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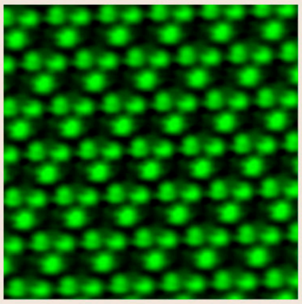

Full strength compacts by extrusion of glassy metal powder at the supercooled liquid state

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We report the production of full strength compacts of metallic glass by warm extrusion of powders at the supercooled liquid state just above the glass transition temperature. The alloy used was $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ (at. %) which has the lowest viscosity among Zr-based metallic glasses with large supercooled liquid region. The tensile strength and Young's modulus of the glassy powder compacts were 1520 MPa and 80 GPa, respectively, which are similar to that obtained in the as-cast bulk alloy and melt-spun ribbon. This opens up possibilities of producing high strength amorphous alloys with complex shapes. © *1995 American Institute of Physics.*

Production methods of bulk glassy alloys can be classified into two main processes. The first is the consolidation of metallic glass powders.[1–7] The second is the casting method such as metallic mold casting.[8–10] Although the latter method is simpler, the size of the obtained samples is, in general, limited to less than 20 mm in diameter and the alloy is restricted to the compositions with a high glass-forming ability. On the contrary, no such restriction exists for the powder consolidation which enables production of complex shapes. A number of studies have previously been carried out on the consolidation of metallic glass powders using various consolidation techniques such as warm extrusion,[1–3] static high pressure compaction,[4] explosive compaction,[5,6] and dynamic compaction.[7] The strength of these amorphous alloy compacts was comparable to bulk alloys only under condition of compressive loading.[2,11] The amorphous alloy compacts with the same tensile strength as the melt-spun ribbons or the as-cast bulk alloys have not been realized and constitute the major challenge for the development of these alloys.

Recently, a number of glassy alloys with a wide supercooled liquid region (above 60 K) and high glass-forming ability have been discovered in Zr-based alloys.[9,10,12] In this communication, we report the development of a consolidation process of glassy metal powders which yield identical mechanical properties as that of bulk alloys using a $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ (at. %) glass as a model material. $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ glassy powder was produced by a helium-gas atomization at a dynamic pressure of 9.8 MPa and a molten-alloy temperature of 1473 K using a guide tube with a hole diameter of 2.0 mm. The average particle size of the atomized powder was 35 μm. It was confirmed by x-ray diffractometry and differential scanning calorimetry (DSC) measurement that the powder with a particle size below 150 μm consists of only an amorphous single phase.

The $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ glass shows a minimum in viscosity of 1.8×10^9 Pa s at 696 K ($T_{v\,\mathrm{min}}$). The corresponding glass transition T_g and crystallization temperatures T_x are 652 and 735 K, respectively, measured at a continuous heating rate of 0.33 K s^{-1} with DSC. The time-temperature-transformation curve of the alloy is shown in Fig. 1. As can be seen, the duration τ of maintaining the amorphous phase without decomposition at 696 K is 1700 s. This provides the time window for the processing of the alloy at this temperature. Further, the maximum temperature T_d for maintaining enough ductility (bending by 180°) of the melt-spun alloy after a 60 s soaking time was found to be 728 K. This is taken as the higher temperature limit for the consolidation process to avoid embrittlement because the time of the extrusion is about 60 s.

The powder production and its consolidation were conducted in a closed powder metallurgy processing system which has been developed by the present authors.[2] In this system, the oxygen and moisture components can be maintained at less than 1 ppm level. In the consolidation process, billets for extrusion were produced in the following way. The $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ glassy powder with a particle size below 75 μm was cold pressed into a copper can with an inner diameter of 20 mm and an outer diameter of 23 mm, and then degassed at 573 K for 900 s. Subsequently, the billets were sealed, heated at the extrusion temperatures, and then

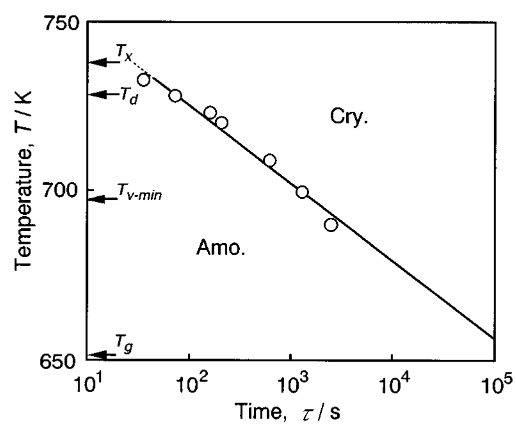

FIG. 1. Time-temperature-transformation curve for the onset of crystallization of $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ glassy alloy.

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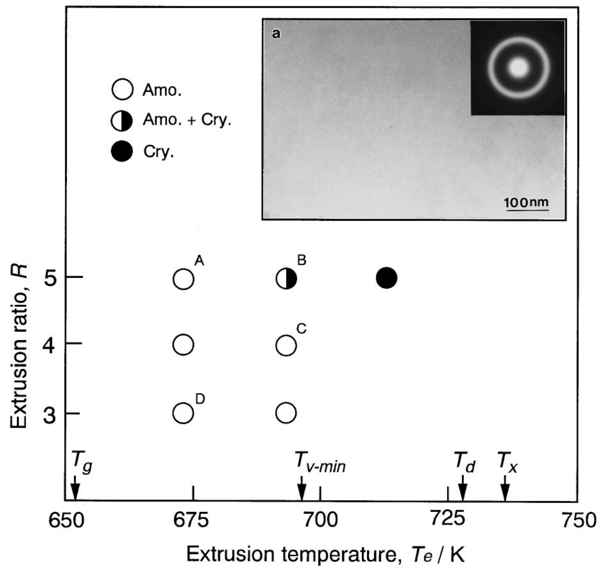

FIG. 2. Effects of extrusion temperature T_e and extrusion ratio R on the structure of $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ compacts. Bright-field electron micrographs and selected-area-diffraction patterns of $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ glassy compact A produced at T_e=673 K and R=5 are also shown in the inset.

extruded at a ram speed V_e of 1.0 mm s^{-1}. The extruded billets were immediately quenched in the water.

The rise ΔT_e in the temperature of the billet during extrusion, which is derived by work-induced heat generation, is given by[3]

$$\Delta T_e = 1.1 \times 10^4 V_e^{0.64} P_e/(\rho C_p). \tag{1}$$

Here, V_e, P_e, ρ, and C_p are ram speed, pressure generated during extrusion, density, and specific heat, respectively. The density ρ of the alloy is measured to be 6.70 g cm^{-3} and the C_p is estimated to be 30 J mol^{-1} K^{-1}. For a typical P_e of 1.0 GPa and V_e=1.0 mm s^{-1}, the temperature rise ΔT_e is estimated to be 55 K. Therefore, the optimum extrusion temperature from this analysis was around 673 K which was arrived at by subtracting ΔT_e from T_d. The density of compacts depends on *P/Y* (*P*: pressure, *Y*: flow stress of the alloy). The pressure P_e generated during extrusion can be expressed as[3]

$$P_e = Y(A \ln R + B), \tag{2}$$

where R is extrusion ratio, and A and B are factors depending on the die shape and friction. This equation implies that the density of the extruded samples is dominated mainly by the R. The extrusion ratio R above 2.5 is necessary for obtaining compacts with a density above 99%.[3] Accordingly, we attempted extruding with extrusion ratios R of 3, 4, and 5.

The results of the consolidations are shown in Fig. 2. The compacts with an amorphous single phase and a high density (more than 99%) were obtained at the extrusion conditions marked with the open symbols in Fig. 2. The full density glassy alloy compact could be obtained at fairly large domain of extrusion temperatures and ratios. The inset (a) shows the transmission electron microscopic evidence of the fully glassy state of the compact. This is processed at a temperature of 673 K and an extrusion ratio of 5. Transmission electron microscopy (TEM) investigation reveals that only at higher temperatures of 693 K and above and at an extrusion ratio of 5, we obtain precipitation of crystalline phase. Table I summarizes the results of four compacts marked in Fig. 2 including the processing conditions. Figure 3(a) shows the scanning electron micrographs (SEM) of the polished cross-sectional surfaces of the glassy compact A produced at R=5 and T_e=673 K. No cavity or pore can be observed in these compacts. The density of each compact was measured by the Archimedean method using tetrabromoethane. These are also summarized in Table I. In all cases nearly full density has been achieved.

The test pieces for the tensile test were machined by spark-type cutting method. The thickness, the width, and the gauge length of the test pieces were 1.5, 3.0, and 8.4 mm, respectively. The tensile test was conducted using an Instron testing machine at room temperature and a strain rate of 5×10^{-4} s^{-1}. For comparison, the $Zn_{65}Al_{10}Ni_{10}Cu_{15}$ glassy bulk alloy with a transverse cross section of 1.5×5.0 mm^2 produced by injection casting of the melt into copper mold was also tested using a test piece with the same size as the compacts. Table I includes the summary of the results from the four compacts together with the results of as-cast and melt-spun alloys. The tensile strength and the Young's modulus of the glassy compact A are 1520 MPa and 80 GPa, respectively. These tensile properties are similar to those of the as-cast glassy bulk alloys and the melt-spun glassy ribbons. The glassy compacts C and D have a tensile strength of 1420 MPa. A slight decrease in the strength is related to smaller extrusion ratio which results in the poor particle bonding. On the other hand, the partially crystallized compact B has a much smaller strength of 930 MPa and a higher Young's modulus of 117 GPa. Figure 3(b) shows the tensile fracture surfaces of the glassy compact A with the highest strength of 1520 MPa. The tensile fracture of this compact

TABLE I. Tensile properties and density of $Zr_{65}Al_{10}Ni_{10}Cu_{15}$ compacts in comparison with as-cast glassy bulk alloy and melt-spun glassy ribbon.

Specimen	Extrusion condition R	T_e (K)	Structure	Tensile strength σ_f (MPa)	Young' modulus E (GPa)	Relative density D (%)
Compact A	5	673	Amorphous	1520	80	99.4
Compact B	5	693	Amorphous+crystalline	930	117	99.9
Compact C	4	693	Amorphous	1420	78	99.5
Compact D	3	673	Amorphous	1420	78	99.1
As-cast sheet			Amorphous	1570	80	99.9
Melt-spun ribbon			Amorphous	1440	⋯	100

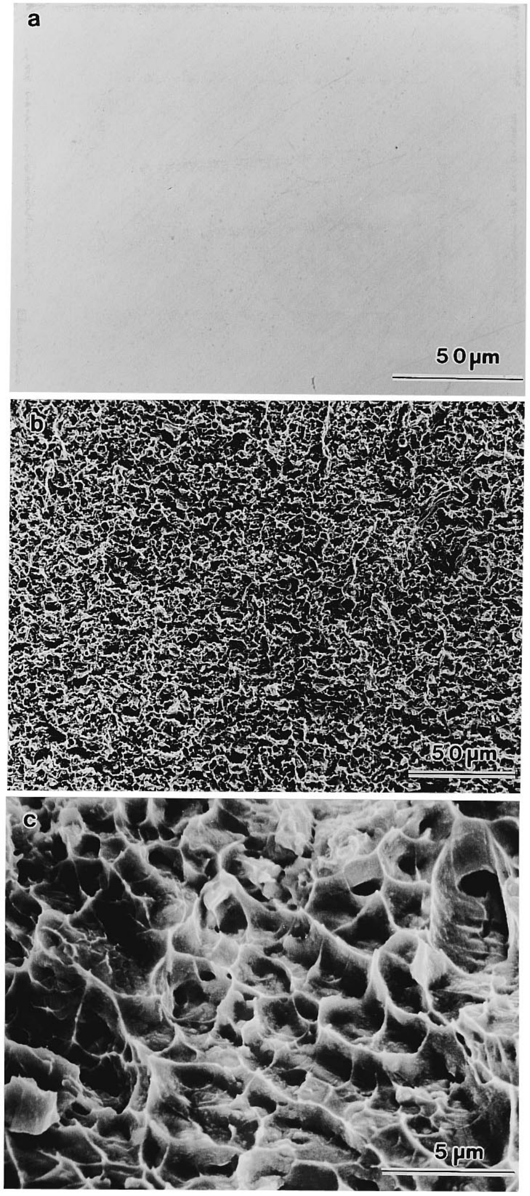

FIG. 3. Scanning electron micrographs of the polished cross-sectional surface (a) and the tensile fracture surfaces (b) and (c) for the $Zn_{65}Al_{10}Ni_{10}Cu_{15}$ glassy compact A produced at T_e=673 K and R=5.

took place along the maximum shear plane which is inclined to the tensile direction by 45°. No fracture occurs along the prior powder particle boundary. This indicates the achievement of strong bonding between the particles. Vein patterns, which is a characteristic fracture pattern for amorphous alloys with good bending ductility, are also observed over the whole fracture surface as shown in a magnified SEM micrograph [Fig. 3(c)]. The glassy compacts seems to maintain good ductility without embrittlement.

To summarize, we have established by this study that the glassy metal powder compacts with almost the same tensile strength as the melt-spun ribbons or the as-cast bulk alloys can be produced. The high tensile strength was derived from the extrusion at supercooled liquid state through a closed powder metallurgy processing system. The system prevents the surface oxidation of the powder particles resulting in strong particle bonding. On the other hand, the flow stress and the work-induced heat generation are small on extruding at the supercooled liquid state. The supercooled liquid state enables extrusion at a larger extrusion ratio and the combined effect leads to strong particle bonding.

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