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SANS of bulk metallic ZrTiCuNiBe glasses

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Abstract

The evolution of decomposition and succeeding primary crystallization in the bulk amorphous $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ alloy have been studied by small angle neutron scattering. Samples annealed isothermally in the supercooled liquid and in the solid state exhibit correlation peaks indicating quasiperiodic inhomogeneities in the scattering length density. The peak positions vary as a function of temperature as predicted by the linear Cahn-Hilliard theory of spinodal decomposition. Variations of the Be-Ti composition ratio of the alloy leads to significant changes in scattering signals. The initially homogeneous alloy separates into two amorphous phases. In the decomposed regions, crystallization probability increases leading to polymorphic crystallization.

Keywords: SANS, bulk metallic glasses, decomposition, crystallization

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A unique family of multicomponent metallic glasses exhibits an excellent glass forming ability and a wide supercooled liquid region [1,2]. Their high thermal stability against crystallization has opened for the first time the opportunity to investigate thermodynamic and atomic transport properties in the supercooled liquid state of a metal [3-5]. Since these glasses exhibit high reduced glass temperatures, nucleation of crystalline phases from the homogenous multicomponent systems is suppressed to a great extent and the thermal stability is limited by alternative processes. We have reported the pathway to crystallization in $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ earlier [6,7]. First, this alloy phase separates, and in the decomposed regions crystallization probability increases leading to polymorphic nanocrystallization. Coarsening of the decomposed structure in the later stages of isothermal annealing is prevented by this nanocrystallization. This paper gives additional information on small-angle neutron scattering (SANS) investigations of the time- and temperature evolution of the decomposition process. In addition, the effect of the Be-Ti composition ratio on decomposition is also considered.

Amorphous $ZrTiCuNiBe$ samples were prepared from a mixture of the pure elements by induction melting on a water-cooled silver boat under Ti gettered Ar atmosphere. Disks with a thickness of 2.7mm were cut from these samples and isothermally annealed at temperatures between 613K and 673K for different times. SANS measurements were carried out at the Intense Pulsed Neutron Source at Argonne National Laboratory using the TOF small angle diffractometer [8].

Fig. 1 shows SANS data of $\text{Zr}_{41.2}\text{Ti}_{13.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{22.5}$ samples annealed at different temperatures between 613K and 673K. Data up to 663K show interference peaks, indicating spatially correlated arrangements of inhomogeneities. These data follow a relation $Q_{\text{max}}^2 \propto T$ (Q_{max} the peak position) consistent with the linear Cahn- Hilliard theory for spinodal decomposition [9] (Fig. 2). At 670K $Q_{\text{max}}=0$, and this probably is the spinodal temperature of $\text{Zr}_{41.2}\text{Ti}_{13.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{22.5}$. Accordingly, the sample annealed at 673K does not exhibit any peak. However, in the low Q region the $S(Q)$ varies as Q^{-1} implying the existence of a broad distribution of inhomogeneities. We assume that this sample is still located within the miscibility gap but outside the spinodal region of $\text{Zr}_{41.2}\text{Ti}_{13.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{22.5}$ (Fig. 3).

To investigate several states along the Be-Ti composition axis inside the miscibility gap of the system (Fig. 3), SANS data of $\text{Zr}_{41.2}\text{Ti}_x\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{36.3-x}$ samples ($x=8.8$, $x=13.8$, and $x=16.3$) annealed at 633K for 300min were measured (Fig. 4). All samples are partially crystallized (nanocrystals in an amorphous matrix) after this annealing. Correlation peaks are observed (with the spatial arrangement of nanocrystals given by that of the correlated decomposed regions) for samples with $x=13.8$ and $x=16.3$. However, the sample with $x=8.8$ does not exhibit a peak upon annealing. This sample is not located in the spinodal region. Possibly it is even outside the miscibility gap and thus shows thermally activated homogenous crystallization upon annealing. Interestingly, the $\text{Zr}_{41.2}\text{Ti}_{8.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{27.5}$ alloy has the same Be-Ti composition ratio as the $\text{Zr}_{46.7}\text{Ti}_{8.3}\text{Cu}_{7.5}\text{Ni}_{10}\text{Be}_{27.5}$ alloy that does not show decomposition at all [10].

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Figure captions

Fig.1: SANS of annealed $\text{Zr}_{41.2}\text{Ti}_{13.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{22.5}$ alloy samples. The line denotes a power law $S(Q) \propto Q^{-1}$.

Fig. 2: $Q_{\max}^2$ vs. temperature for $\text{Zr}_{41.2}\text{Ti}_{13.8}\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{22.5}$.

T_g is the glass transition temperature, T_c the spinodal temperature.

Fig.3: Schematic phase diagram of $\text{Zr}_{41.2}\text{Ti}_x\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{36.3-x}$

(1): $x=8.8$, (2): $x=13.8$ (3): $x=16.3$

Fig. 4: SANS of annealed $\text{Zr}_{41.2}\text{Ti}_x\text{Cu}_{12.5}\text{Ni}_{10}\text{Be}_{36.3-x}$ samples with compositions marked in Fig. 3.

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