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Liquid -to-glass transition in bulk glass-forming $Cu_{55-x}Zr_{45}Ag_x$ alloys using molecular dynamic simulations

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Abstract. We report results from molecular dynamics (MD) studies concerning the microscopic structure of the ternary, bulk metallic glass-forming Cu_{55-x} $Zr_{45}Ag_x$ (x=0,10,20) alloys using tight-binding potentials. Understanding of the nature of Glass Forming Ability (GFA) of studied alloys, GFA parameters, glass transition temperature (T-g), melting temperature (T-m), reduced glass transition temperature (T-g/T-m), the supercooled liquid region and other parameters were simulated and compared with experiments. The computed pair distribution functions reproduce well experimental x-ray data of Inoue and co-workers. Structure analysis of the Cu-Zr-Ag alloy based on MD simulation will be also presented.

1 Introduction

Bulk metallic glasses (BMGs) are promising materials because of their superior properties and relatively lower materials cost. Their low glass-forming ability (GFA) and poor plasticity are points that need further improvement. Recent studies have been interested in order to develop new BMG with improved glass forming ability (GFA). Related to this the Cu-Zr binary alloy system is regarded as a good model system for the investigations on the effect of minor elements added for improvement of both GFA and plasticity [1-7]. Recently, BMG formation in Cu-Zr-Ag alloys has been studied by Inoe, Mattern and co-workers [1-3, 5-7]. The glass forming ability (GFA), crystallization kinetics and structural changes in the family of Cu_{55-x} $Zr_{45}Ag_x$ alloys have also been analyzed by adding Ag to Cu-Zr alloys [1-7]. On the other hand, a few simple criteria were developed in the literature of metallic glass research to explain the GFA of alloys [8-10]. In addition to this, there are less computational approaches proposed to explain the GFA of alloys [8]. According to our knowledge, there are no published simulated structural results for the analysis of GFA ability of the $Cu_{55-x}Zr_{45}Ag_x$ (x=0, 10, 20) alloys. There has been only one reported result of Monte Carlo (MC) simulations performed by the modified embedded atom type (MEAM) potentials [11]. It has been shown that, the MEAM potential reproduced the occurrence of the phase separation of Ag rich phases in supercooled Cu-Zr-Ag liquid alloy that is in a good agreement with experimental data [4] and CALPHAD-type thermodynamic calculation [12]. However, recent thermodynamic calculations [13] have predicted the amorphous formation diagram of the Cu-Zr-Ag system based on the available experimental data [4, 5]. Thus the miscibility gap of the liquid phase that was not considered in the calculations of He et al. [12] has been re-optimized by Kang and Jung [13].

In the present work, the GFA ability of ternary $Cu_{55-x}Zr_{45}Ag_x$ (x=0, 10, 20) systems are firstly evaluated by molecular dynamics (MD) calculations in which the atomic interactions are modeled with a many body type potential as tight binding (TB) potential (called as n-body Gupta potential) proposed by Cleri and Rosato [14]. In order to investigate the liquid to glass transition in Cu-Zr-Ag BMGs on an atomic level, TB potentials for the pure Cu, Zr and Ag has been used to newly developed for the ternary Cu-Zr-Ag system. This potential yields the liquid to glass transition in the ternary $Cu_{55-x}Zr_{45}Ag_x$ (x=0, 10 ,20) alloys, in a good agreement with recent calculations of Kang and Jung [13].and also experimental data [4].

2 Method

As a first step in MD simulations, it is necessary to describe the atomic motions. In this study we have used a quantum mechanics based TB potential [12] to describe the atomic motions and to explain the structural properties. In TB approach, the total potential energy of a system is given the following form :

$$E = \sum_i \left[A_{ij} \sum_{j \neq i} \exp\left[-p_{ij}\left(\frac{r_{ij}}{r_{0ij}} - 1\right)\right] - \left(\xi_{ij}^2 \sum_{j \neq i} \left[-2q_{ij}\left(\frac{r_{ij}}{r_{0ij}} - 1\right)\right] \right)^{1/2} \right] \quad (1)$$

Up to date no works dedicated the GFA or BMG forming of the Cu-Zr-Ag ternary system by MD simulations with TB potentials.

The presented $Cu_{55-x}Zr_{45}Ag_x$ (x=0, 10 ,20) alloys also show a typical feature of glass formation, that quenching from liquid to 300K at the cooling rate of 0.5K/ps leads to the metallic glasses. As known that splitting of the first maximum in the total PDF indicates the lower GFA. The experimental and simulated data are in good agreement for $Cu_{55-x}Zr_{45}Ag_x$ (x=10, 20) alloys

MD results are agree well with the Inoue's empirical rule [10] for the increasing of T_g at the cooling rates of 5 and 0.5K/ps. The simulated glass transition temperature T_g of $Cu_{45}Zr_{45}Ag_{10}$ ternary alloy is about 680K at the cooling rate of 0.5K/ps that is 4K lower than the reported experimental value given in Ref [1].

For the MD simulations in the cooling rates of 5 and 0.5K/ps the T_g values of the ternary alloys remains almost unchanged with increasing Ag content to $Cu_{55}Zr_{45}$ binary alloy, as indicated experimentally in Ref.[7]. The MD results are agree well with experimental and thermodynamic data [6,13]. It has been shown experimentally that Ag content significantly affects the liquidus temperature T_l and melting behaviour of these alloys. The lowest T_l and highest T_g/T_l was observed for $Cu_{45}Zr_{45}Ag_{10}$ alloy case while the MD simulations predict that the highest value of T_{rg} is achieved for $Cu_{55}Zr_{45}$ alloy.

According to the recent thermodynamic data given in Ref. [13], the presented compositions of $Cu_{55-x}Zr_{45}Ag_x$ (x=0, 10, 20) alloys have been shown as fully amorphous sate that is in good agreement with our MD results.

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