys have mechanical properties such as elastic constants and moduli, as well as intrinsic hardness, that are higher than pure aluminium metal. From the six different alloys studied, the alloy with highest intrinsic hardness is $AlAg_3$ while with the lowest value is $CuAl_3$. The values of the mechanical properties of the alloys are inversely proportional to the percentage of aluminium in the alloy.

The research demonstrates that the percentage of aluminium in the alloy has a very significant influence on the mechanical properties of a given alloy. By alloying elements like copper, gold, and silver with aluminium, superior alloys with improved mechanical properties can be produced. This creates alloys of aluminium with better mechanical properties that improves the drawbacks of the soft pure aluminium metal while extending the applications in diversified fields.

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