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Plasticity improvement of a bulk amorphous alloy based on its viscoelastic nature

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This study examines the fundamental issue of whether an elastic shear stress imposed on amorphous alloys causes structural changes during quasistatic loading. In order to deliver elastic shear stress to the workpieces, a quasistatic shear deforming device was designed. With repeated application of elastic shear stress, the plasticity of the shear-deformed amorphous alloys increased while the yield strength decreased. These behaviors were analyzed from the perspective of the structural changes associated with the viscoelastic nature of amorphous alloys.

Keywords: Bulk amorphous alloy; Plasticity; Free volume; ECAP

Recently, there have been a number of attempts to identify the origin of the plasticity of amorphous alloys by defining the free volume as a structural defect within them [1–3]. Argon and Kuo [4] suggested that the shear load promotes the generation of excess free volume in the local shear zone, resulting in an increase in the plasticity [5]. Egami et al. [6] observed that annealing reduces the excess free volume in amorphous alloys, causing their embrittlement. A common feature of these earlier results is that the plasticity of amorphous alloys is intimately related to the free volume fraction.

The fact that excess free volume is formed by the application of shear stress [1] implicitly suggests that mechanical working could be used as a technique for enhancing the plasticity of amorphous alloys. Indeed, a substantial improvement in the plasticity of amorphous alloys was achieved by cold rolling [7]. In this process, excess free volume, along with numerous shear bands, is readily created at the sub-surface regions, on which a simple shear stress associated with the roll contact is imposed. However, rolling is unable to deliver uniform shear stress along the thickness direction of the sample, while it inevitably reduces its thickness. Equal channel angular pressing (ECAP) is an alternative technique that can readily create excess free volume, thus improving the plasticity. This is because ECAP not only enables a relatively uniform simple shear stress to be delivered to the samples, but also allows multi-passage operation without varying the sample thickness. Therefore, it is thought that the amount of excess free volume can be controlled via ECAP by changing either the number of passages or the die angle (i.e. magnitude of the shear stress).

When the shear stress applied during ECAP exceeds the yield strength of an amorphous alloy, it sometimes causes excessive plastic deformation, resulting in the premature failure of the workpiece. Therefore, ECAP has to be performed in the elastic regime, so that it can deliver elastic shear stress to the workpiece. When ECAP is conducted in the elastic regime of an amorphous alloy, the questions arise as to whether (i) excess free volume is generated even at a stress level below the yield strength and (ii) the free volume that is created is maintained even after the removal of the applied stress.

Several pioneering works have suggested that elastic stress causes irreversible structural changes in amorphous alloys. Based on a simple theoretical model of an amorphous alloy, Argon [2] and Falk and Langer [3] postulated that structural changes can be induced by the application of shear stress. Wakeda et al. [8] and Ogata et al. [9] used definitive three-dimensional molecular dynamics simulations to demonstrate that structural changes may occur even in the elastic regime and are accompanied by the formation of excess free

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volume. In addition, Taub and Spaepen [10], from the elastostatic tensile tests of amorphous wires at elevated temperatures, observed an unrecoverable viscoelastic strain, suggesting that the characteristics of the structural changes associated with elastostatic loading are irreversible. These earlier results indicate that elastic stress causes irreversible structural changes that create excess free volume, and this provides the motivation for the present study.

This study reports on the concept of utilizing a shear deforming technique to enhance the plasticity of bulk amorphous alloys without reducing the sample dimension. A method to design an ECAP die, which can deliver elastic shear stress to generate excess free volume, is proposed. The variations in the mechanical properties and free volume fractions were measured as a function of the number of ECAP passages. On the basis of these results, we demonstrate experimentally that elastic shear stress can generate excess free volume in amorphous alloys, which in turn affects their mechanical properties.

A monolithic amorphous alloy with a composition of $Zr_{57}Cu_{20}Al_{10}Ni_8Ti_5$ was chosen as the sample alloy, since it can be cast into bulk form with minimal porosity. The alloy was cast into a copper mold to prepare platelet samples with dimensions of 6 mm (width) × 20 mm (length) × 1.5 mm (thickness). The geometry of the ECAP die was designed using the finite element (FE) method, so that it could maximize the creation rate of free volume while preventing the plastic deformation of the alloy. The properties of the sample used for the FE analysis were determined by the uniaxial compression test. Two-dimensional calculations based on the Mohr–Column criteria were made on four-noded plane stress elements. The ECAP die was assumed to be rigid and frictionless. ECAP was carried out at room temperature on workpieces with dimensions of 4 mm × 10 mm × 1.5 mm with a feeding speed of 0.06 mm min^{-1}. The ECAPed samples were then machined to rectangular rods with dimensions of 1 mm × 1 mm × 2 mm, and subjected to uniaxial compression tests at a constant strain rate of $\sim 10^{-4}$ s^{-1}. The increment of the relative free volume associated with the number of ECAP passages was determined by differential scanning calorimetry [11] (DSC; Perkin-Elmer DSC7, USA) at a heating rate of 40 K min^{-1}. The activation energies for overall crystallization were calculated using the non-isothermal Kissinger analysis [12] of the thermograms obtained at different heating rates of 10, 15, 20, 25 and 30 K min^{-1}.

The samples used in this study were fully amorphous and exhibited a yield strength and plastic strain of 1.78 GPa and 1.2%, respectively, indicating that they would be expected to yield at a shear stress of ∼890 MPa. An ECAP die was designed to deliver a shear stress slightly smaller than ∼890 MPa to the samples, in order to maximize the generation of excess free volume within them, while preventing them from becoming fractured.

Figure 1(a) shows an example of the FE calculations, showing how the shear stress is distributed along the thickness direction of the sample during the ECAP operation through the die with an internal angle of Φ. A series of simulations similar to that shown in Figure 1(a) were conducted by varying the die angle, in order to determine the appropriate die angle for imparting a

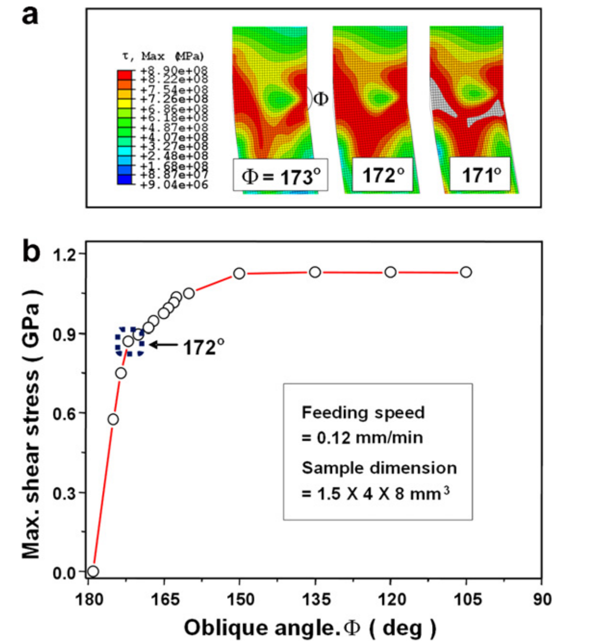

Figure 1. (a) Examples of finite element calculation for a die angle Φ. (b) Variations in the maximum shear stress imposed on the samples as a function of the die angle Φ.

shear stress of ∼890 MPa to the samples. Figure 1(b) shows the variations in the calculated maximum shear stress as a function of the die angle, suggesting that a shear stress slightly smaller than ∼890 MPa is obtained at a die angle of ∼172°. As such, the use of an ECAP die with $\Phi = 172°$ would maximize the generation rate of excess free volume without inducing plastic deformation.

ECAP was applied repeatedly to the samples in the same direction using a solid die consisting of two rectangular cross-sectioned channels with dimensions of 4 mm (w) × 1.5 mm (t) and an internal angle of $\Phi = 172°$. The ECAPed samples were then observed using scanning electron microscopy in order to confirm whether or not they were devoid of shear bands. No visible evidence for the formation of shear bands in the amorphous alloy was observed in the ECAPed samples, even after multiple passages. Figure 2 shows the compressive flow

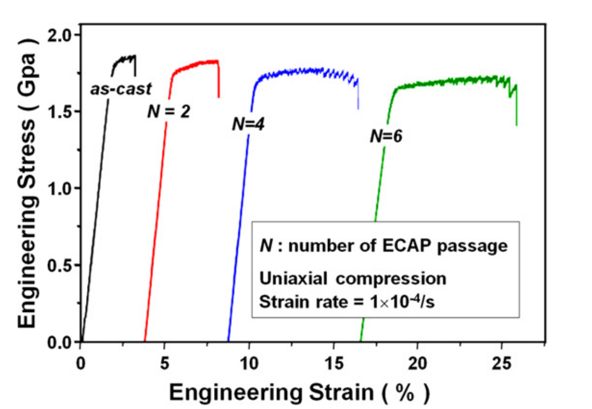

Figure 2. (a) Representative stress–strain curves obtained from theas-cast and ECAPed samples with various number of passages of ECAP.