Computer Modelling & New Technologies, 2002, Volume 6, No.1, 47-50
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SILICON-ALUMINUM BONDING IN *Al* ALLOYS

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Ab initio calculation was performed to investigate the nature of Si-Al bonding in Al based alloys. Total electronic energy E_{tot} for different configurations of the model cluster Si_2Al_6 was calculated. When the model cluster consists of two perfect tetrahedrons there is a strong influence of the Si-Si distance on the Si-Al adiabatic potential. The equilibrium distance between Si and Al atoms increases with the length of Si-Si bond increasing. It was concluded that description of Si clusters in Al matrix demands an account of the angle depending part of Si-Al interaction.
Keywords: Si-Al alloys, *ab-initio* band calculations

1. Introduction

The nature of silicon-aluminum interaction is of great interest because Si-Al interatomic potentials are used for description of equilibrium configurations of Si precipitates in Al-based alloys. The important problem of nuclear industry is linked to peculiarities of these materials. Si precipitates are accumulated in Al-based alloys in conditions of reactor irradiation. Radiation-induced processes lead to degradation of reactor materials and to decreasing their exploitation recourse. Computer simulation of these processes allows to predict the changes of mechanical characteristics of Al-based alloys under reactor irradiation. The task is reduced to understanding mechanisms of Si precipitates formation and appearance of mechanical stresses in reactor constructions [1-5]. The Molecular Dynamics (MD) method gives a useful insight into these problems. But it is necessary to have adequate interatomic potentials for Si-Al interaction which depend on sizes and structure of Si clusters in Al lattice. In this paper we investigated Si-Al bonding in Al matrix. The model cluster (MC) consists of two Si atoms and six Al atoms (Figure 1).

We performed calculations of total-energy of MC using density-functional theory and pseudopotential approach [4-7]. Only the chemically active valence electrons are dealt with explicitly. The chemically inert core electrons are eliminated within the frozen core approximation [8], being considered together with the nuclei as rigid non-polarizeable ion cores. In turn, all electrostatic and quantum-mechanical interactions of the valence electrons with the ion cores (the nuclear Coulomb attraction screened by core electrons, Pauli repulsion and exchange-correlation between core and valence electrons) are accounted for by angular momentum dependent pseudopotentials. The valence electrons are described by smooth pseudo-orbitals, which play the same role as the true orbitals, but avoid the nodal structure near the nuclei that keeps core and valence states orthogonal in all -electron framework. The respective Pauli repulsion largely cancels the attractive parts of the true potential in the core region, and is built into therefore rather weak pseudopotential [9].

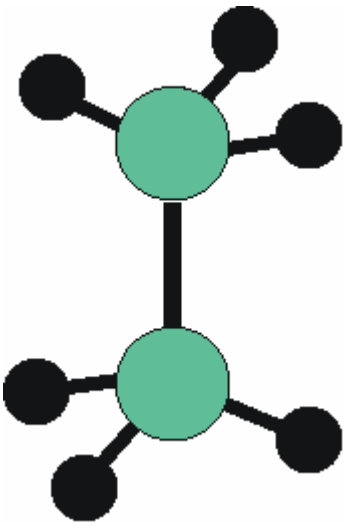

Figure 1. The tetrahedral configuration of model cluster: ● - Si atoms, ● - Al atoms

The scheme for the electronic energy minimization based on iteration of wave functions is the steepest descent approach [6]. A more efficient scheme is based on a second order equation of motion [7]. In our study the computer code FHI98MD [10] is used.

2. Description of the model

The covalent radius of Si is a little less than of Al [11]. A substitutional Si atom in Al lattice does not create visible microstresses but results a small decreasing of lattice parameter [12]. The situation changes in the case of formation of Si cluster in Al matrix. Even if two Si atoms are united to Si_2 molecule a tendency to form hybrid atomic bonds with neighbor atom is revealed. We have studied Si_2Al_6 cluster *(Figure 1)* in Al lattice which is a fragment of a diamond-like lattice consisting of two tetrahedrons. Si-Al adiabatic potential was calculated as a dependence of the total electronic energy E_{tot} of the cluster on the distance of Si-Al atoms. Characteristics of Si-Al bonds were changed when the distance between Si atoms increased. The disturbed tetrahedrons were chosen using correlation:

$$h_i = N(a_i s + b_i p_x + c_i p_y + d_i p_z) \ . \qquad (1)$$

Here s and p_x, p_y, p_z are corresponding $s-$ and p - orbitals. N is a normalizing coefficient. If to introduce the ratio between contributions of $2p$ - and $2s$ - orbitals to i-th hybrid orbital λ_i^2:

$$\lambda_i^2 = \frac{b_i^2 + c_i^2 + d_i^2}{a_i^2} \ , \qquad (2)$$

one can get a correlation [13]:

$$\cos\alpha_{ij} = -1/\lambda_i\lambda_j \qquad (3)$$

(3) leads to $\cos\alpha_{ij} \le 0$ and $\alpha_{ij} \ge 90^o$. It means that an angle between two sp^k - hybrid orbitals is not less than 90^o. So we have taken angles between chemical bonds from minimal value 90^o to tetrahedral ones $109^o58'$. Calculation of Si-Al interatomic potentials was performed by displacement of Al atoms along directions of Si-Al bonds as it is shown in *Figure1* by pointers. For each position of Al atom E_{tot} was calculated

3. Results and discussion

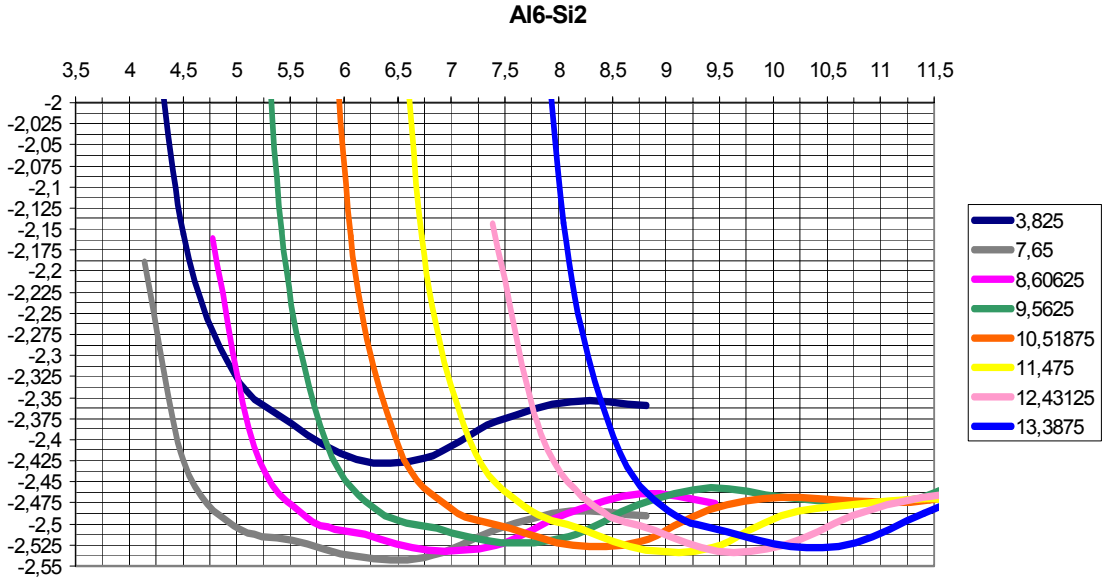

Figure 2. The typical adiabatic potentials

In the *Figure 2* one can see typical adiabatic potentials calculated for different lengths of Si-Si bond in the case of tetrahedral atom configuration of cluster (*Figure 1*). Calculation results represented in the *Figure 3* correspond to disordered tetrahedrons of the model cluster. It is seen that in the case of two ideal tetrahedrons r_{min} increases with increasing the length of Si-Si bond r_{Si}. This dependence is shown

in the Table 1. Si-Al bonds weaken when the length of Si-Si bond increases. In the case of disordered tetrahedrons r_{min} doesn't depend on the value r_{Si} (*Figure 3*).

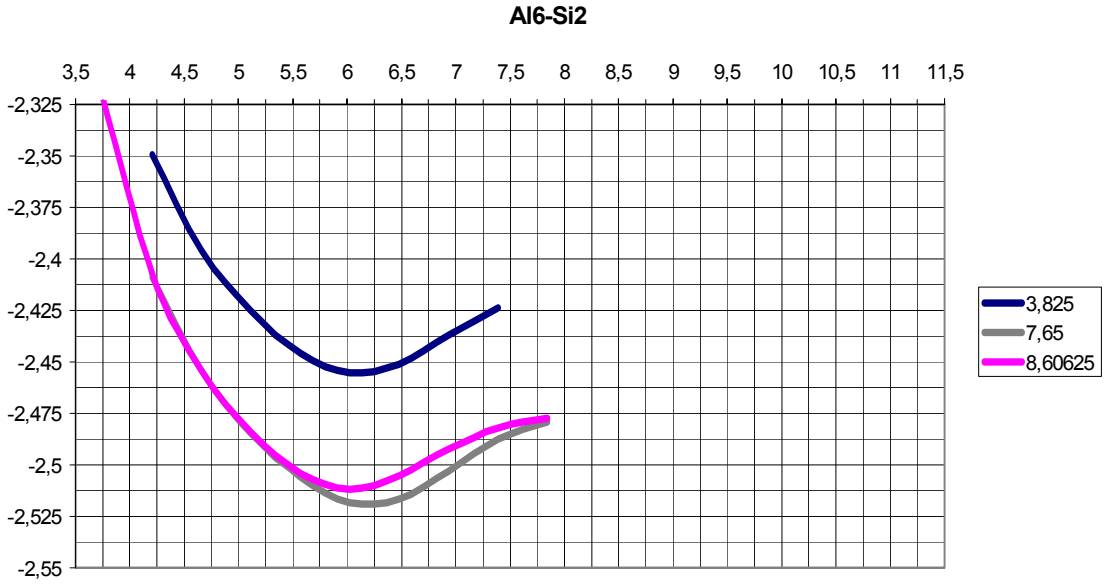

Figure 3. Adiabatic potentials for disordered Si- Al tetrahedrons of the model cluster

TABLE 1. Influence of Si-Si bond length on the minimum of Si-Al potential

r_{Si} , a.u.	3.83	7.65	8.61	9.56	10.52	11.48	12.43	13.39
r_{min} , a.u.	6.25	6.50	7.00	7.75	8.50	9.00	9.75	10.00

The obtained results show that Al atoms which are disposed around Si cluster participate in formation of tetrahedral sp^3-hybrid bonds. It seems that in metal matrix Al atoms can add an additional *p*-electron and interact with Si atoms forming orbitals, which are close to sp^3 - orbilals in Si. Thus Si-Al potentials, which are obtained for separate Si atoms in Al crystal, cannot be applied for Si clusters calculations in Al matrix. In the last case it is necessary to use three-particle potentials with orientation dependence. The ability to create hybrid bonds by Al atoms is mentioned in [14].

4. Conclusion

Ab initio calculation of total electronic energy for Si_2Al_6 cluster in Al matrix was performed. The adiabatic potentials for Si-Al interaction were calculated for different lengths of Si-Si bond. It was found that Al atoms participate in formation of sp^3 hybrid bonds with Si. This result shows that in the case of Si cluster formation in Al crystal the potentials, which are used for description of Si-Al interaction, must be angle depending.

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Received on the 17th of May 2002