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Atomic packing and medium-range order in Ni_3Al metallic glass

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ABSTRACT

In this report, classic molecular dynamics simulations based on the embedded atom method (EAM) are carried out to study the atomic arrangement in Ni_3Al metallic glass. The short-range order (SRO) and medium-range order (MRO) in the rapidly quenched Ni_3Al alloy are characterized by the both structural analysis methods; Radial distribution function (RDF) and Voronoi tessellation. From RDF, we found a splitting in the second peak that reflects the atomic packing beyond the nearest neighbors and it is a characteristic of the MRO. Furthermore, the transition from the supercooled state to the glassy state of Ni_3Al alloy leads to an increased population of icosahedral-like clusters and their spatial connectivity via vertex-sharing (VS), edge-sharing (ES), face-sharing (FS) and intercross-sharing (IS). Interestingly, by comparing the position ratios of subpeaks in RDF with those of the shared cluster distance to the first peak in RDF, we suggest that the first subpeak originates from the face-sharing clusters while the second one results from the vertex-sharing clusters. However, the edge-sharing is hidden between the two subpeaks due to its smaller fraction in the system.

1. Introduction

The shaping of metallic glasses (MGs) has been, and continues to be, one of the core subjects of condensed matter physics and materials chemistry. Over the last decade, recent developments in the processing techniques, characterizations methods and computational materials science have been carried out leading to the opening up of new avenues for controlled study of the structure of metallic glasses, and the new properties offered by these materials. Since the first discovery of $Au_{75}Si_{25}$ metallic glass [1] in the 1960s by rapidly quenching of the liquid melt, both theoretical and experimental techniques have been performed to study these materials. The glass transition is still a complex process due to their structural, thermodynamic and dynamic transitions but the search for metallic glasses has not stopped. In this context, several experimental techniques have been used to form metallic glasses. Among them, we cite the melt spinning [2], liquid splat-quenching [3] and pulsed laser quenching [4]. These techniques are only able to produce multicomponent metallic glasses (binary, ternary…). More recently, Zhong et al. [5] have succeeded in producing monatomic metallic glasses through ultrafast liquid quenching; they have applied this method on Ta, V, W and Mo monatomic metallic glasses. On the other hand, the modeling methods such as ab-initio molecular dynamics simulations and reverse Monte Carlo (RMC) are highly accurate and show good ability in describing the structures and dynamics of amorphous alloys.

It has been generally accepted that any molten alloy cooled to solid state will be amorphous if the cooling rate is in the order of the critical rate $R_c = 10^6$ K/s for metallic alloys [6–9] and in the order of the critical rate $R_c = 10^{12}$ K/s for pure metals. For the former materials, the glass transition has not been possible without extremely high cooling rates because of the low glass-forming ability. Thus, a composition favoring the formation of metallic glasses will have a better glass forming ability (GFA). As Inoue [10] mentioned, three famous empirical features affect the glass forming ability: i.e. multicomponent systems consist of two or more elements with significant atomic size ratios above 12% and negative heats of mixing. This explains the importance of the choice of the elements and the proportions on the stability and the density of the clusters, and therefore on the glass properties.

Bulk metallic glasses show a variety of outstanding properties which are due to their chemical composition and structural characteristics including smooth surface, absence of grain boundaries, second-order glass transition. Some Pd and Nd-based amorphous alloys have hard magnetic behavior at room temperature, and they have been commonly used as recording disks, solenoid actuator, permanent magnet motors in modern cars, and in the dermatological field [11–14]. On the other hand, the smooth surface and negative heat of mixing of metallic glasses make them ideal for applications in optical transmittance or reflectivity of devices used to harvest solar energy [15]. Moreover, these materials play an important role in the biomedical field such as

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the proper use in the orthopedic applications. The most commonly metallic glasses used for orthopedics include Ti, CoCr and Zr-based BMGs due to their excellent mechanical properties and their high corrosion resistance [16,17]. In this work, we have chosen to study the intermetallic Ni-Al compounds, which is interesting in the industry field. It can enhance the performance and stability of catalysts, turbine blades, ferroelectric capacitors and vanes due to their several attractive properties including excellent oxidation resistance up to 1573 K, as well as good thermal conductivity and high chemical stability [18–21]. In this sense, experimental and computational techniques have tried to develop advanced NiAl materials with engineering applications. The mechanical alloying method was used by Zadorozhnyy et al. [22] to fabricate NiAl intermetallic for coatings. They showed that the microstructure of mechanically alloyed coatings depends on the substrate hardness. Cao et al. [23] used the molecular dynamics simulation and they found that the NiAl alloy's hardness was greater than that of pure Ni metal. Recently, we have performed molecular dynamics (MD) simulation to study the vitrification of single-component metallic glasses of Al and Ni [24,25]. We found that the icosahedral cluster type is important for the glass forming ability in both systems. In the present work, we focus on the mixture of Ni and Al to give new insights into the atomic level structure of the amorphous Ni_3Al alloy using molecular dynamics (MD) simulation. Various structural analysis methods are used to investigate the evolutions of the different clusters in the medium range order (MRO) and their connections during the glass transition. Even less understood is the origin of the splitting in RDF. A previous simulation work [26] has shown that both of the subpeaks are due to the face- and vertex shared connection between atomic clusters. In another paper [27], it is shown that the first subpeak is caused by the face-sharing of icosahedral clusters while the edge- and vertex-sharing contribute to the second subpeak in RDF. From our analysis, we describe the correlation between the splitting of second peak in the RDF and icosahedral-like structures in the glassy state. The cooling rate used in our study is much higher than any realistic experimental cooling rate, so our findings cannot be reliably compared to any experimental observations to reveal other features of structural properties of metallic glasses.

In the next section of this paper (Section 2), we present a brief review of the MD technique and the structural analysis methods used to unravel the basic local cluster in Ni_3Al alloy. In a third section, some detailed structural characteristics of amorphous Ni_3Al will be discussed and a special attention will be paid to relationship of linked clusters with the splitting in RDF. In a fourth and last section, we close this work by emphasizing our main conclusions.

2. Simulation conditions and analysis methods

To delineate the atomic arrangements from short to medium range order of binary Ni-Al BMG, the molecular dynamic (MD) simulations was carried out by using the LAMMPS code (large scale atomic/molecular massively parallel simulator) [28,29]. The starting configuration of the model system that represents the Ni_3Al compound has L12 ordered structure containing 13,500 atoms (10125 Ni and 3375 Al) in a 3D cubic box of size 15 Å × 15 Å × 15 Å with periodic boundary conditions applied along the three directions. We mentioned that this box size was considered after an examination of different systems with different number of atoms. The system was first heated-up from 200 K to 3500 K, which is well above the melting temperature. The heating process is performed under the isothermal isobaric (NPT) ensemble where atom number (N), pressure (P), and temperature (T) are conserved. Then, the supercell was relaxed at the temperature of 3500 K for 100 ps under the NVT ensemble (i.e. the number of particles N, the system volume V and the temperature are kept constant). The temperature was then gradually decreased from 3500 K to 200 K at the isothermal isobaric (NPT) ensemble at a uniform cooling rate of 10^{13} K/s.

The interatomic potentials is the most important issue in molecular dynamics simulation. For describing the interaction between Ni–Ni, Ni–Al, Al–Al, we use the force fields in the generalized form of the embedded-atom method (EAM) [30]; which is better able to reproduce some basic features of cubic metallic systems. The EAM is a semi-empirical potential based on concepts coming from density functional theory. This potential represents the total energy E_{tot} of a collection of atoms, which consist of an embedding energy function and a pairwise potential function, and is given by [31–33]:

$$E_{tot} = \sum_i F_i(\rho_i) + \frac{1}{2}\sum_{i\neq j}\varphi_{ij}(r_{ij}) \quad ; \tag{1}$$

here, $\varphi_{ij}(r_{ij})$ is the suitable pairwise potential with r_{ij} represents the distance between atoms i and j. ρ_i is the host density associated with atom i which is closely approximated by the total atomic densities given by:

$$\rho_i = \sum_{i\neq j}\rho_j^{\alpha}(r_{ij}) \quad ; \tag{2}$$

The EAM potential for Ni-Al system was constructed by fitting to properties of individual elements Al and Ni followed by fitting a cross-interaction potential Ni-Al.

For both Ni and Al, the electron density is given by:

$$\rho(r) = \psi\left(\frac{r-r_c}{h}\right)[A_0 z^y e^{\gamma z}(1 + B_0 e^{-\gamma z} + C_0)]; \tag{3}$$

where $z = r - r_0$.

The pair-interaction function is parameterized in a generalized Lennard–Jones form, which is expressed as:

$$\varphi(\mathrm{r}) = \left[\frac{V_0}{(b_0 - b_1)}\left(\frac{b_2}{z^{b1}} - \frac{b_1}{z^{b2}}\right) + \delta\right]\psi\left(\frac{\mathrm{r\text{-}r_c}}{\mathrm{h}}\right), \tag{4}$$

where $z = r/r_1$.

The cross potential representing the interactions between Ni and Al atoms was constructed also by employing the generalized Lennard–Jones form given by Eq. (4). The optimized values of the fitting parameters for the pairwise and embedding functions are adapted from reference [30].

3. Results and discussion

In this section, we present our results concerning the atomic configurations for our simulated system. For this reason, we have used a range of analysis techniques to effectively characterize the obtained metallic glass and extract the key structural features relevant to the fundamentals of glass formation. Among them, we have firstly used the radial distribution function (RDF) to verify that the Ni_3Al alloy is indeed solidified as a glass. In the case of the crystal, thanks to the periodicity and the lattice symmetries, an X-ray or neutron diffraction measurement is sufficient to characterize the structure of the crystal. By against, the structural study of disordered materials such as liquid and glass is limited to the determination of the short to medium range scale by the radial distribution function. The RDF describes how density varies as a function of the distance r from a reference particle and it can be calculated as [34]:

$$g(r) = \frac{V}{N^2}\left\langle\sum_{i=1}^{N}\frac{n(r)}{4\pi r^2\Delta r}\right\rangle; \tag{5}$$

where N is the number of atoms, V represents the volume of the system and $n(r)$ denotes the number of particles found in the shell from r to $r + \Delta r$. Fig. 1 illustrates the RDF curves for different temperatures during the cooling process of Ni_3Al alloy. At high temperatures (above 1500 K), the calculated RDF shows a pronounced peak at the shortest distance and the oscillations that follow it have an intensity that decreases, showing the liquid state because there is no correlation