



Formation and structure of Cu–Zr–Al ternary metallic glasses investigated by ion beam mixing and calculation

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ARTICLE INFO

Article history:

Received 9 October 2011
Received in revised form 10 January 2012
Accepted 10 January 2012
Available online 28 January 2012

Keywords:

Cu–Zr–Al system
Ion beam mixing
Amorphous phase
Structure factor

ABSTRACT

The $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ amorphous alloys are synthesized in metallic multilayered films by ion beam mixing in the present study. Formation mechanism of amorphous phases is discussed in terms of the atomic collision theory. To further analyze the structure and composition of the obtained Cu–Zr–Al metallic glasses, structure factor $S(q)$ is calculated based on the results of molecular dynamics simulations. It is found that the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ metallic glasses are mainly consisted of Al–Zr and Al–Al, Cu–Cu amorphous phases, respectively. The calculated results turn out to match well with the experimental observation.

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1. Introduction

Since the first finding of millimeter-diameter glassy rods in the Pd–Cu–Si system [1], the bulk metallic glasses (BMGs) have attracted tremendous attention due to their superior mechanical properties and excellent corrosion resistance [2–5]. In particular, there has been a huge interest in the Cu–Zr–Al system currently, which is expected to be an advantaged candidate for BMGs because of its high glass-forming ability (GFA) [6,7]. A series of Cu–Zr–Al based BMGs or BMG composites have also been obtained in experiments. The obtained BMGs exhibit extraordinarily high strength and good compressive ductility [8–11]. To clarify the atomic structures and compositions of the Cu–Zr–Al BMGs, investigations have been conducted from both experimental and theoretical aspect, such as using molecular dynamics (MD) simulations by Wang et al. [12] and in situ synchrotron X-ray scattering measurements by Fan et al. [13].

To study the glass-forming-ability or glass-forming-region of the Cu–Zr–Al system, the long range smoothed second-moment-approximation of tight-binding potential for the Cu–Zr–Al system has been constructed by fitting the physical properties of Cu, Zr, Al and their compounds [14]. Based on the constructed potential, MD simulations were carried out using solid solution model to compare the relative stability of the crystalline solid solution versus its disordered counterpart as a function of solute concentration. Simulations reveal that the physical origin of metallic glass

formation is crystalline lattice collapsing while solute concentration exceeding critical value, thus predicting a hexagonal composition region, within which the Cu–Zr–Al ternary metallic glass formation is energetically favored, as shown in Fig. 1 [14].

For the experimentally obtained Cu–Zr–Al metallic glasses, the great majority of their compositions fall within the predicted glass-forming favored region. Further inspecting Fig. 1, one may find that the experimental work has so far been focused on the Cu–Zr-based system with minor addition of Al [15–17]. In the present study, two Cu–Zr–Al multilayered films were designed with the overall compositions of $\text{Cu}_{12}\text{Zr}_{40}\text{Al}_{47}$ and $\text{Cu}_{32}\text{Zr}_{6}\text{Al}_{62}$, which are exactly in the predicted hexagonal region. On one hand, we can study the metallic glass formation in Al-rich composition, because whether the Cu–Zr–Al metallic glasses can be obtained in the Al–Zr-based or the Al–Cu-based system by experiment is still an open issue. On the other hand, the experimental results can also serve as a comparison with the MD simulations.

2. Cu–Zr–Al ternary metallic glasses formed by ion beam mixing

Considering the individual and total thickness of Cu–Zr–Al multilayered films, the samples were both designed to be 8 layers. Two sets of initial multilayered films were prepared by depositing alternatively pure Cu (99.99%), Zr (99.99%) and Al (99.99%) at a rate of 0.2 Å/s onto NaCl single crystal substrates in an e-gun evaporation system with a vacuum level on the order of 10^{-7} Pa. The as-deposited Cu–Zr–Al multilayered films were then irradiated by 200 keV xenon ions in an implanter with a vacuum level better than 5×10^{-4} Pa and the irradiation dose was in a range from

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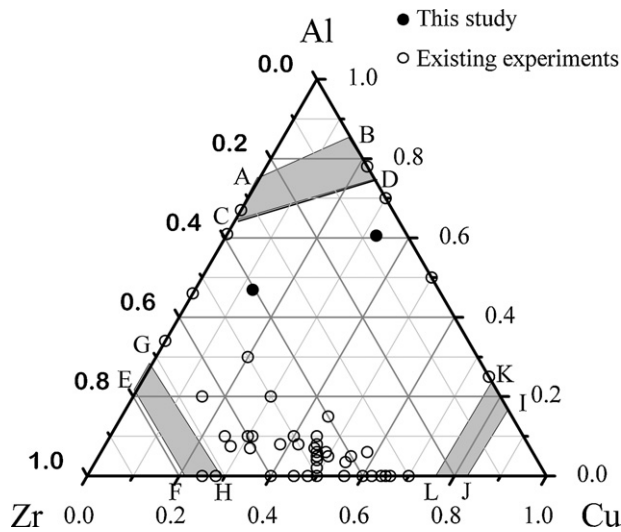


Fig. 1. The crystal-amorphous state phase diagram derived from MD simulations for the Cu–Zr–Al ternary system [11], and the “blank” region delimited by the contour line CDKLHG predicts the formation of amorphous phase.

8×10^{14} to 5×10^{15} Xe^+/cm^2 . During irradiation, the sample holder was always cooled by liquid nitrogen (77 K) and the ion current density was confined to be about $2 \mu\text{A cm}^{-2}$ to avoid overheating effect. The as-deposited and irradiated Cu–Zr–Al samples were examined by transmission electron microscopy (TEM) and high resolution transmission electron microscopy (HRTEM) for structural characterization. X-ray fluorescence (XRF) was used to determine the compositions of the samples.

The composition of the two sets of as-deposited samples are determined by XRF to be $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$, which are exactly located in the amorphous formation region predicted by MD simulation [14]. Metallic glasses were obtained in the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ samples upon ion beam mixing (IBM) in irradiation dose ranges of 3×10^{15} to 5×10^{15} and 1×10^{15} to 5×10^{15} Xe^+/cm^2 , respectively. The HRTEM graphic and the selected area diffraction (SAD) pattern of the irradiated $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ sample are shown in Fig. 2(a) and (b), respectively. From Fig. 2(a) and (b), it can be seen that a unique amorphous phase was synthesized in the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ samples. Fig. 2(a) presents the micro-morphology of an homogeneous metallic glass, and Fig. 2(b) shows the halo due to the amorphous structure. In the cases of the $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ irradiated sample, the bright field image and selected area diffraction (SAD) pattern are shown in Fig. 2(c) and (d), respectively. Fig. 2(c) shows the bright field image with a contrast that can be explained by the presence of an amorphous dual-phase resulting from a phase separation. The SAD pattern of Fig. 2(d) shows a unique broad halo as a consequence of the overlapped halos of the two amorphous phases, indicating a quite similar structure of the two metallic glasses. For further analysis, energy-dispersive spectrum (EDS) was utilized to determine the composition of the separated phases formed in the $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ samples, however, the composition differences could not be detected possibly due to the limitation of instrument accuracy.

By comparing the halos shown in Fig. 2(b) and (d), it can be found that their locations are varied, showing that there are some structural or compositional differences existing between the two amorphous phases. Based on the camera constant and measured radius of halos in SAD patterns, the average distances of nearest-neighbor atoms were estimated to be 2.30 Å and 2.76 Å for the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ metallic glasses,

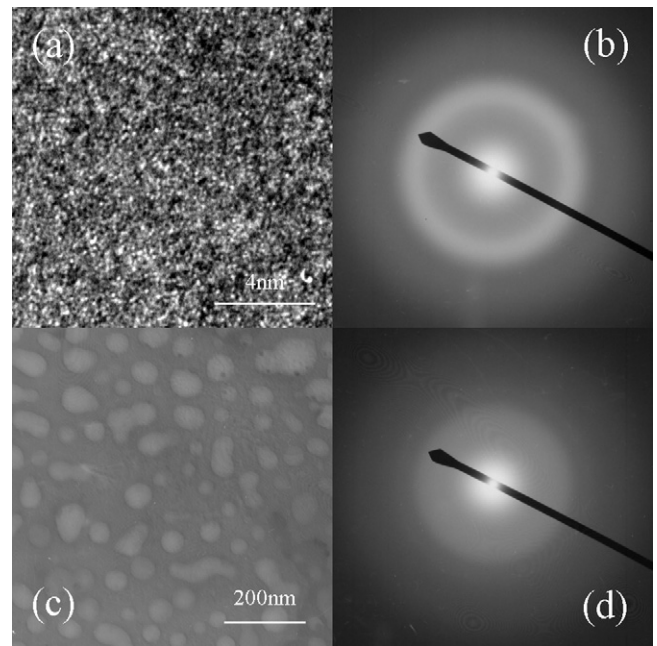


Fig. 2. HRTEM image (a) with the corresponding SAD pattern (b) of the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ alloy and bright field image (c) with the corresponding SAD pattern (d) of the $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ alloy after an irradiation dose of 3×10^{15} Xe^+/cm^2 .

respectively. The structures and compositions of the two metallic glasses will be discussed in detailed in Section 3.

According to the atomic collision theory [18,19], the process of IBM in the Cu–Zr–Al multilayered films occurs as follows: initially, a sequence of ballistic collisions induce the interfacial mixing in the Cu–Zr–Al multilayered films upon ion irradiation, and after irradiation to an adequate dose, atomic mixing would result in forming a highly energetic and disordered Cu–Zr–Al atomic mixture. When the atomic collision cascade is eventually terminated, the highly energetic mixture has to relax toward equilibrium. However, the relaxation period is extremely short, lasting for about 10^{-10} s, so only a very minor atomic rearrangement could take place. Thus, the complete relaxation of the highly energetic state may become frustrated, and therefore the disordered state was preserved, leading to the formation of amorphous phases in the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ samples.

3. Structural quantities of the Cu–Zr–Al metallic glasses

As shown above, the Cu–Zr–Al amorphous alloys were obtained by IBM in the $\text{Cu}_{12.6}\text{Zr}_{40.5}\text{Al}_{46.9}$ and $\text{Cu}_{32.7}\text{Zr}_{6.7}\text{Al}_{60.6}$ samples, and the compositions were right in the predicted glass-forming favored region by MD simulations. Interestingly, some structural or compositional differences were also found in the two amorphous phases synthesized by IBM. To develop a better understanding of the atom configurations in the amorphous alloys, structure factor $S(q)$ [20,21] as a function of wave vector q , was derived for the two Cu–Zr–Al alloys.

From the atom positions obtained in MD simulations, the structure factor $S(q)$ can be calculated as following. Specifically, in a cubic box, we may examine fluctuations for which $k = (2\pi/L)(k_x, k_y, k_z)$, where L is the box length and k_x, k_y and k_z are integers. One quantity of interest is the spatial Fourier transform of the number density [22]

$$\rho(k) = \sum_{i=1}^N \exp(i\vec{k} \cdot \vec{r}_i) \quad (1)$$

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