

Influence of Ni-Al interphase boundary orientation on the interdiffusion rate at temperatures above aluminum melting point: a molecular dynamics study

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Abstract. The influence of the orientation of Ni-Al interphase boundary relative to the nickel crystal lattice on the intensity of interdiffusion at temperatures above the melting temperature of aluminum was studied by the method of molecular dynamics. It was observed that the boundary between the crystalline and liquid phases was, in fact, shifted by two or three atomic planes into the aluminum phase – a thin layer of aluminum near the boundary retained a crystalline structure that repeated the nickel lattice. As the temperature increased, the thickness of the Al crystalline layer near the boundary decreased. The concentration curves were obtained after modeling interdiffusion at various temperatures for (111) and (001) orientations of the interphase boundary with respect to the Ni lattice. The parts of the curves responsible for the diffusion of Al atoms into crystalline Ni turned out to be similar for both orientations. However, the more sloping parts related to the diffusion of Ni atoms into liquid Al differed: diffusion of Ni atoms into Al proceeded more intensively at the (111) boundary orientation and slower at orientation (001).

Keywords: molecular dynamics, diffusion, interphase boundary, nickel, aluminum

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1. Introduction

Intermetallic compounds of Ni-Al system and alloys based on them, due to the combination of properties such as low density, high yield strength at elevated temperatures, good resistance to oxidation and corrosion, have a high potential for their use as high-temperature structural materials [1-4]. The technology for producing intermetallic compounds and alloys is based on diffusion, the process of which in such systems is complex and multifactorial. In the diffusion zone at the interface of two metals, for example, in the process of combustion synthesis, both solid ordered and disordered phases can be present simultaneously, as well as liquid mixtures with different content of components [5-7]. Knowledge of diffusion kinetics, characteristics, and mechanisms of diffusion in metal systems, in particular in the Ni-Al system, is necessary for a more detailed understanding of the processes occurring at high-

temperature synthesis and is also of great importance for the entire field of research and creation of intermetallic compounds and binary alloys.

In previous works [8, 9], we studied the self-diffusion of individual Ni, Ti, and Al atoms in liquid alloys of Ni-Al and Ti-Al two-component systems. Ordered and disordered alloys with compositions $A_{75}B_{25}$, $A_{50}B_{50}$, and $A_{25}B_{75}$ (A and B are alloy components), as well as pure metals, were considered. In particular, the characteristics of self-diffusion were obtained for the considered systems: the diffusion activation energy and the pre-exponential factor in the corresponding Arrhenius equation.

This work is devoted to the molecular-dynamics study of the influence of the orientation of the interfacial boundary relative to the crystal lattice of Ni on the rate of interdiffusion in the solid-liquid phase contact of Ni and Al, i.e. at temperatures above the melting point of Al, but below the melting point of Ni.

2. Description of the model

EAM potentials from [10] were used to describe interatomic interactions in Ni-Al system in the molecular dynamics model. These potentials were obtained in [10] on the basis of comparison with both experimental and first-principle data on various properties and structures of pure metals and intermetallic compounds NiAl and Ni_3Al . They have proven themselves in various studies and have been successfully tested in a wide range of mechanical and structural-energy properties of the alloys of the systems under consideration [9-11].

The computational cells contained about 90 thousand atoms and had the form of rectangular parallelepipeds (Fig. 1). The interphase boundary was created at the center of the computational cell along the yz-plane. Along the y and z axes, an infinite repetition of the cell was simulated, that is, periodic boundary conditions were imposed. In this case, the dimensions along these axes were selected in such a way that they were multiple, with the minimum possible deviation, of the repetition periods of both Ni and Al crystal lattices (initially, aluminum was created in a crystalline state). The unequal thermal expansion of metals for each specific temperature at which the simulation was carried out was also taken into account. Rigid conditions were imposed along the x-axis: the atoms at the left and right ends of the computational cell (colored in gray in Fig. 1) remained immobile during the computer experiment.

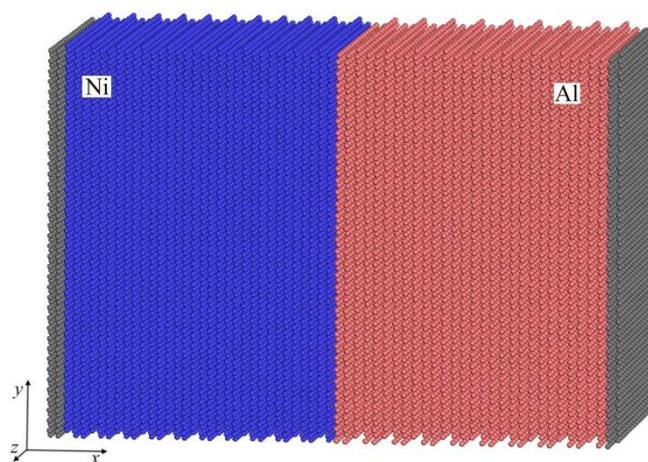


Fig. 1. Computational cell for simulating interdiffusion at the Ni-Al boundary before setting the temperature (gray atoms at the ends of the cell remained immobile during the simulation)

Both metals were initially created in the crystalline state. In this work, a solid-liquid phase contact was considered, that is, aluminum was in a liquid state during the simulation and its initial orientation did not matter. Nevertheless, to confirm this and refine the results,

we considered four initial orientations of the interphase boundary relative to the fcc crystal lattices of Ni and Al: 1) (111):(111), 2) (111):(001), 3) (001):(001), 4) (001):(111). The (111) plane is the most densely packed - its filling density with atoms (if their radius is taken equal to half the distance to the nearest neighbors in an ideal crystal) is 90.7%. The filling density of the (001) plane is 78.6%.

The temperature in the model was set through the initial velocities of atoms according to the Maxwell distribution. To keep the temperature constant during the simulation, a Nose-Hoover thermostat was used. The time integration step in the molecular dynamics method was 2 fs [12-14]. The potentials used, among other things, describe well the melting points of Ni and Al: 1710 and 990 K in our model respectively (reference values: 1726 and 933 K). The temperature varied in the range from 1200 K to 1800 K. The melting of aluminum in the model occurred much faster than interdiffusion, in a few picoseconds. Simulation of interdiffusion was carried out for several hundred picoseconds.

3. Results and discussion

After the melting of aluminum, a part of it near the interphase boundary remained in the crystalline state, repeating the nickel lattice. That is, the interface between the solid and liquid phases actually moved two or three atomic planes deep into the aluminum. Figure 2 shows examples of this phenomenon. This is explained by the fact that the Ni-Al bond is much stronger than the Al-Al bond [10], and therefore, in particular, the melting temperatures of intermetallic compounds of the Ni-Al system significantly exceed the melting temperature of aluminum [1,2]. Therefore, for the destruction of Ni-Al bonds at the interphase boundary, the considered temperatures are not enough. Because of this, the detachment of Ni atoms and their entrainment into liquid aluminum at the initial stage of the computer experiment, obviously, becomes more complicated and does not occur as quickly as it would be in direct contact of nickel with the liquid phase. Subsequently, despite the mixing of the components and blurring of the boundary, this effect persisted (Fig. 3), but became less noticeable with increasing temperature.

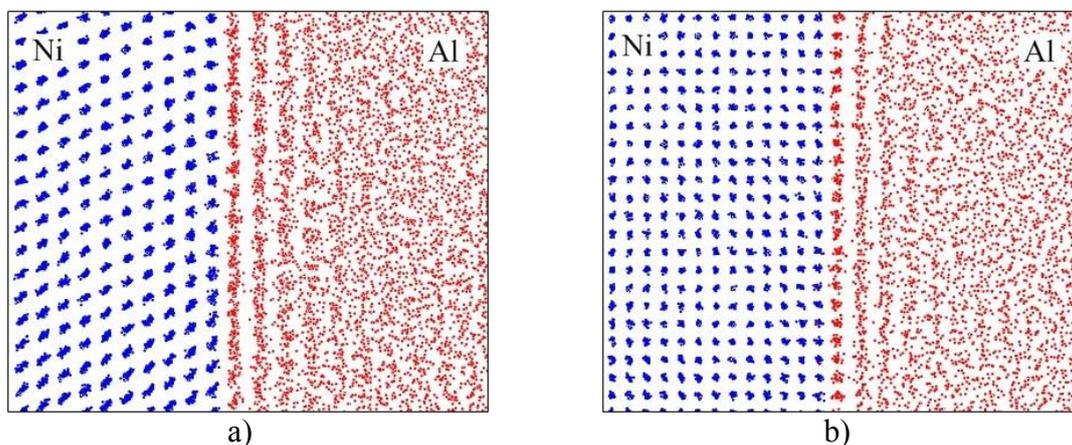


Fig. 2. Formation of a crystal structure in Al near the interphase boundary at a temperature above the melting point of aluminum (1300 K): a) (111) orientation; b) (001) orientation

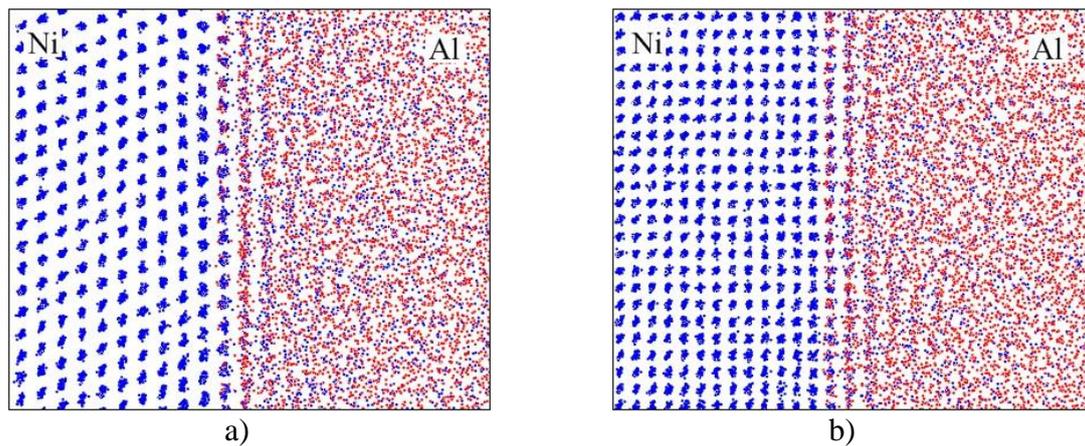


Fig. 3. Distribution of atoms in computational cells after simulation for 500 ps at a temperature of 1400 K: a) (111) orientation; b) (001) orientation

Despite the phenomenon described above, the mutual diffusion at the Ni-Al interface during solid-liquid contact was intense enough to be modeled using the molecular dynamics method. A significant predominance of the diffusion of Ni atoms deep into liquid Al was observed compared to the diffusion of Al atoms into crystalline Ni (Fig. 3), which is explained, first of all, by the difference in the aggregate states of Ni and Al.

Figure 4 shows the concentration curves obtained at temperatures of 1400 and 1600 K after simulation for 500 and 300 ps, respectively, for the four interphase boundary orientations considered. The curves were plotted by analyzing the concentration of Ni atoms in a layer 5 Å thick when this layer was displaced by 1 Å along the x-axis. The obtained concentration curves are qualitatively similar to those obtained experimentally for this system [6,7].

The relatively steep left part is the diffusion of Al atoms deep into the Ni crystal lattice. Obviously, it proceeds much more slowly than the diffusion of Ni atoms into liquid Al (the right, more sloping part on the concentration curves). In connection with the above-described phenomenon of the preservation of the crystal structure in the aluminum phase in a small layer near the interface, the concentration curves at a temperature of 1400 K show a change from the steep to the sloping part at concentrations well below 50% (Fig. 4a). However, as the temperature increased, this boundary shifted and approached 50% (Fig. 4b).

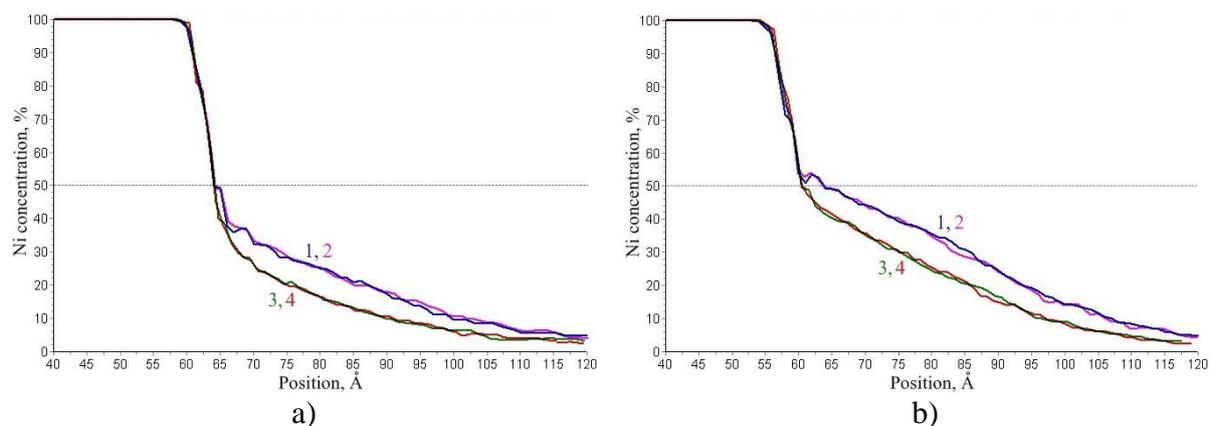


Fig. 4. Distribution of the concentration of Ni atoms in the computational cells: a) after 500 ps at a temperature of 1400 K; b) after 300 ps at a temperature of 1600 K. The numbers indicate the considered orientations: 1) (111):(111), 2) (111):(001), 3) (001):(001), 4) (001):(111)

The left parts, as can be seen, turned out to be identical and have no difference for all considered interphase boundary orientations. Also similar, as expected, were the results obtained for the same orientation of the boundary relative to the Ni lattice, but for different initial orientations of the Al lattice (1, 2 and 3, 4 in the figures). However, on the right part for the (111) and (001) orientations, the curves did not coincide: for the (111) orientation, in all cases, the concentration curves were located above the (001) curves, which primarily indicates an easier detachment of Ni atoms from the boundary with the orientation (111) and entrainment in aluminum compared to the (001) orientation. That is, with the (111) orientation, interdiffusion proceeds more intensively than with the (001) orientation.

A similar effect of the orientation of the interface is observed when the crystallization front moves in metals: crystallization from the interface, which has a relatively looser (001) orientation in the fcc lattice, moves 1.3-1.5 times faster than from the interface, which has the densest orientation (111) [15-18]. This behavior can be explained by the difference in the depth of potential wells in which atoms are located at the interface. Near the interface between metals in solid-liquid-phase contact, as well as at the liquid-crystal interface, the main role is played by the difference between the free energies of an atom near the interface in the liquid phase and "built-in" into the crystal boundary. This value can be compared with the energy of an adatom on the corresponding free surface of the crystal, or with the activation energy of its migration over the given surface. For example, using computer simulations, in [19] it was shown that the activation energy of diffusion of adatoms over the free (001) surface of fcc metals is almost two times higher than over the (111) surface.

4. Conclusion

The method of molecular dynamics was used to study the influence of the orientation of the interphase boundary relative to the nickel crystal lattice on the intensity of interdiffusion at temperatures above the temperature of aluminum, that is, at solid-liquid-phase contact of metals. It was noticed that near the interphase boundary a small layer of aluminum with a thickness of two or three atomic planes remained in the crystalline state, repeating the nickel lattice, and therefore the boundary between the crystal and the liquid metal actually shifted deeper into the aluminum. In the process of interdiffusion and blurring of the boundary, this effect persisted, but with increasing temperature, the thickness of the crystalline layer near the boundary decreased.

For the considered orientations (111) and (001) of the interphase boundary with respect to the Ni lattice, the concentration curves were obtained after modeling interdiffusion at different temperatures. The parts of the curves responsible for the diffusion of Al atoms into crystalline Ni turned out to be similar for both orientations. However, the more sloping parts related to the diffusion of Ni atoms into liquid Al differed: diffusion of Ni atoms into Al proceeded more intensively at the (111) boundary orientation and slower with a relatively "loose" orientation (001), which, apparently, is due to the difference in the energies of the Ni atoms in the aluminum phase and those embedded in the boundary of crystalline nickel.

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