Formation and Properties of Zr-(Ti, Nb)-Cu-Ni-Al Bulk Metallic Glasses

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The effect of composition variations and of B or Si addition on the glass forming ability and thermal stability of Zr-M-Cu-Ni-Al (M = Ti, Nb) bulk metallic glass-forming alloys have been investigated. The structural, acoustic, thermal and elastic properties of the bulk metallic glasses (BMGs) are studied by X-ray diffractometry (XRD), differential scanning calorimetry (DSC) and ultrasonic method. A small amount of boron and silicon addition can significantly affects the GFA and crystallization behaviors and microstructural nature of the glass-forming alloys. The small changes of the density and acoustic velocities in the BMGs relative to its corresponding crystallized state indicate a similar electronic state and similar atomic interactions between the glassy and crystalline states.

(Received September 4, 2000; Accepted December 26, 2000)

Keywords: bulk metallic glass, formation, properties, addition, microstructure

1. Introduction

Metallic glasses have long represented an intriguing class of materials.¹⁾ The recent discovery of bulk metallic glasses (BMG)¹⁻⁴⁾ offers excellent opportunities for the understanding of the metallic glassy state and for investigating the physical properties of the metallic glasses by using various methods. This discovery is significant for both fundamental and practical reasons. Zr-M-Cu-Ni-Al bulk metallic glass-forming systems, where M = Ti or Nb, is one of the new families of BMGs which have been found to exhibit exceptional glass forming ability (GFA), excellent thermal stability and unique physical properties.⁵⁻¹⁰⁾ The GFA and processability are comparable with those of silicate glasses. Therefore the BMGs can be processed by common methods available in a foundry.

These bulk metallic glass formers are very robust against heterogeneous nucleation at the surface or at interfaces. This leads to the development of bulk metallic glassy matrix composites by using the addition of crystalline materials.¹¹⁻¹³⁾ It is found that a small change of Nb composition results in a substantially improved GFA and ductility of the glass forming system.¹⁴⁾ A little dissolution of carbon (about 1 at%) into the Zr-Ti-Cu-Ni-Be metallic glass system can enlarge the thermal stability and hardness of the BMG.^{15,16)} The improved thermal stability of the matrix by dissolution of reinforcement has also been observed for Si addition in Cu-Ti-Zr-Ni BMG.^{17,18)} Recently, synthesis of a bulk ZrTiCuNiBe metallic glass matrix with ZrC particles¹⁹⁾ and a bulk Zr₅₅Al₁₀Ni₁₅Cu₃₀ metallic glassy matrix composite with ZrC particles²⁰⁾ have been reported. Directly introducing ceramics such as SiC, WC and ZrC particles into the BMGs also yields composites. This is a new way for synthesis of metallic glassy matrix composite alloys with improved mechanical properties. However, not much information exists on the effects of the metalloid Si and B addition on the glass forming alloys. It may be expected that Si and B would result in crys-

tallization, because they can react with metal constituents and form compounds with high melting temperature. On the other hand, B and Si with small atomic size may tighten the alloy structure and then stabilize the amorphous alloy against crystallization. Hence, studies of the effects of B and Si on the Zr-M-Cu-Ni-Al alloys are of special interest.

In this paper, the Zr-(Ti, Nb)-Cu-Ni-Al bulk metallic glasses have been obtained. The effects of the metalloid elements Si, B on the GFA and thermal stability of the systems have been investigated. X-ray diffractometry (XRD), differential scanning calorimetry (DSC) and ultrasonic measurements are applied to study the structure, the GFA and the thermal properties of the BMGs. Ultrasonic measurements provide a powerful tool for studying the structure of a matter. The larger geometry of the BMGs is very suitable for acoustic measurements. The differences between glassy and crystalline states in acoustic feature and long-wavelength phonons have significance for understanding of the unique microstructural characteristics and physical properties of the BMG.

2. Experimental

Ingots (about 50 grams) with nomination composition were prepared by arc-melting elemental metals in a Ti-gettered argon atmosphere. The purity of the elements used in preparing the specimens was as follow: Cu (99.9%), Ti (99.98%), Al (99.9%), Ni (99.9%), Zr (99.99%). The oxygen content of the Zr crystal bar was about 300 ppm. To reach a high homogeneity the specimens were remelted for several times. The ingots were crushed into small pieces, and melted by electromagnetic-levitation melting, and then die-cast into a copper mould under Ar atmosphere. This results in cylinders with 3 mm in diameter and 50 mm in length. The ejection temperature was about 200 to 300 K higher than the melting temperature of the alloy, and the ejection pressure was 60 kPa. Cross sections of the cast rods were cut and examined by XRD and DSC. The amorphous nature and the structure of these as-cast specimens were checked by XRD using a Philips PW 1050 x-ray diffractometer with Co K α radiation. The thermal stability and properties of the BMGs were

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examined using DSC at a heating rate of 20 K/min under Ar atmosphere (Perkin-Elmer DSC7). The melting temperature of the alloy was measured using high temperature DSC. The glassy rod was cut to a cylinder with a length of 7 mm for ultrasonic measurements. The ends of the cylinder were carefully polished flat and parallel before ultrasonic measurement. The acoustic velocities were measured by using a pulse echo overlap method.²¹⁾ The excitation and detection of the ultrasonic pulses were provided by X- or Y-cut (for longitudinal and transverse waves, respectively) 10 MHz quartz transducers. The travel time of ultrasonic waves propagating through the sample with a 10 MHz frequency was measured using a MATEC 6600 ultrasonic system with a measuring sensitive of 0.5 ns.

3. Results and Discussion

3.1 Zr-Ti-Cu-Ni-Al alloy

Figure 1 presents XRD patterns of the starting $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ master alloy, the amorphous $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ as-cast cylinder and of $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}$ -based alloys derived from the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG composition. Since it is known that Ti can be replaced by Nb, Cr or Ta,²²⁾ we have tried to substitute Fe for Ni (Fe and Ni have similar properties and atomic radius), and replaced Al by Mg in the alloys. As shown in Fig. 1, for these replacements crystalline peaks are superimposed on the broad diffuse scattering maxima of the amorphous phase, indicating only partial amorphization can be achieved for these alloys. These results indicate that the GFA of the alloy is sensitive to the composition, composition and cooling rate.

Figure 2 presents XRD patterns for the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$

Fig. 1 XRD patterns of the starting $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ master alloy, the amorphous $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ as-cast cylinder, and of $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}$ -based alloys derived from the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG composition.

Fig. 2 XRD patterns for the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ alloy with B and Si addition.

alloy with B and Si addition. 1 at%B addition does not change the amorphous structure within the detection limit of XRD. For more B addition (3 at%), strong crystalline diffraction peaks indicating the formation of crystalline boride can be seen. The situation for Si addition is markedly different from that of B addition. 1 at%Si addition leads to crystallization of the excellent glassy forming alloy and induces silicide formation, as shown in Fig. 2. Si, which can be absorbed easily from the quartz tube during melting and heat treatments of the alloy, is more reactive with the components and has great negative effect on the GFA than B addition.

Figure 3 displays a comparison of DSC traces for the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG and the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ alloy with 1 at%B or Si addition. All traces exhibit an endothermic characteristic of a glass transition followed by exothermic crystallization peaks at higher temperature. A remarkable feature of the BMG is a distinct large supercooled liquid region (SLR) as indicated in Fig. 3. The onset glass transition temperature, T_g , the onset temperature of the first crystallization event, T_{x1} , and the SLR defined as $\Delta T = T_{x1} - T_g$ of the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG are 653 K, 715 K and 62 K, respectively. The high temperature DSC shows that the melting temperature, T_m , of the alloy is 1080 K. This gives a reduced glass transition temperature $T_{rg} = T_g/T_m = 0.60$. The crystallization of the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG proceeds by a multistage crystallization process. The DSC trace exhibits three main heat release peaks. Xing *et al.*²³⁾ found that the first exothermic peak, which is sensitive to isothermal annealing near T_g , is related to a transformation from a short-range ordered (SRO) structure to a medium-range order (MRO) atomic arrangement of clusters/domains, yielding extremely fine primary quasicrystalline precipitates. The second peak mainly involves grain growth and decomposition of the primary quasicrystals. Figure 3 shows that B and Si addi-

Fig. 3 A comparison of DSC traces for the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG and the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ alloy with 1 at% B or Si addition.

tion results in a marked change of the DSC traces. The first peak is sensitive to the small amount addition of B and Si, and almost merges with the second one. The crystallization temperature increases by about 15 K. The addition of B and Si with larger negative enthalpy of mixing causes a change of the crystallization mode from a multistage process towards a single-stage-like transformation. We infer that B and Si addition may have the same role as the isothermal annealing on the microstructure and induce the disappearance of the first peak resulting from the microstructure transformation from SRO to MRO. Presumably, the B and Si react with metallic components and form nuclei, but the extremely slow atomic mobility and high activation energy²²⁾ for the precipitation of the crystalline phase(s) limit the growth of the nuclei and leads to a homogeneous distribution of 0.5 to 2 nm nuclei/MRO domains which can not be detected by XRD. This phenomenon has also been found in the ZrTiCuNiBe BMG upon annealing under high-pressure.²⁴⁾

3.1.1 Zr–Nb–Cu–Ni–Al alloy

Nb has a much high melting temperature ($T_m = 2741$ K) and is not so prone to reaction with oxygen compared with Ti ($T_m = 1941$ K). It is found that the GFA of the Zr–Cu–Ni–Al alloy is very sensitive to the content of oxygen.²⁵⁾ More than 0.48 at% oxygen will result in the formation of crystalline phases. Since Ti easily reacts with oxygen during the melting process, the addition of Ti in the Zr–Cu–Ni–Al system may increase the oxygen content in the alloy. It is expected that substitution of Nb for Ti may increase the thermal stability of the BMG and improve the GFA of the ZrCuNiAl glass forming system, because the Nb addition may lead to a lower oxygen content in the alloy. The experiments confirm that the $\text{Zr}_{57}\text{Nb}_5\text{Cu}_{15.5}\text{Ni}_{12.5}\text{Al}_{10}$ BMG is easier to be obtained compared to the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG under the same preparation conditions. Figure 4 shows XRD patterns of $\text{Zr}_{57}\text{Nb}_5\text{Cu}_{15.5}\text{Ni}_{12.5}\text{Al}_{10}$, $\text{Zr}_{60}\text{Nb}_3\text{Cu}_{14}\text{Ni}_{13}\text{Al}_{10}$, and $\text{Zr}_{55}\text{Nb}_9\text{Cu}_{15}\text{Ni}_{11}\text{Al}_{10}$ alloys with different composi-

Fig. 4 XRD patterns of $\text{Zr}_{57}\text{Nb}_5\text{Cu}_{15.5}\text{Ni}_{12.5}\text{Al}_{10}$, $\text{Zr}_{60}\text{Nb}_3\text{Cu}_{14}\text{Ni}_{13}\text{Al}_{10}$, and $\text{Zr}_{55}\text{Nb}_9\text{Cu}_{15}\text{Ni}_{11}\text{Al}_{10}$ alloys with different composition.

Fig. 5 DSC traces of the Zr–Nb–Cu–Ni–Al alloys with different Nb contents.

tion. An amorphous alloy can only be obtained for the $\text{Zr}_{57}\text{Nb}_5\text{Cu}_{15.5}\text{Ni}_{12.5}\text{Al}_{10}$. A small change in Nb composition results in a substantially changed GFA as shown in the figure. Less or more Nb results in the formation of metallic glass-based composites with crystalline particles. Figure 5 shows typical DSC traces of the Zr–Nb–Cu–Ni–Al alloys. The crystallization of the $\text{Zr}_{57}\text{Nb}_5\text{Cu}_{15.5}\text{Ni}_{12.5}\text{Al}_{10}$ BMG proceeds through one single DSC event, which is markedly different from the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$ BMG. Both the glass transition temperature and the crystallization temperature are increased compared with the values found for the $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$

Fig. 6 XRD patterns of the alloys with B and Si addition. 1 at%B or Si addition induces the formation of crystalline phases.

BMG. Hence, T_g , T_x , the SLR and the crystallization behavior are very sensitive to the Nb content. A decrease of the Nb content to 3 at% lowers T_g and T_x to 648 and 728 K, respectively, but the SLR, $\Delta T = T_x - T_g$, increases to 80 K. The partially amorphous $Zr_{60}Nb_3Cu_{14}Ni_{13}Al_{10}$ and $Zr_{55}Nb_9Cu_{15}Ni_{11}Al_{10}$ alloys also have a different crystallization behavior. Already small changes in the Nb content cause a change from the single-stage crystallization mode to a multistage reaction sequence. Figure 6 presents XRD patterns

of the alloys with B and Si addition. 1 at%B or Si addition induces the formation of crystalline phases. The as-cast alloy contains a mixture of amorphous phase and crystalline phase. The crystalline diffraction peaks are of large full width at half maximum indicating the fine grain size of the crystalline phases.

Table 1 presents the elastic data obtained from acoustic measurements. For comparison, the elastic data of other Zr-based BMGs are also listed in Table 1. The elastic constants calculated from the acoustic velocities are very close to that obtained by other methods.^{3,22)} The Debye temperature θ_D and the elastic constants of the $Zr_{57}Ti_5Cu_{20}Ni_8Al_{10}$ and the $Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10}$ BMGs are close to the data found for other Zr-based BMGs. These results indicate that the BMGs investigated in this study exhibit the same excellent properties as the other Zr-based BMGs already investigated. It should be noted that the elastic properties of the BMGs containing additional B and of the metallic glass composites do not significantly deviate from the data of the as-prepared $Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10}$ BMG.

Table 2 compares the relative changes of the transverse velocity, v_s , Debye temperature, θ_D , shear modulus G , density ρ , longitudinal velocity v_l , and bulk modulus K for the glassy and the crystallized states (obtained after annealing at 773 K in vacuum for 1 hour) of the Zr–(Ti, Nb)–Cu–Ni–Al BMGs. It should be note that the crystallized state might not be equilibrium or final crystallized state, because the crystallization kinetics of the BMGs is very slow, and the BMGs which have various states depending on the cooling rate are not relaxed state. The corresponding data for the $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ BMG are also listed for comparison.²⁷⁾ The $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ BMG shows large changes in ρ (1.1%), v_s (13.5%), θ_D (13.4%), and G (30.3%)

Table 1 The acoustic and elastic constants calculated from the ultrasonic data for $Zr_{57}Ti_5Cu_{20}Ni_8Al_{10}$, $Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10}$ and other Zr-based BMGs. *a* stands for the amorphous structure, *c* stands for the composite.

Sample	ρ (g/cm ³)	V_l (km/s)	V_s (km/s)	E (GPa)	G (GPa)	K (GPa)	σ	θ_D (K)	Structure	Ref.
$Zr_{57}Ti_5Cu_{20}Ni_8Al_{10}$	6.55	4.623	2.149	82.0	30.1	99.2	0.36	270.3	<i>a</i>	This work
$Zr_{60}Nb_3Cu_{14}Ni_{13}Al_{10}$	6.69	4.780	2.149	84.8	30.9	111.7	0.37	270.4	<i>c</i>	This work
$Zr_{55}Nb_9Cu_{15}Ni_{11}Al_{10}$	6.87	4.880	2.250	93.7	34.3	115.7	0.36	283.4	<i>c</i>	This work
$(Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10})B_1$	6.77	4.681	2.106	82.5	30.0	108.3	0.37	266.3	<i>a</i>	This work
$Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10}$	6.81	4.683	2.165	87.1	31.9	106.7	0.36	273.9	<i>a</i>	This work
$Zr_{48}Nb_8Cu_{14}Ni_{12}Be_{18}$	6.7	4.950	2.260	93.9	34.3	118.3	0.37	295.0	<i>a</i>	26
$Zr_{48}Nb_8Cu_{12}Fe_8Be_{24}$	6.43	4.933	2.740	95.7	35.2	113.4	0.36	306.0	<i>a</i>	26
$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$	6.125	5.174	2.472	101.2	37.4	114.1	0.35	326.8	<i>a</i>	27

Table 2 A comparison of the density and properties of the as-prepared amorphous state (density, ρ_0 , acoustic velocities, v_l^0 , v_s^0 , elastic constant G_0 , Debye temperature θ_{D0}) and crystallized state (density ρ , acoustic velocities, v_l , v_s , elastic constant G , Debye temperature, θ_D) of the $Zr_{57}Ti_5Cu_{20}Ni_8Al_{10}$, $Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10}$, and $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ BMGs. The elastic modulei, and the Debye temperature are calculated from acoustic data.

State	$\frac{\Delta\rho}{\rho_0}$ (%)	$\frac{\Delta v_l}{v_l^0}$ (%)	$\frac{\Delta v_s}{v_s^0}$ (%)	$\frac{\Delta G}{G_0}$ (%)	$\frac{\Delta K}{K_0}$ (%)	$\frac{\Delta\theta_D}{\theta_{D0}}$ (%)
$Zr_{57}Ti_5Cu_{20}Ni_8Al_{10}$	0.65	3.8	1.4	5.6	12.7	2.4
$Zr_{57}Nb_5Cu_{15.5}Ni_{12.5}Al_{10}$	0.60	1.0	0.5	3.1	3.6	1.2
$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$	1.1	5.2	13.5	30.3	3.9	13.4

and small changes in v_l (5.2%) and K (3.9%) between the amorphous and crystallized states, which indicates a softening of the transverse elastic modulus (*i.e.* the softening of the long wavelength transverse phonons) in the BMG relative to its crystallized state. A striking softening of transverse acoustic phonons relative to the crystalline state has been observed in various glasses including oxide glasses,²⁸⁾ PdNiP and PdNiCuP²⁹⁾ BMGs, conventional metallic glasses with poor GFA³⁰⁾ and amorphization of thin films induced by irradiation.³¹⁾ However, only small relative changes in the v_s (1.4%), v_l (3.8%), θ_D (2.4%), and G (5.6%) and K (3.9%) between the glassy and crystallized states for the Zr₅₇Ti₅Cu₂₀Ni₈Al₁₀ BMG are observed (shown in Table 2). The density is not obviously changed after crystallization ($\frac{\Delta\rho}{\rho_0} = 0.65\%$). This result is in good agreement with data from Inoue.²²⁾ A similar situation is found for the Zr₅₇Nb₅Cu_{15.5}Ni_{12.5}Al₁₀ BMG.

These results indicate no softening of the shear modulus in the Zr–(Ti, Nb)–Cu–Ni–Al BMGs relative to its crystallized state. It is known that the density and acoustic velocities are very sensitive to the microstructure. The softening phenomenon in the glasses is related to the change of their local atomic structure. Lam and Okamoto³¹⁾ pointed out that for the softening associated with static atomic displacement and anharmonic vibration in a solid, the effect of the static atomic displacement is nearly twice as large as that of anharmonic vibration. The static atomic displacement is a general measure of the chemical and topological disorder and, hence, can be used as a general disorder parameter for characterizing a glassy solid.³¹⁾ The increased magnitude of non-uniform static atomic displacements leads to lower frequencies of transverse phonons. The disorder of the structure mainly results in the softening of the transverse phonons in the glasses, the softening mainly being attributed to the unique microstructural characteristics of the BMG.²⁷⁾ The small changes of the density and acoustic velocities in the BMGs relative to their corresponding crystallized state indicates that a similar electronic state and similar atomic interactions between the glassy and crystalline states exist.

4. Conclusions

Bulk Zr–M–Cu–Ni–Al (M = Ti, Nb) metallic glasses are obtained by casting method. Their thermal stability and glass forming ability as well as the effects of the metalloid elements Si, B have been investigated. It is found that the GFA and crystallization behavior of the alloys are very sensitive to the addition of B and Si. Unlike other glasses, no softening of transverse phonons relative to the crystallized state has been observed in the Zr-based BMGs. The small changes of the density and acoustic velocities in the BMG relative to its corresponding crystallized state indicate a similar electronic

state and similar atomic interactions between the glassy and crystalline states.

Acknowledgments

The authors thank W. Gude for help with the sample preparation, and L. Schultz for stimulating discussions. W. H. Wang is grateful for the financial support from the National Natural Science Foundation of China.

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