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Glass formation and structural properties of $Zr_{50}Cu_{50-x}Al_x$ bulk metallic glasses investigated by molecular dynamics simulations

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ABSTRACT

Temperature effects on the structural evolution and the glass formation of $Zr_{50}Cu_{50-x}Al_x$ (x = 0, 10, 20, 30, 40, and 50) in the liquid and glassy states are studied by classical molecular dynamics simulations. In order to perform a comprehensive comparison and analysis, we consider the Honeycutt-Andersen indices, Voronoi analysis, radial distribution functions, coordination numbers, enthalpy, specific heat, and self-diffusion coefficients in our classical simulations in conjunction with the many body tight binding and embedded atom method potentials. The simulated structural properties were found to be in good agreement with available experimental data for Al poor concentration. We may conclude that the Al is a key element in glass transition and icosahedral ordering in considered systems, Zr-Cu-Al alloys have the best GFA until the concentration of Al in ternary alloy reaches the value of 20% and the parameters of TB model potentials for Al need to improve to explain the aggregation of Al atoms in ternary Zr-Cu-Al alloy.

Contents

1. Introduction

The first successful preparation of a metallic glass was reported by Duwez's group in 1960 [1]. The first bulk metallic glass (BMGs)

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was the Pd-Cu-Si alloy prepared by Chen in 1974 [2]. The successful synthesis of these BMGs has opened the door for the rapid development of BMGs in the last decades. Recently, studies in particular on Cu-based and Zr-based BMGs increased significantly. Among them Zr-based BMGs alloys have many interesting properties, which are considered as potential structure materials [3]. Compared to other alloys, Zr-based BMGs alloys have become the most successful and promising for the discovery and application of BMGs. Furthermore, Zr-based BMGs alloys exhibit large supercooled liquid regime $\Delta T_x = T_x - T_g$, where T_x and T_g are the crystallization and glass transition temperatures, respectively [4,5]. The binary Zr-Cu alloys system are known to form glassy phase on quenching in a wide range of compositions [6]. In order to investigate atomic structures, thermal stability, topological order, chemical and mechanical properties of the binary Zr-Cu systems in a wide compositional range, a number of studies both theoretically and experimentally have been carried out [7–9].

The effect of Al on the thermal stability of supercooled liquid state, the local structures of binary Zr-Cu and ternary Zr-Cu-Al metallic glasses have been studied by Sato et al. using X-ray diffraction and X-ray absorption fine structure (EXAFS) measurement [10]. Recently, Georgarakis et al. [11] showed that atomic structure of Zr–Cu glassy alloys and detection of deviations from ideal solution behavior with Al addition by x-ray diffraction using synchrotron light in transmission, and the addition of Al to Zr–Cu BMGs changes the atomic structure in the short and medium range order because of the strongly attractive interaction between Al and Zr atoms. The atomic structure of ternary Zr–Cu–Al metallic glasses have been previously studied by X-ray diffraction using high-energy synchrotron radiation [8,12,13]. The thermal stability, glass forming ability (GFA), volume and viscosity of Zr–Cu–Al BMGs alloys have been investigated by using X-ray diffractometry (XRD) [14–17]. Ma et al. [18] have studied the phase transformation from the B2 to B19 of $Cu_{46}Zr_{46}Al_8$ BMG via cryogenic treatment. Antonowicz at al. [9] have investigated the local atomic structure of Zr–Cu–Al amorphous alloys by using EXAFS method. A recent study regarding the influence of substitution of Cu by Al on the martensitic transformation temperatures in $Zr_{50}Cu_{50-x}Al_x$ alloys ($x = 0, 2, 4, 6, 8, 10$) have been reported by Meng et al. [19]. They have concluded that the crystalline to amorphous transformation of these alloys are affected by high pressure torsion effects [20]. In addition, Hermann et al. [21] have studied the cooling rate dependence of icosahedral short-range order in ternary Cu–Zr–Al BMGs alloy. Bo et al. [22] have calculated some thermodynamic properties of undercooled liquid alloys for Zr–Cu–Al system and the authors have deduced that it is not appropriate to explain the GFA with only the thermodynamic properties of liquid phase. On the theoretical side, molecular dynamics (MD) simulations with EAM potentials have been performed to study the atomic structure of $Cu_{46}Zr_{47}Al_7$ BMG alloy by Cheng et al. [23]. Authors have reported that percentage of Al in alloy leads to increased population of ideal icosahedra. Also, Wang et al. [24] have used the same method to investigate the effects of the concentration of Zr in $Zr_xCu_{90-x}Al_{10}$ ($20 \leq x \leq 70$) and showed that the small addition of Al concentration in Zr-Cu alloys exhibits more prominent icosahedral ordering. Deb Nath [25] has studied effects of concentration of Zr on the stiffness and strength of same series of BMGs.

These studies aforementioned have concentrated to specific Zr-Cu-Al systems whose content of Zr and Cu is almost equal and minor addition of Al is considered. In this work, we have studied the concentration dependent structural and thermodynamic properties of $Zr_{50}Cu_{50-x}Al_x$ ($0 \leq x \leq 50$) alloys using many body tight-binding (TB) potential coupled with MD simulations. Our aim is to give a comprehensive analysis, a complete view of structural evolution of $Zr_{50}Cu_{50-x}Al_x$ ($0 \leq x \leq 50$) alloys. In order to check the transferability of TB potential to glassy systems considered in present work, we compare the TB results with two widely-used EAM forms proposed by Zhou et al. [26] and Cheng et al. [23]. The simulations were performed via well-known simulation code of DL_POLY [27]. In our previous studies, we showed that the TB potential was successful in determining the GFA and structural properties of ternary $Cu_{50}Ti_{25}Zr_{25}$ [28], $Zr_{50}Cu_{20}Fe_{20}$ [29] and binary $Cu_{50}Ti_{50}$ [30], $Zr_{70}Pd_{30}$ [31] alloys. In order to investigate the effect of Al content within Zr-Cu-Al BMG alloy on glass transition and icosahedra short range order, the energetic curves, the pair analysis technique and the radial distribution functions are considered.

2. Simulation details

2.1. Potential functions

The reliability of the MD simulation depends on the interatomic potential that can explain better the atomic interactions of the system. In the potential function of TB which is an effective model for classical MD simulations, the total cohesive energy E_C^i at an atomic site i is written as

$$E_C^i = \sum_i \left(E_R^i + E_B^i \right) \tag{1}$$

Based on the TB model [32], the energy of a single atom can be divided into two parts. One is the attractive potential to bind atoms together:

$$E_B^i = -\left\{ \sum_j \xi_{\alpha\beta}^2 exp\left[-2q_{\alpha\beta}\left(\frac{r_{ij}}{r_o^{\alpha\beta}} - 1 \right) \right] \right\}^{1/2} \tag{2}$$

The other is the repulsive potential:

$$E_R^i = \sum_j A_{\alpha\beta} exp\left[-p_{\alpha\beta}\left(\frac{r_{ij}}{r_o^{\alpha\beta}} - 1 \right) \right] \tag{3}$$

where r_{ij} is the distance between atoms i and j; and $r_o^{\alpha\beta}$ is the nearest neighbors distance in the α and β lattice. The α and β variables represent different lattice unlike neighboring atoms. A, p, ζ, and q are model parameters connected with the physical properties of the elements. The parameters of $A_{\alpha\alpha}$, $p_{\alpha\alpha}$, $\xi_{\alpha\alpha}$ and $q_{\alpha\alpha}$ ($\alpha = \beta$) for pure solid elements taken from Cleri et al. [32] are given in Table 1. The parameters $r_o^{\alpha\alpha}$ are taken to be the nearest neighbor distance of pure elements. To describe the pair interaction between different types of atoms ($\alpha \neq \beta$), for the hetero-interactions, an arithmetic mean was taken for the parameters of $p_{\alpha\beta}$, $q_{\alpha\beta}$ and $r_o^{\alpha\beta}$, while a geometric mean was taken for parameters related to the strength, $A_{\alpha\beta}$ and $\xi_{\alpha\beta}$.

2.2. Computational procedure and analysis method

Classical MD simulations with TB and EAM potentials were performed by using DL_POLY simulation package with the isobaric

Table 1
TB potential parameters for pure Zr, Cu, and Al metals.

Metal	A(eV)	ξ(eV)	p	q	r_0(A)
Zr-Zr	0.1934	2.2792	8.2500	2.249	3.170
Cu-Cu	0.0855	1.2240	10.960	2.278	2.556
Al-Al	0.1221	1.3160	8.6120	2.516	2.863