

Thermophysical properties of undercooled liquid Ni-Zr alloys: Melting temperature, density, excess volume and thermal expansion



P. Lü, K. Zhou, X. Cai, H.P. Wang*

MOE Key Laboratory of Space Applied Physics and Chemistry, Department of Applied Physics, Northwestern Polytechnical University, Xi'an 710072, PR China

ARTICLE INFO

Article history:

Received 14 November 2016

Received in revised form 27 March 2017

Accepted 6 April 2017

Keywords:

Thermophysical properties

Undercooling

Liquid alloys

Ni-Zr alloys

ABSTRACT

The thermophysical properties of undercooled liquid Ni-Zr binary alloys were investigated by molecular dynamics simulation combined with a Finnis-Sinclair (F-S) potential, including melting temperature, density, excess volume and thermal expansion. The melting temperatures were obtained by the evolution of crystal-liquid-crystal sandwich model, where there exist rather low differences of 4.14% for Ni_{77.8}Zr_{22.2} alloy and 3.98% for Ni₅₀Zr₅₀ alloy when they were compared with the reported values. The calculated densities of liquid Ni-Zr alloys increase with the decrease of temperature, which agree well with the reported experimental values except for the Ni-rich composition alloys. Thus, the reported experimental density of liquid Ni_{77.8}Zr_{22.2} alloy was employed to re-gauge the current F-S potential and the densities of the Ni-rich composition alloys were recalculated by the re-gauged potential. This binary liquid alloy system shows a negative excess volume, which could be attributed to the strong attractive interactions between Ni and Zr atoms. It is indicated that the Ni-Zr alloy system seriously deviates from the ideal solution, and the accuracy would be very low if the thermophysical properties are estimated by Neumann-Kopp rule. Meanwhile, the thermal expansion coefficients were also derived on the basis of the density data, which increase with the enhancement of temperature except for liquid Ni_{77.8}Zr_{22.2} alloy.

© 2017 Elsevier B.V. All rights reserved.

1. Introduction

The thermophysical properties of undercooled liquid alloys, such as melting temperature, density and thermal expansion, are fundamental parameters in the fields of materials science and condensed matter physics, which have aroused great interests [1–3]. The melting temperature plays a vital role in determining whether the liquid alloy is in normal state or undercooled state [4]. The densities of liquid alloys are of considerable importance in understanding solidification processes, mass transport, atomic structure, thermal convection and performing numerical simulations [5–9]. Thermal expansion, which can be derived from the density as a function of temperature, is also important for performing engineering design and studying the phase transformation [10,11]. However, the knowledge of these properties of undercooled liquid alloys is scarcely known compared with that of pure liquid metals and normal liquid alloys. This is because that any contact between melts and container walls will trigger immediately nucleation, and thus the metastable state of undercooled melts is difficult to be achieved and kept. To date, these thermophysical properties can

be obtained mainly by the experimental and computing methods. Containerless processing techniques, for example, electromagnetic levitation and electrostatic levitation, are employed to experimentally determine these properties, which are the best methods to obtain reliable data. Nevertheless, large time consumes and financial costs are always required during the experimental measurements, and thus the efficiency is rather low. More importantly, high undercooling is hard to achieve although the contact between melts and container walls has been completely avoided. As an alternative, the molecular dynamics (MD) simulation together with a reasonable potential is considered to be a powerful method to investigate the thermophysical properties of liquid metals [12,13]. It is easier to obtain high undercoolings and to evaluate the thermophysical properties for pure metals and simple alloys, especially for some dilute alloys. But for the complicated alloys, there still exist various problems to obtain reliable thermophysical properties.

As a typical metallic glass system, Ni-Zr binary system has aroused great interests because of its good glass forming ability in a wide composition range and abundant intermetallic compounds [14,15]. Most works focus on its structure and dynamics properties. Recently, Georganakakis et al. [16] investigated the local structure of Zr-Ni metallic glasses and found their structures cannot be approached with an ideal solution model. Holland-Moritz

* Corresponding author.

E-mail address: hpwang@nwpu.edu.cn (H.P. Wang).

et al. [17] revealed a large activation energy for Ni-diffusion in liquid $\text{Ni}_{36}\text{Zr}_{64}$, which may result from a peculiar short order of the Ni-Zr melts. Whereas, only a small amount of thermophysical properties of undercooled Ni-Zr alloys have been reported. And these properties of undercooled liquid Ni-Zr alloys are scarce and in lack of detailed and systematic studies, which is quit essential for fundamental researches and important to improving industrial processing.

In this paper, we presented the calculations of the thermophysical properties of various Ni-Zr liquid alloys including melting temperature, density, and thermal expansion. We used the reported experimental density of liquid $\text{Ni}_{77.8}\text{Zr}_{22.2}$ alloy to re-gauge the current F-S potential and then recalculated the density for Ni-rich alloys with the gauged potential. Comparisons between the experimental and calculated data were performed to evaluate the present calculations. Meanwhile, the origin of the excess volume in liquid Ni-Zr binary alloy system was also discussed.

2. Method

The inter-atomic potential is the fundament of molecular dynamic simulation. The F-S potential proposed by Finnis and Sinclair [18] has been believed to be successful for describing atomic interactions of metallic systems and applied in the simulation of liquid structure, liquid-glass transition and thermodynamic properties. In the F-S, the total energy E of the system is given by

$$E = \sum_{i=1}^{N-1} \sum_{j=i+1}^N \varphi^{\alpha_i \alpha_j}(r_{ij}) + \sum_{i=1}^N F^{\alpha_i}(\rho_i), \quad (1)$$

where F is the embedding energy which is a function of the atomic electron density ρ_i , N the number of atoms in the system, $\varphi^{\alpha_i \alpha_j}(r_{ij})$ the pair potential interaction, α_i the element type of atoms i , r_{ij} the distance between atoms i and j . The electron density ρ_i is calculated by $\rho_i = \sum_j f^{\alpha_i \alpha_j}(r_{ij})$. The functions $\varphi^{\alpha_i \alpha_j}(r_{ij})$ and $f^{\alpha_i \alpha_j}(r_{ij})$ are the results of empirical modeling. Recently, a F-S potential for Ni-Zr binary alloy system was put forward by Mendeleev [19], which was developed to match properties of Ni, Zr, NiZr and NiZr_2 . This potential has been employed to simulate the solid-liquid interface properties of Ni-Zr B33 phase successfully and is also applied in this work.

2.1. Melting temperature

For the melting temperature, simulations were performed in orthorhombic boxes with periodic boundary conditions in three dimensions. We used a sandwich model [20] to simulate the melting behavior of metals. To generate the initial configuration of crystal-liquid-crystal coexisting structure, an orthorhombic box was employed in the simulation, which was swarmed with atoms with stable lattice. The crystal-liquid-crystal multi-layers were arranged in the z direction. The liquid phase was generated by heating the central half of crystal in z direction at 1000 K above the melting temperature (far beyond the melting temperature) for 100,000 steps. The solid phase was obtained by equilibrating the rest of the crystal around the experimental melting temperature for 100,000 steps. As a result, the coexisting structure was constructed and annealed at given temperatures for 5,000,000 steps, which was monitored during this process. The simulated melting temperature can be estimated from the evolution of the coexisting structure, which was determined according to the change of energy of per atom. If the given temperature is higher than the melting temperature, the liquid phase will grow on the expense of the solid phase, and contrariwise lower than the melting temperature.

2.2. Density

For the simulation of density of liquid alloys, 4000 atoms were put into a cubic box with the periodic boundary conditions in three dimensions. The system was under constant temperature and constant pressure. NPT algorithm was employed in the process of the simulations. The time step was set to 1 fs and the pressure was set to 1 bar. The temperature was adjusted every 50 steps. To obtain an equilibrium liquid state, the system started at 3000 K, which was far beyond the melting temperature. The initial temperatures were kept constant for 100,000 steps. Then, the system was cooled with the cooling rate of 10^{12} K/s in 100 K temperature interval. At each temperature, 100,000 steps were performed for equilibrium. All the MD simulations were performed with LAMMPS.

3. Results and discussion

3.1. Melting temperature

The simulated melting temperature can be determined according to the change of internal energy of per atom. Fig. 1 shows the variations of internal energy per atom of pure Ni with time at two desired temperatures. It is obvious that the internal energy per atom of pure Ni increases in the first place and stabilizes subsequently at 1730 K presented in Fig. 1(a), indicating that the coexisting structure melts into the liquid state completely. Nevertheless, at 1725 K, the internal energy per atom decreases before it stabilizes, which gives an indication of the crystallization of the coexisting structure. To further confirm the states of the final structure at the desired temperatures, the pair distribution function is calculated according to the following equation:

$$g(r) = \frac{V \langle n_i(r, r + \Delta r) \rangle}{4\pi r^2 \Delta r N}, \quad (2)$$

where V is the system volume, $n_i(r, r + \Delta r)$ the atom number around the i th atom in a spherical shell between r and $r + \Delta r$, and N the

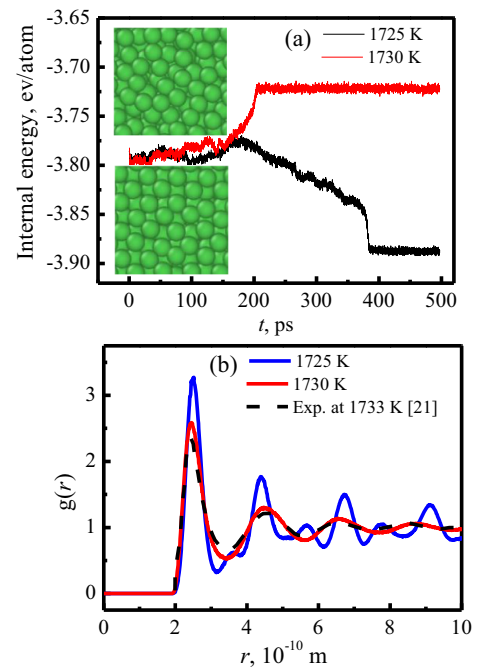


Fig. 1. (a) Variation of internal energy per atom for sandwich structure of Ni during simulation; (b) pair distribution functions of Ni at two different temperatures. The insets present the snapshots of atom positions at two different temperatures.

Download English Version:

<https://daneshyari.com/en/article/5453183>

Download Persian Version:

<https://daneshyari.com/article/5453183>

[Daneshyari.com](https://daneshyari.com)