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Reaction characteristics of Ni–Al nanolayers by molecular dynamics simulation

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ABSTRACT

We have performed molecular dynamics simulations to investigate the reaction characteristics of Ni–Al nanolayers by varying ignition temperature and bilayer thickness with three different compositions (1:1, 3:1, and 1:3) of Ni to Al. The overall sequence of reaction pathway was found to be unchanged by stoichiometry, but the reaction rate and the extents of intermixing varied by case. Also, the reaction kinetics and thermodynamics were quantitatively investigated by various structural and reaction conditions. Through this systematic study, the reaction characteristics of Ni–Al nanolayers were theoretically quantified, which can provide an insight into the fabrication of advanced Ni–Al nanolayer systems.

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Introduction

Reactive multilayer foils are exothermic materials self-propagated and reacted by thermal or mechanical stimuli from external environment [1–4]. The reactive multilayer foils, which consist of alternating layers of two or more solid reactants, are manufactured by vapor deposition methods via sputtering [5–7], mechanical processing techniques such as rolling [8–10], compacting [11], and etc. They exhibit specific characteristics of large heat of mixing, fast burn rate, and high power [12], thus have many potential applications in military and industrial fields such as eco-friendly lead-free primers and detonators [13,14], welding or joining [15,16], soldering [17], heat source for clinical usage [18], and etc. There have been many studies including fundamental ones because of their complex reaction pathways involving melting, mass and thermal diffusions, and phase transition in initiation and progression of self-propagating exothermic reactions [19]. Especially, Ni–Al systems have attracted much attention because of high thermal and acidic stabilities in the formation process of intermetallic phase [20,21]. Recently, several experiments made

significant progresses of the layer systems in nanoscale in terms of heat of reactions and speed of reaction by using high-end characterization methods such as differential scanning calorimetry (DSC) and in-situ XRD [22–24]. However, experimental approach to the nanoscale system still has a limit to interpret the reaction phenomena originated from the atomistic level.

In that respect, molecular simulation approach came into play an important role; molecular dynamics (MD) simulation is a suitable computational technique to observe physical and chemical phenomena in the atomic scale. In particular, by using the embedded atom method (EAM) potential developed by Purja Pun and Mishin [25], fundamental studies of Ni–Al multilayer system were actively conducted. For example, Zhao et al. elucidated a mechanism of shock-induced alloying reaction involving pore effects [18]. Cherukara et al. studied exothermicity, reaction speed, and structural change in terms of temperature, periodic length, and defects [26]. A few important factors such as stoichiometry and ignition temperature have strong influence on reaction but crucial data are still largely deficient in this area. Especially, the quantitative correlation between structural properties and reaction characteristic is very much critical information yet missing.

There are representative phases in Ni–Al binary systems, which are Ni₃Al, NiAl, and NiAl₃ [27]. Out of them, B2–NiAl phase is energetically the most stable while Al-rich NiAl₃ is unstable [25]. Thus, by the change of stoichiometry, the product phase is expected

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to be varied. In this study, therefore, we set up the Ni–Al bulk nanolayers to have three stoichiometric ratios of NiAl (1:1), Ni₃Al (3:1), and NiAl₃ (1:3), where the bilayer thickness are changed, under the variation of ignition temperature. The reaction thermodynamics of model systems was explored in terms of the heat of reaction, which was obtained by estimating the enthalpy change in the reaction process. Also, the reaction kinetics was investigated by estimating the reaction time and diffusion coefficients as a function of ignition temperature and bilayer thickness. In summary, the reaction kinetics and thermodynamics were quantitatively elucidated in terms of the variation of ignition temperature, stoichiometry, and bilayer thickness in Ni–Al nanolayers.

Simulation models and details

Fig. 1 shows the model systems of Ni–Al nanolayers used in the MD simulations. The Ni–Al nanolayers were constructed to confront the most stable (111) surfaces of Ni and Al [28]. Two supercells of Ni $6 \times 10 \times 1$ and Al $5 \times 9 \times 1$ were first put together in order to minimize the lattice mismatch (i.e., $\sim 2\%$). Then, by controlling the number of atoms, three different stoichiometric systems (i.e., NiAl, Ni₃Al, and NiAl₃) were modeled to have the different values of bilayer thickness (i.e., 10, 20, and 30 nm) in z-direction, which contain from 17,280 to 63,360 atoms. Note that simulation box lengths in x- and y-directions were kept for 5.1 nm. The information of the model systems used in the MD simulation is provided in Table S1 in the Supporting information.

In order to perform the simulation, we have used EAM potential developed by Purja Pun and Mishin [25] in the LAMMPS package [29]. In order to mimic heating and igniting processes, NVT (i.e., canonical – constant temperature) and NVE (i.e., microcanonical – constant energy) ensembles were consecutively applied in the MD simulation. First, MD with NVT ensemble was run for initial 5 ps at 1000, 1400, and 1700 K. This step can be considered as a thermal shock, starting from no mixing at the interface. The Nose–Hoover thermostat with coupling constants of 0.1 ps was used to control

the temperature. Next, MD with NVE ensemble was run until the temperature and pressure values converged, indicating the alloying reaction was completed. In this calculation process, thermal energy accumulated in Ni and Al atoms in the heating (NVT MD) process was transformed into kinetic energy, which in turn triggers mixing of Ni and Al atoms, at conserved total energy. The time step for all MD simulation was set to be 1 fs.

When thermal shock is applied to the Ni–Al nanolayers, temperature rises by mixing of Ni and Al, which undergoes phase transition from solid to liquid. In order to quantify the reaction kinetics, the reaction time (τ) was estimated at the point where final temperature became constant (i.e., the average temperature for last 0.1 ns simulation). Subsequently, the τ 's were compared with respect to the ignition temperature, bilayer thickness, and stoichiometry.

Results and discussion

Reaction mechanism by stoichiometry

Fig. 2 shows the mixing behaviour in the reaction process of NiAl 20 nm system with the ignition temperature of 1000 K. A series of snapshots from NVE MD simulation are shown in Fig. 2(a), and the time evolution of overall temperature and pressure in the system is shown in Fig. 2(b). As reported in previous studies [30–32], the exothermic reaction pathway of Ni–Al nanolayers can be discretely divided into several reaction steps. Initially, the mixing between Ni and Al occurred at the interface and during this stage, the temperature and pressure were slowly varied. In the second stage (from 3.15 ns), the Al region began to melt with a continuous rise in temperature and the rapid increase in pressure. Note that since the total volume of the system is fixed, the structural expansion by melting induces the pressure to increase and the melting transition in the Al region was completed as indicated by sudden dip of temperature or pronounced peak of pressure, also as reported by Zhao et al. [32].

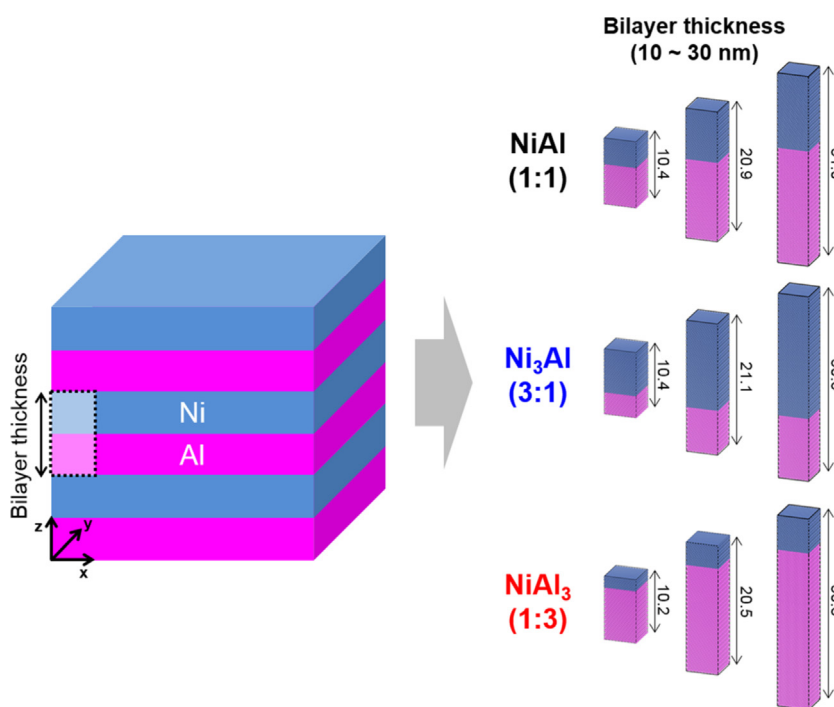


Fig. 1. Schematics of model systems used in MD simulations. Three stoichiometric ratios (i.e., NiAl, Ni₃Al, and NiAl₃) with the variation of bilayer thickness (i.e., 10, 20, and 30 nm) were considered. Ni and Al atoms are coloured as dark blue and pink, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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