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SIGNIFICANT TENSILE PLASTICITY OF COLD ROLLED $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BULK GLASSY ALLOYS

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Abstract. In order to obtain ductile bulk glassy alloys (BGAs) with macroscopic tensile elongation, cold rolling was examined using $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs. Cold rolled structure is characterized by its unique fine-layered structure, which is composed of deformed band regions and non-deformed block regions. Since the deformed shear band region can be deformed lower shear stress than that of non-deformed region, cold rolled bulk glassy alloy can deform homogeneously. On the other hand, cold rolling ability is also important factor to fabricate the cold rolled glassy alloys with sufficient reproducibility. Density, which strongly influences on the mechanical properties of cast BGAs, is used to estimate the cold rolling ability. As the result, we can achieve the cold rolled ductile $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA exhibiting significant tensile elongation.

1. INTRODUCTION

Glassy alloys have two distinct features that result from their aperiodic glassy structure and flexible metallic bonding. One special trait of glassy alloys is their mechanical property. Glassy alloys in metal-metal type system usually exhibit ductile metallic bonding nature. Ductile crystalline alloys can exhibit superior plasticity due to systematic dislocation movement. The deformation mechanism of glassy alloys is attributed to a unique adiabatic shear-band movement [1,2]. Microscopic fracture-surface images reveal a vein pattern caused by low viscosity [3] in the shear band. Therefore, once a shear band begins moving, it results in a final fracture with minimal uniform plastic deformation. The lack of uniform plastic deformability of glassy alloys has been considered to limit the toughness [4].

Since 1998, special multicomponent alloys have recently been found to exhibit high glass-forming ability [5]. Bulk glassy alloys have new potential applications as structural material [6,7]. Even in such an advanced state, one must pay attention to cast defects [8-10] in these structural BGAs. Crystalline inclusions, which act as crack-initiation sites and enhance crack propagation, must be reduced to examine the intrinsic features of BGAs [11]. While an as-cast sample exhibits a single glassy phase, the glass structure depends on composition and cooling-rate. Therefore, the control of glass structure is important to obtain consistent quality [12]. In this study, we measure the density to evaluate the quality of glassy alloys.

Ductile glassy alloys, the features of which are derived from significant elastic strain-energy storage, require sufficient elastic deformability to relax the localized stress immediately. A substantial elastic strain, up to 2% of a glassy alloy, may be associated with sufficient stress-relaxation ability [13]. Therefore, the ductility of glassy alloys originates from the high stress-relaxation ability. However, the improvement of plasticity and ductility of BGAs is

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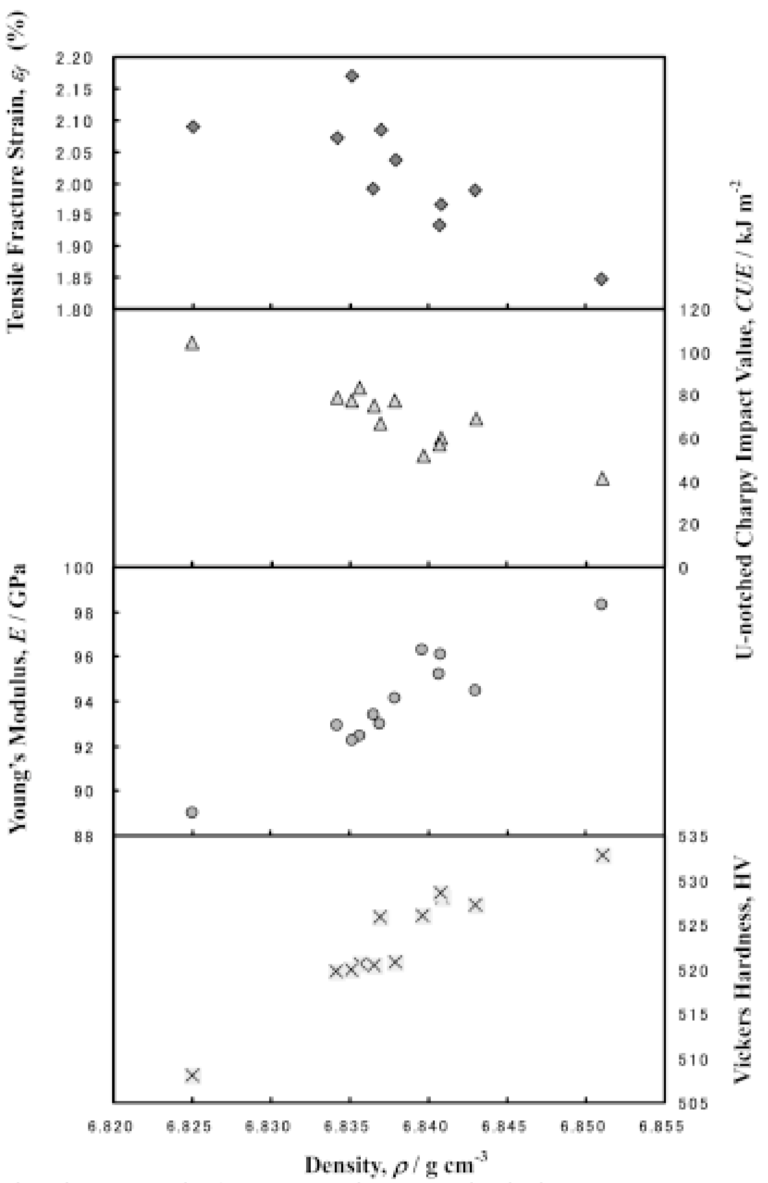

Fig. 1. The effect of density on tensile fracture strain, U-notched Charpy Impact Value, Young's modulus, and Vickers hardness of cast $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs.

difficult problem, because of having a apparent work softening phenomena due to viscoelastisity phenomena in shear band [14] even at the room temperature. Though, the initiation and propagation of shear bands are important factor to decide the mechanical feature of BMGs at room temperature. In order to improve the plasticity of BGAs, pre-introduced shear band structure is effective on the viewpoint of localized stress relaxation. Cold rolled structure is characterized by its unique fine-layered structure, which is composed of deformed shear band regions and non-deformed block regions [15]. Since the deformed shear band region can be deformed lower shear stress than that of non-deformed region, cold rolled bulk glassy alloy can deform homogeneously [16]. On the other hand, some glassy alloy cannot be cold rolled with insufficient ductility. Estimation of cold rolling ability is also important factor to fabricate the fine shear band structure by cold rolling. Fine cold rolled structure promotes the significant improvement of plasticity [17] of bulk glassy alloys.

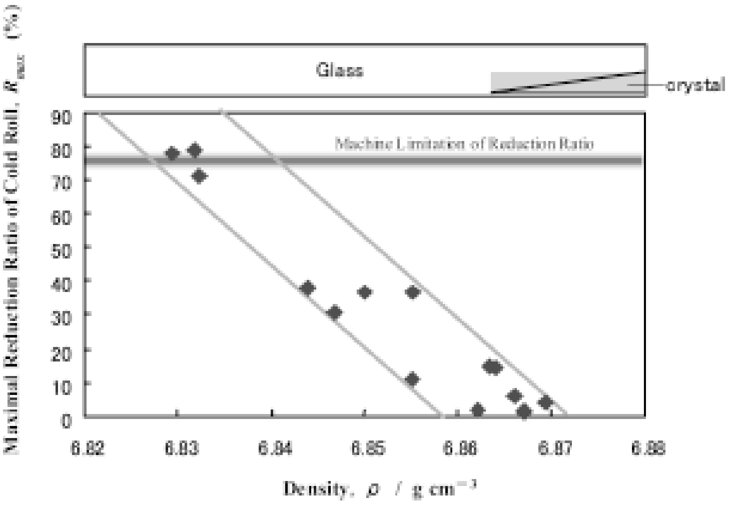

Fig. 2. Relationship between the density and maximal reduction ratio of cold roll. Upper part means the ratio of crystallized region. The maximum reduction ratio of the cold roll machine is about 77%.

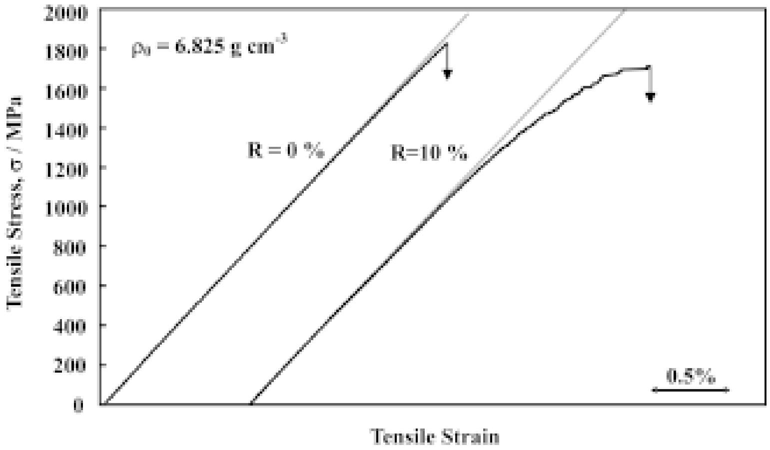

Fig. 3. Tensile stress and strain curve of non-rolled and 10% cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs.

The objective of this paper is estimation of cold rolling ability by using the density, and we also fabricated ductile cold-rolled bulk glassy alloy with distinct tensile elongation at room temperature.

2. EXPERIMENTAL PROCEDURE

Master-alloy ingots of quaternary $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ alloys were prepared by arc-melting mixtures of pure Zr, Cu, Ni, and Al in an argon atmosphere. We used a special Zr crystal rod with an oxygen concentration of less than 0.05 atomic percent (at.%) to maintain a low oxygen concentration in the alloys. $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs were cast into rod-shaped (∅8 x 60 mm) for mechanical test and plate-shaped (3 x 20 x 60 mm) for cold rolling with tilt-casting technique [11]. This casting method has the advantage of restricting the formation of cold shuts, which act as crack-initiation sites and also enhance crack propagation. Densities were measured by the Archimedes method using two shaped samples (∅8 x 20 mm) for mechanical test and (3 x 10 x 20 mm) for cold rolling. The measurement limit of an electronic balance is 0.1 mg, and the fluid for Archimedes method was purified water. We also verified the cast structure using optical microscopy (OM) and scanning-electron microscopy (SEM) to confirm the single glassy phase. The phase characterization and occupation rate of crystallized region were examined by X-ray diffractometry. Tensile specimens are prepared plate shape (2 mm thick) with 5 mm in gage width and 10 mm in gage length. Cold-rolling process is

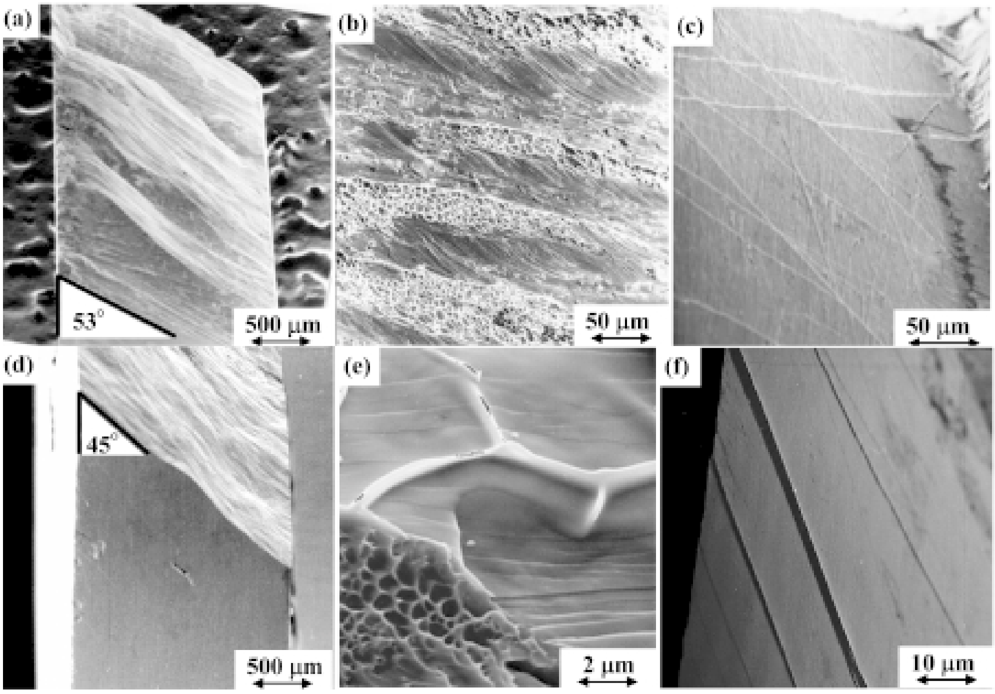

Fig. 4. SEM images of tensile fractured surface (a), its magnified image (b), and side view (c) of non-rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs. And SEM images of tensile fractured surface (d), its magnified image (e), and side view (f) of 10% cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs.

done using a 1 MN class four-roll-high rolling machine with a work roll diameter of 100 mm, and rolling reduction for one pass is controlled less than 0.5% to avoid fracture during cold rolling. Tensile tests were performed using an Instron 5582 testing machine to measure the Young's modulus. A Charpy impact test was also performed in air using a size-reduced sample that was 55 mm long, 10 mm wide, and 5 mm thick, U-notched to a 2 mm depth.

3. RESULTS AND DISCUSSION

Glass is solidified at a glass-transition temperature; however, the free volume in a glassy alloy may change with the cooling rate. The quantity of free volume may also be changed by the alloy composition. It is difficult to determine the absolute value of the free volume, and therefore we measured the density of cast BGAs to estimate the glass structure change in the same composition. The density change of cast BGAs is mainly caused by the difference in quenched in free volume. Fig. 1 shows the changes of tensile fracture strain, U-notched Charpy impact value (CUE), Young's modulus and Vickers hardness with a function of the density of as cast $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs with several casting conditions. The relationship between the density and tensile fracture shows negative proportionality relation, meaning the embrittlement during the density increasing. The relationship between the density and CUE shows also negative proportionality relation with a large declination, meaning the significant embrittlement for the impact fracture. On the other hand, the relationship between the density and Young's modulus shows positive proportionality relation, and tensile fracture strain decreases with density. Since high correlation with density is probably originate to glass structural change, the property can be regarded as an intrinsic property. We conclude that the tensile strength is not an intrinsic property in bulk glassy alloy because it has no correlation with density. Hardness also increases linearly with the density increasing. Consequently, the density increasing due to structural relaxation is not simple volume shrinkage but typical atomic bond state change to be rigidity of glass structure, that phenomenon is not usually seen in common crystalline alloys. Therefore, in order to obtain ductile glassy alloy, we have to control the density of cast bulk glassy alloy. Low density of glass structure is mainly originated to the quenched in open volume from liquid state called as free volume.

In order to obtain the fine shear band lamellar structure of cold rolled BGAs, cold rolling ability is an important factor, which is probably enhanced by the degradation of density value. Fig. 2 shows the relationship between the density and maximal reduction ratio before failure of cold roll of cast $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs. Cold rolling ability increases significantly with a decrescence of density. Since the relation also takes positive proportionality relation, we can estimate the cold rolling ability of the cast $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs by the value of density. In this study, we determined the standard density of a $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA to be 6.825±0.005 g cm^{-3}, which value can be realize under sufficient cooling rate of tilt cast rod sample (∅8 x 60 mm). As the result, we could fabricate plastic cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs with fine lamellar shear band structure by controlling the density of cast alloy before cold rolling.

By making full use of pre-introduced shear band function; plasticity is significantly improved to realize the tensile elongation under the tensile testing at room temperature. Fig. 3 shows the tensile stress and strain curves of non-rolled and 10% cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs. We can observe significant tensile plastic elongation in cold rolled stress-strain curve as about 0.5%. On the contrary in non-rolled sample, we can observe the tensile plastic elongation about 0.03%. Tensile fractured surfaces of both samples were also examined to clarify the origin of difference in tensile elongations. Fig. 4 shows SEM images of tensile fractured non-rolled (a-c) and 10% cold rolled (d-f) $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs. Fracture surface is formed by one shear band plane with tilt angle about 53 degree from the tensile direction meaning plane strain condition for non-rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA as shown in Fig. 4a. The cold rolled sample fractured rather smooth shear band plane, which was introduced by cold rolling, therefore the tilt angle about 45 degree from tensile direction. Tensile fractured surface of non-rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA shows periodic band structure with rub pattern and vein pattern, whose structure is originated to the typical wavy-curved shear band morphology. The wavy shear band, whose movement is restricted by the barrier to rub convex regions, is usually seen [18] in ductile BGAs with low density. Such a barrier for shear band movement may cause a little tensile elongation in non-rolled BGAs as shown in Fig. 3. On the other hand, well-grown vein patterns are also seen in all over tensile fractured surface of cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA. However, the fine shear band steps, which are caused by reoperation of pre-introduced shear band during the tensile testing, characterize the cold rolled tensile fractured surface as shown in Fig. 4e. Accordingly, coordinated movement of pre-introduced shear bands brings about the apparent plastic deformation during the tensile test. Therefore, the side view of cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA shows a lot of parallel shear band marks with visible steps as shown in Fig. 4f. On the other hand, the side view of non-rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGA exhibits crossed shear bands with a little step width.

4. SUMARRY

We attempted to control the density and cold rolled structure of $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs to achieve the tensile elongation. The results obtained can be summarized as follows.

(1) The density $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs has linear relationship with cold rolling ability, tensile fracture strain, Charpy impact value for positive proportion and with Young’s modulus and hardness for negative proportion, respectively.

(2) By controlling the density of cast $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs, we could control the cold rolling process. Cold rolled $Zr_{50}Cu_{30}Ni_{10}Al_{10}$ BGAs exhibit significant tensile elongation as about 0.5%.

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