

EFFECT OF Al ATOM FORCING OUT BY Si IN Al-Si SOLID SOLUTIONS

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It is shown by computer simulation of the Al-Si solid solution that at the first stages of Si precipitates formation the small Si clusters arise in the tetrahedral configurations. This process is accompanied by the effect of Al atom forcing out from the lattice site by one of the Si atoms of nuclei.

1. Introduction

One of essential results of reactor irradiation of pure Aluminum and Aluminum alloys to high neutron doses is Silicon precipitates formation [1-3]. Silicon is produced by means of reactions $^{27}\text{Al}(n,\gamma)\rightarrow^{28}\text{Al}\rightarrow^{28}\text{Si}\rightarrow\beta$. The cross section for Silicon formation is so large, that the amount of transmutation Silicon by far exceeds its solubility in Aluminum. Thus, it is clear that Silicon must precipitate and may be observed as visible particles. In vicinity of grain boundaries and other imperfections of Al crystal lattice Silicon dissolves better and these regions become supersaturated with Silicon, which precipitates into the large flakes [1]. Both quenching and nuclear reactor experiments allow to conclude that at the first stages of Si precipitates formation accumulations of few Si atoms appear with large probability. But initial configurations of these accumulations in the most cases have not the tetrahedral structure.

A treatment of Al-Si alloys at elevated temperatures ($> 500^\circ\text{C}$) in the quenching experiments leads to an intensive mixing of Si atoms in Al lattice. Diffusion coefficients for Si atoms in Al-Si alloys with concentration of Si about 0.5 at % and at temperatures near $500^\circ\text{C} \approx 10^{-9} \text{ cm}^2\text{s}^{-1}$ [2]. The mean distance between Si atoms in the Al-0.5at % Si alloy is $x < 30 \text{ \AA}$. This distance may be got over by Si atom for the time $t \approx x^2 / D \approx 10^{-4} \text{ s}$.

In [2] the specimens were treated at the 580°C during the 40 min. Si atom can get over the distance x about 10^7 times for this time. So the mixing of Si atoms in quenching experiments is really intensive. In the moment of quenching we have the random disposition of Si atoms in Al matrix and because of fluctuations of Si atoms density we may suggest that they have a space distribution with 0, 1, 2, 3, etc Si atoms in the given site of Al lattice. The probability to find one Si atom in the volume which contains three neighboring lattice nodes is $\sim 3 \cdot 10^{20}/10^{22} \sim 3 \cdot 10^{-2}$. The probability to find three Si atoms in the same volume is $(3 \cdot 10^{-2})^3 \sim 2.7 \cdot 10^{-5}$. So we obtained that $10^{20} \cdot 2.7 \cdot 10^{-5} \sim 2.7 \cdot 10^{15}$ Si atoms in the quenching moment find oneself in positions of Si_3 clusters. However these Si clusters may have at first distorted tetrahedral configurations with stressed chemical bonds. Such Si_3 configurations aspire to change the angles between chemical bonds and to bring nearer them to $\sim 109^\circ$. The aim of this paper is to explain how the distorted Si accumulations transform to the nuclei with tetrahedral configurations and later on form Si precipitates with diamond like structure.

2. Description of the model

The model was constructed on the basis of the classical MD scheme [3]. The program allowed to investigate the behavior of about two thousands atoms in Al cluster with small Si_3 cluster inside and to obtain

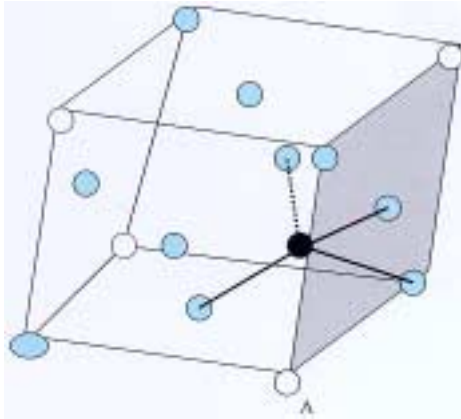
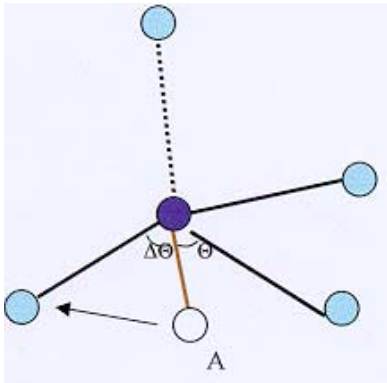


Figure 1. Fragment of FCC Al lattice with tetrahedral Si₃ nucleus. The grey circles are nodes where Si atoms have to dispose to form tetrahedral structures. The black circle shows the interstitial site in FCC lattice for the Si atom in the center of tetrahedron. The initial disposition of Si atom in the node A leads to formation of stressed bonds.

a high degree of precision. The starting configuration was a parallelepiped which described the Al crystal. The disposition of Si atoms in the FCC Al lattice is shown in the Fig. 1. The model temperature was 300 K. Periodic boundary conditions were used in three dimensions. Interaction of Al atoms was described by the potential of Morse-Abel type for FCC lattice [4]. For Si atoms the potential of Stillinger-Weber was used [3]. Si-Al interaction was described by potential function with parameters which were average between parameters of

potentials for Al and Si. This approximation is reasonable to use if to take into account that in the main the nearest Al atoms determine the displacements of Si atoms in their configuration shown in Fig. 1. Besides that by quantum-chemical estimations analogous to described in [5] it was shown that the bounding energy of Si-Al molecule is close to above mentioned average parameter.

In the limits of the considered model it was interesting to investigate the behavior of distorted Si cluster shown in Fig.2. This situation corresponds to the disposition of Si atoms which may arise in quenching processes of Al-Si alloys or in conditions of neutron irradiation of Al metal. The angles between chemical bonds move aside from the tetrahedral ones. As a result of computer simulation of such situation we observed the transition of Si atom to the site which was occupied by Al atom as it is shown by a pointer in Fig. 2. The



forcing out of the Al atom by Si atom is caused by aspiration of stressed chemical bonds to restore the tetrahedral angles. It is obtained that the energy which corresponds to transition of Si atom shown in Fig. 2 is equal to 0.75 eV. This is a value of energy of the vacation formation in Al [2]. The effect of forcing out of Al atom by Si atom in Al lattice plays a significant role in Si precipitates formation and may be useful for understanding of degradation mechanisms of Al-Si alloys in real conditions.

Figure 2. A Si tetrahedron from the Figure 1 $\Theta = 100^\circ$, $\Delta\Theta = 9^\circ$

3. Discussion

In [2] the results are described which show that it is necessary in quenching experiments to have a pre-aging time 10 - 15 s to develop the finely distributed precipitates for different solute concentrations. Let us suggest that τ and E are the time and the activation energy which are necessary to rebuild the primary Si₃ configuration to the good tetrahedral one (Fig.2). Then:

$$\tau = \tau_0 \exp(E/kT), \tag{1}$$

where $\tau_0 = 10^{-12}$ s is an effective period of atom oscillations. If to take $\tau = 10$ sec [2] we get for $T = 300$ K $E = 0.75$ eV. So we see that the activation energy which is got from experiments [2] coincides with the value of energy of restoration of tetrahedral angles between chemical bonds.

It is important to note that the further development of the Si nuclei in Al depends on the their stability. The stability of Si clusters is determined by their size and form. Theoretical and experimental investigations have led to conclusion that the stability parameters of Si clusters have a quasi-periodic dependence on cluster size [6]. Besides that, it was established that in each case the dependence of energy characteristics of Si clusters

on their size is determined by the real structure of Si clusters and by the degree of their disordering. The nature of chemical bonds which saturate the free valences of the periphery cluster atoms also plays a significant role in the stabilization of Si clusters [7]. Therefore the mechanisms of Si precipitates formation in Al matrix may be understood on the basis of simultaneous consideration of the thermodynamic conditions of Al-Si phases behavior and quantum-chemical features of Si clusters in Al lattice.

4. Conclusion

The disordered Si clusters, which arise in Al-Si solutions and alloys as nuclei for Si precipitates growth at the pre-aging stages restore their tetrahedral structure. Simultaneously the effect of Al atom forcing out from its lattice site by Si atom has place.

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