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A molecular dynamics study on the chemical reaction of Ni/Al reactive intermetallics

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ABSTRACT
The chemical reaction mechanism of Ni/Al composites, referring to the exothermic mechanism and intermetallic-forming mechanism, is investigated by using molecular dynamics simulation. During the exothermic process, the influences of Ni/Al atomic ratios and crystallographic orientations on the exothermic reaction are systematically investigated. The exothermic mechanism can be explained by the atomic diffusion that increases the quantity of Ni–Al chemical bonds. There are two pathways to form the intermetallic phase during the chemical reaction. One is the atomic diffusion that forms the B2 NiAl phase at the interface. The other way is quenching the sample to the room temperature, but the type of intermetallic phases depends on the Ni/Al atomic ratio and ignition temperature.

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I. INTRODUCTION
Intermolecular reactive composites (IRCs), as a new class of energetic materials, can realize highly exothermic self-sustained chemical reactions, when subjected to extreme conditions such as high temperature, electric shock, or physical impact. These materials exhibit some excellent properties, including extremely fast burn rates, high combustion temperatures, and lower volume expansion than traditional energetic materials due to the lack of gaseous products. Therefore, they are promising candidates for a wide range of defense and commercial applications, such as nanoscale energy sources for bio-agent defeat and medical treatment, environmentally clean primers, reactive welding, ultrafast fuses, and smart thermal batteries. Among the IRCs, the investigations of Al/Ni reactive systems have already attracted extensive attention both theoretically and experimentally in recent years because of the ideal combination of mechanical and physical properties as well as the high energy release capability. In addition, intermetallic-forming reactions between Al and Ni could occur under the conditions of certain temperatures or shock compression conditions. However, until now the chemical reaction mechanism (exothermic mechanism and intermetallic-forming mechanism) of the Ni/Al system is still one of the long-standing and open scientific questions.

Experimental determination of the Ni/Al chemical reaction mechanism is challenging because of the involved characteristic length (nanometers) and timescale (nanoseconds). Complementing experimental efforts, molecular dynamics (MD) simulation, which can reveal atomic-scale structure evolution and interpret the relevant experiments at the microscopic level, is an effective method to clarify the chemical reaction mechanism. Cherukara et al. proposed the reaction mechanisms in thermally ignited samples of core/shell Ni/Al wires. They found that the samples ignited at low temperatures (900 K or less) follow a multi-stage reaction process involving solid-phase diffusion and formation of the B2 NiAl phase, while samples ignited at higher temperatures follow a direct reaction path with accelerated diffusion of Ni into molten Al, leading to a completely molten final state. Cherukara et al. characterized the exothermic chemistry of Ni/Al nanolaminates for various temperatures, periodic lengths, and in the presence of extended defects. They also investigated the thermomechanical response and chemical process under shock loading of a granular material consisting of laminated Ni/Al grains. Wu and Zhao analyzed the complete reaction pathway of nanostructure Ni/Al clad particles with an Ni/Al atomic ratio of 3:1 under thermal loading. They confirmed that the B2 NiAl compound is an intermediate product and finally transforms into a Ni3Al compound. Sandoval et al. studied the reactive process of Ni/Al bilayer systems at 800 K in both isothermal and isenthalpic conditions. They found that a disordered phase always forms upon mixing as a
precursor to a more stable nanocrystalline B2 NiAl phase. Weingarten and Rice investigated the influence of pressure on atomic mixing and subsequent exothermic alloying reaction. All their results demonstrated that MD simulation is an effective method to investigate the Ni/Al chemical reaction. However, so far the influences of Ni/Al atomic ratios and crystallographic orientations on the chemical reaction have not been well studied. What is more, the reaction mechanism, including the exothermic reaction and intermetallic formation, still needs further investigation.

In this paper, the chemical reaction of the Ni/Al system is modeled by using MD simulation to clarify the reaction mechanism. Section II of this presentation addresses the methodology related to MD simulations and the analysis methods. The results and discussion, about the temperature profiles, morphological evolutions, atomic structures, exothermic mechanism, and intermetallic formation mechanism, are presented in Sec. III. Based on the simulation results, the conclusions about the Ni/Al chemical reaction are summarized in Sec. IV.

II. METHODOLOGY

The Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) is used to simulate the reaction process of the Ni/Al system. An embedded atom method potential developed by Purja Pun and Mishin is used to describe the interatomic interactions. The potential is based on the previously developed accurate potential for pure Ni and Al. At the same time, this potential can also accurately reproduce a variety of physical properties of the NiAl and Ni₃Al phases and shows reasonable agreement with experimental and ab initio data for phase stability across the Ni/Al phase diagram. Using this interatomic potential, the melting points of Al, Ni, and B2 NiAl phases are, respectively, 1042 K, 1701 K, and 1859 K after quenching the sample to the room temperature. The contact surfaces of Ni and Al are both (0 0 1) planes. In this paper, five models, which are made of Ni block sandwiched between two blocks of single crystal Al, are respectively built, as shown in Fig. 1.

To investigate the influence of the Ni/Al atomic ratio on the chemical reaction, three sandwiched models, with the Ni/Al atomic ratios of 1:3 (#1), 1:1 (#2), and 3:1 (#3) are, respectively, analyzed. The contact surfaces of Ni and Al are both (0 0 1) planes. In this potential, the lattice constants of Ni (lₙi) and Al (lₐl) are, respectively, 3.52 Å and 4.05 Å at 300 K. Because of the incommensurate unit cell sizes of the Ni and Al crystals, the system sizes in the x- and y-directions have to be chosen carefully to avoid any severe deformation. In the three simulations, 45 Ni unit cells and 39 Al unit cells in the x- and y-directions are fitted resulting in a mismatch of about 0.25%.

To investigate the influence of crystallographic orientations on the chemical reaction, three sandwiched models, with the crystallographic orientations of (100) (#2), (110) (#4), and (111) (#5) are, respectively, analyzed. The Ni/Al atomic ratios are 1:1 in the three models. The (110) laminates are created from unit cells oriented along [001], [110], and [110], and to minimize cell mismatch we replicate the Ni unit cell 45 times along [001] and 32 times along [110], while the Al unit cell is replicated 28 and 24 times, respectively. This leads to cell mismatch of 0.28% along [001] and 0.67% along [110]. The (111) laminates are created from unit cells oriented along [110], [112], and [111], and to minimize cell mismatch we replicate the Ni unit cell 32 times along [110] and 28 times along [112], while the Al unit cell is replicated 28 and 24 times, respectively. This leads to cell mismatch of 0.67% along [110] and 0.17% along [112].

In the five models, the dimensions for the Ni blocks are, respectively, 158.4 × 158.4 × 38.7 Å³, 158.4 × 158.4 × 84.5 Å³, 158.4 × 158.4 × 140.8 Å³, 158.4 × 158.4 × 159.3 Å³, and 158.4 × 159.9 Å³. The dimensions for the Al blocks are, respectively, 158.0 × 158.0 × 89.1 Å³, 158.0 × 158.0 × 64.8 Å³, 158.0 × 158.0 × 36.5 Å³, 158.0 × 160.4 × 63.0 Å³, and 160.4 × 158.7 × 63.1 Å³. Totally, the five models, respectively, contain 356 796, 389 088, 433 512, 388 032, and 392 448 atoms.

To create well-equilibrated samples, all the systems are first relaxed under constant-pressure-temperature conditions (NPT ensemble) at 300 K and zero external pressure for 200 ps. To simulate the thermal ignition, the systems are rapidly heated to the desired ignition temperature (T₀) of 900 K in 20 ps, still using the NPT ensemble. Then, the samples are thermalized at T₀ for a further 10 ps. The chemical evolution of the reactive system is then followed under isobaric-isenthalpic conditions (NPH ensemble) for the duration of reaction. The initial configurations of all simulation cells are applied under three-dimensional periodic boundary conditions. For all simulations, a constant integration time step of 2 fs is used.

After simulation, atomic visualizations are performed using the OVITO software. Radial distribution functions (RDFs) are adopted to analyze the atomic structures of both Ni and Al after the exothermic reaction. Common neighbor analysis (CNA) is adopted to investigate the micro-structures of the Ni/Al systems after quenching the sample to the room temperature.

III. RESULTS AND DISCUSSION

A. Temperature profiles

Figure 2 elucidates the five temperature profiles of the Ni/Al systems. As shown in Fig. 2, a small amount of heat is initially released with a slow exothermic rate. When the five Ni/Al systems, respectively, arrive in 1009 K, 978 K, 939 K, 990 K, and 998 K, the exothermic rates increase rapidly, as indicated by the increased slope of the temperature profiles. Weingarten and Rice considered that the increased reaction rate is owing to the melting of Al. However, based on our simulations, we can find that the temperatures for the increased reaction rate are lower than the melting point of Al (1042 K). The inset in Fig. 2 shows the temperature profile of the Ni/Al system in #3 from 0 to 30 ns. Finally, the temperatures of the Ni/Al systems reach the maximum. In the five Ni/Al systems, the maximum temperatures are, respectively, 1434 K, 1397 K, 1647 K, 1890 K, and 1904 K. Then, we can calculate the average exothermic rate from the ignition temperature to the maximum. The average exothermic rates are, respectively, 60.0 K/ns, 117.1 K/ns, 27.3 K/ns, 143.5 K/ns, and 119.5 K/ns. Cherukara et al. also simulated the exothermic of Ni/Al nanolaminates with the Ni/Al atomic ratio of 3:1. Their average exothermic rate is at least an order of magnitude larger than our results, which is...
possibly due to their higher ignition temperature (1200 K) and shorter periodic length along the z-direction.

To realize the influence of the atomic ratio on the exothermic reaction, the temperature curves of the Ni/Al systems with the Ni/Al atomic ratios of 1:3 (#1), 1:1 (#2), and 3:1 (#3) are analyzed. For the three systems, it takes a longer time to arrive in the increased exothermic rate by decreasing the Ni/Al atomic ratio from 3:1 (#3) to 1:3 (#1). When the Ni/Al atomic ratio equal to 3:1 (#3), during the rapid exothermic process, the exothermic rate is reduced with the appearance of a kink at approximately 5 ns. By observing the

FIG. 1. Ni and Al samples prepared for MD simulation. The Ni/Al atomic ratio and lattice direction are, respectively, 1:3 and (100) in #1 (a); 1:1 and (100) in #2 (b); 3:1 and (100) in #3 (c); 1:1 and (110) in #4 (d); and 1:1 and (111) in #5 (e). The green and yellow atoms, respectively, represent the Al and Ni atoms.

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morphological evolutions in Fig. 3(c), this is due to the formation of the B2 NiAl phase at the Ni/Al interface. In the three Ni/Al systems, the maximum temperature and average exothermic rate tend to decrease as the number of Al atoms (#1) or Ni atoms (#3) increases.

To realize the influence of crystallographic orientations on the exothermic reaction, the temperature curves of the Ni/Al systems with the lattice direction of (100) (#2), (110) (#4), and (111) (#5) are analyzed. In the three Ni/Al systems, the exothermic rate is the highest with the lattice direction of (110) but is the lowest with the lattice direction of (111). However, the maximum temperatures are almost the same, which is probably due to the 1:1 Ni/Al atomic ratio in the three samples.

B. Morphological evolutions

To realize the exothermic process, the morphological evolutions of the five Ni/Al systems are investigated in Fig. 3. It is seen that the exothermic reaction is dominantly due to the Ni atoms diffuse into the Al side.

To realize the influence of the Ni/Al atomic ratios on the exothermic reaction, the morphological evolutions of the three Ni/Al systems with the Ni/Al atomic ratios of 1:3 (#1), 1:1 (#2), and 3:1 (#3) are analyzed in Figs. 3(a)–3(c). At \( t = 1 \) ns, some Ni atoms at the Ni/Al interface diffuse into the Al side and gradually change the Al atoms into a disordered state. At \( t = 4 \) ns, as the Ni atoms continuously diffuse into the Al side, the thickness of the central Ni gradually decreases and nearly all the Al atoms arrange in the disordered state. Combined with the temperature profiles in Fig. 2, we can consider that, when all the Al atoms turn into the disordered state, the exothermic rate begins to rapidly increase. At \( t = 6 \) ns, the atoms at the central Ni still keep the crystalline structure. However, a thin layer of the B2 NiAl phase is formed at the Ni/Al interface in the Ni/Al system of #3. Cherukara et al.\(^2\) also observed the formation of the B2 NiAl phase at the Ni/Al interface in the simulation of the core/shell Ni/Al wires. However, in our simulations, the NiAl phases are almost not observed in the Ni/Al system of #1 and #2. By counting the number of Ni and Al atoms in the three systems, the Ni/Al atomic ratios at the Al side are, respectively, 0.15, 0.33, and 0.56. Maybe it is the higher Ni/Al atomic ratio at the Al side that results in the formation of the B2 NiAl phase. At \( t = 9 \) ns, nearly all the Ni and Al atoms are homogeneously mixed in the disordered state in the Ni/Al system of #1 and #2. We can consider that when all the Ni and Al atoms are uniformly mixed, the temperature of the system reaches the maximum. However, for the Ni/Al systems of #3, the central Ni block still remains the crystalline structure, and more B2 NiAl phases are generated at the Ni/Al interface at \( t = 30 \) ns. In this situation, the Ni and Al atoms cannot be totally mixed together, which is due to the B2 NiAl phase that prevents the diffusion of the Ni atoms. In the MD simulation results performed by Cherukara et al.,\(^2\) the B2 NiAl phase is not observed in their Ni/Al system with the atomic ratio of 3:1, which is possible because of the higher ignition temperature (1200 K).

To realize the influence of the crystallographic orientations on the exothermic reaction, the morphological evolutions of the three Ni/Al systems with the lattice direction of (100) (#2), (110) (#4), and (111) (#5) are analyzed in Figs. 3(b), 3(d), and 3(e). As can be seen, the highest exothermic rate in the Ni/Al system with the lattice direction of (110) can be explained by the fastest diffusion process. At \( t = 6 \) ns, almost all the Ni atoms have already diffused into the Al side. However, at this moment, the central Ni atoms still remain the crystalline structures and slowly diffuse into the Al side in the Ni/Al systems with the lattice direction of (100) and (111). The exothermic rate is the lowest in the Ni/Al systems with the lattice direction of (111). This is mainly due to the lowest diffusion rate for the Ni atoms to diffuse into the Al side. Compared with the Ni/Al systems with the lattice direction of (100) and (110), it takes a longer time to change all the Al atoms into the disordered state in the Ni/Al systems with the lattice direction of (111).

In the five models, the sample lengths along the \( z \)-direction are, respectively, 221.0 Å, 218.1 Å, 217.8 Å, 214.6 Å, and 215.6 Å. Based on the morphological evolutions of the five Ni/Al systems, we can speculate that the exothermic rate tends to decrease as the sample length along the \( z \)-direction increases, which is due to the longer time needed to change all the Al atoms into the disordered state. In the MD simulations performed by Cherukara et al.,\(^2\) they also found that the reaction time increases with increasing periodic length due to the larger transport distances.

C. Radial distribution functions

In the five Ni/Al systems, when the temperature of the Ni/Al system reaches the maximum, the atomic structures of both Ni and Al are analyzed by using the partial RDF,\(^2\) as shown in Fig. 4. Before the chemical reaction, the initial models of pure Ni and Al are also investigated in Fig. 4. In the initial models, the positions of the first two peaks sequentially represent their first neighbor distances and their lattice constants. The lattice constants of Ni and Al are, respectively, 3.52 Å and 4.05 Å at 300 K, and the first neighbor distances are 2.49 Å and 2.86 Å at 300 K. In the five Ni/Al systems, the first peaks of both the Ni and Al shift to the left,
FIG. 3. The morphological evolutions of the Ni/Al systems in #1 (a), #2 (b), #3 (c), #4 (d), and #5 (e).
which is probably due to the atomic diffusion that reduces the average distance in each atom.\textsuperscript{20} The second peaks of Ni and Al disappear in the Ni/Al systems of #1, #2, #4, and #5, which means that both the Ni and Al atoms arrange in the disordered state. In these situations, melting takes place in the Ni/Al systems. However, for the Ni atoms in the Ni/Al system of #1, a second split peak can be observed obscurely. The second split peak is a characteristic only of a solid amorphous structure.\textsuperscript{34} Therefore, we can consider that, for the Ni/Al system with the atomic ratio of 1:3 (#1), the Al atoms mainly arrange in the liquid state, but the Ni atoms arrange in the solid state. It should be noted that, in this potential, the melting point of the B2 NiAl phase is 1780 K. During the exothermic process in the Ni/Al system of #3, the maximum temperature (1647 K) is lower than the melting point of the B2 NiAl phase. The B2 NiAl phase at the Ni/Al interface prevents the diffusion of Ni atoms. Therefore, in this condition, both the Ni and Al atoms mainly remain the crystalline structure in the solid state.

D. Exothermic mechanism

Based on the morphological evolutions, we can hypothesize that the exothermic reaction occurs because of the formed Ni–Al chemical bonds. To investigate the exothermic mechanism, the numbers of Ni–Al chemical bonds are calculated. Xiong et al.\textsuperscript{35,36} characterized the chemical reaction during shock loading by calculating the neighboring Ni atoms around each central Al atom. They assumed that the Ni–Al bond is formed if at least one Ni atom is found within 0.25 nm radius. The reaction fraction ($\eta$) is defined as the ratio of reacted Al atoms to all Al atoms in the shocked region. In our opinions, the reaction fraction should be defined as the ratio between the number of Ni–Al chemical bonds to the total chemical bonds in the Ni/Al systems. The number of Ni–Al chemical bonds can be calculated by summing the number of neighbor Ni atoms around each central Al atom. Then, the reaction fraction can be calculated in the following equations:

$$\eta = \frac{n_{\text{Ni–Al}}}{n_{\text{Ni–Ni}} + n_{\text{Al–Al}} + n_{\text{Ni–Al}}}.$$
where $n_{Ni-Ni}$, $n_{Al-Al}$, and $n_{Ni-Al}$ are, respectively, the number of Ni–Ni, Al–Al, and Ni–Al bonds.

Figure 5 demonstrates the relationships between the average reaction fraction and temperature in the five systems. As can be seen, when all the Al atoms turn into the disordered state (approximately 1000 K), the slopes between the temperature and reaction fraction become much higher. In the five systems, the temperature increases almost linearly with the increase of reaction fraction. Increasing the Ni/Al atomic ratio from 1:3 (#1) to 3:1 (#3), more Ni–Al bonds are required to turn all the Al atoms into the disordered state. Therefore, under the same exothermic temperature, the reaction fraction tends to decrease as the increase of the Ni/Al atomic ratio. After all the Al atoms arrange in the disordered state, the slopes between the temperature and reaction fraction are almost the same in the Ni/Al systems of #1 and #2. However, when the Ni/Al atomic ratio equal to 3:1, the slope is slightly changed due to the formation of the B2 NiAl phase. For the Ni/Al systems with the lattice direction of (100) (#2), (110) (#4), and (111) (#5), the relationships between the temperature and reaction fraction are almost the same. Therefore, we can conclude that the exothermic reaction of the Ni/Al systems is because of the formed Ni–Al chemical bonds. The maximum temperature depends on the ratio between the number of Ni–Al bonds and the total chemical bonds in the Ni/Al systems. The system size and lattice direction influence the exothermic rate but do not affect the final temperature.

E. Intermetallic formation mechanism

In our simulations, when the Ni/Al atomic ratio equal to 3:1 (#3), the Ni atoms diffusion results in the formation of the B2 NiAl phase at the Ni/Al interface. However, when the Ni/Al atomic ratio is 1:3 (#1) or 1:1 (#2, #4, and #5), the B2 NiAl phase is almost not observed. Therefore, we hypothesize that maybe quenching the system to the room temperature is another pathway to form the intermetallic phases. To verify this hypothesis, the Ni/Al systems are quenched to the room temperature with a cooling rate of $10^{11}$ K/s at zero external pressure.

Figures 6(a)–6(c) demonstrate the morphologies of the Ni/Al system in #1, #2, and #3 after quenching to the room temperature. Figures 6(e)–6(g), respectively, reveal their micro-structures by
CNA. When the Ni/Al atomic ratio equal to 1:3 (#1), the Ni and Al atoms are homogeneously mixed in the disordered state [Fig. 6(a)] and form the amorphous phase [Fig. 6(e)]. However, in the experiments conducted by Ma et al., the reaction product is composed of a mixture of three phases: NiAl3, Ni2Al3, and Al. The formation of the amorphous phase in our simulation is probably due to the solid amorphous Ni atoms [Fig. 4(a)] when the temperature reaches the maximum. When the Ni/Al atomic ratio equal to 1:1 (#2), nano-crystalline B2 NiAl phases are produced [Figs. 6(b) and 6(f)]. The fractions of BCC and disordered atoms are, respectively, 63.4% and 36.0%, and the percentages of FCC and HCP structures are almost zero. It should be mentioned that the disordered atoms are mainly the grain boundaries and some amorphous atoms. In the shock-compression experiments, Kelly and Thadhami also observed the B2 NiAl phase in the fully reacted samples with the Ni/Al atomic ratio of 1:1. When the Ni/Al atomic ratio equal to 3:1, the B2 NiAl phases are formed at the Ni/Al interface [Figs. 6(c) and 6(g)]. In this situation, the central Ni still remains the crystalline structure, and some amorphous structures are generated at the Al side. The fractions of FCC, BCC, and disordered atoms are, respectively, 53.0%, 34.3%, and 11.9%, and the percentages of HCP structures are almost zero. This is because of the lower ignition temperature (900 K) in our simulations. In the experimental results performed by Ma et al., the Ni3Al phase as the reaction product is generated. We hypothesize that, if the Ni/Al system is ignited at higher temperature, maybe the melting of the Ni/Al system results in the formation of the Ni3Al phase. To verify this hypothesis, we simulate the exothermic reaction of the Ni/Al system with the Ni/Al atomic ratio of 3:1 under the ignition temperature of 1200 K. Figures 6(d) and 6(h) show their morphologies and micro-structures after quenching to the room temperature. In this situation, Ni3Al phases with twin structures are generated in the Ni/Al system. The fractions of FCC, BCC, HCP, and disordered atoms are, respectively, 41.1%, 17.2%, 31.3%, and 10.4%. Therefore, we can conclude that there are two pathways to form the intermetallic phase during the chemical reaction. One is the atomic diffusion that forms the B2 NiAl phase at the interface. The other way is quenching the sample to the room temperature, but the type of intermetallic phases depends on the Ni/Al atomic ratio and ignition temperature.

IV. CONCLUSIONS

In this paper, the reaction mechanism of the Ni/Al system is investigated through MD simulation. The results are summarized as follows:

1. The exothermic mechanism is because of the atomic diffusion that increases the number of Ni-Al chemical bonds. When all the Ni and Al atoms are uniformly mixed, the exothermic temperature reaches the maximum.
2. The exothermic rate and maximum temperature depend on the Ni/Al atomic ratio. They are the highest with the Ni/Al atomic ratio of 1:1 but tend to decrease by increasing the number of Ni or Al atoms.
3. The lattice direction influences the exothermic rate but does not affect the final temperature.
4. There are two pathways to form intermetallic phases during the chemical reaction. One is the atomic diffusion that generates the B2 NiAl phase near the interface. The other way is quenching the sample to the room temperature, but the type of intermetallic phases relies on the Ni/Al atomic ratio and ignition temperature.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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