Free Volume in a cold-worked Zr₅₅Cu₃₀Ni₅Al₁₀ Bulk Metallic Glass

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1 Introduction

It was reported that the volume fluctuation called the free volume is introduced into a plastically deformed amorphous alloy and metallic glass [1, 2]. However, the change in the glass structure of a deformed metallic glass has been insufficiently studied for recent years. The discovery of bulk metallic glasses (BMGs) gives us much opportunity to elucidate the structural properties in a deformed metallic glass. The high intensity x-ray of PF enables us to obtain new information of the details of plastically deformed BMGs.

2 Experiment

The Zr₅₅Cu₃₀Ni₅Al₁₀ BMG was prepared by the tilted cupper mold-casting technique [3] with a size of 14 mm in diameter and 30 mm in length. Two disks with a thickness of about 2 mm were cut from the rod shape BMG. One piece was thinned down into a fraction of about 90% by cold working at room temperature and another was regarded as the standard sample. The structure of both samples was investigated by a normal scattering technique with x-ray energy of selected 15.000 keV at BL7C station. The scattering experiment was performed with a vertical type two-axis goniometer and a SSD (Ge) detector. Then the scattering angle (2θ) was changed from 4 to 140 degrees. The coherent intensity of scattered photons was estimated from the diffracted beam and the structure factor, S(Q), was derived, where Q is the magnitude of the scattering vector.

3 Results and Discussion



Fig. 1: Qi(Q)'s of as-cast, deformed BMGs and the difference of S(Q).



Fig. 2: G(r)'s of as-cast, deformed BMGs and the difference of G(r).

The reduced radial distribution function, G(r), was calculated by Fourier transformation of S(Q). Figure 1 exhibits, Qi(Q)=Q(S(Q)-1), of standard as-cast and 10% cold-worked BMGs and the difference of S(Q), $\Delta S(Q)$ $=S_{cw}(Q)-S_{as}(Q).$ The change in S(Q) representatively appears around its first peak, i.e. a peak shift to lower side and a small broadening of peak. The corresponding G(r)'s are shown in Fig. 2 together with the difference, $\Delta G(r) = G_{cw}(r) - G_{as}(r)$. The overall behavior in $\Delta G(r)$ is definitely in anti-phase to both $G_{as}(r)$ and $G_{cw}(r)$, meaning that the amplitude of $G_{cw}(r)$ became smaller than the $G_{as}(r)$. The result suggests that the topological disorder of the as-cast BMG increased by cold working. The change in the glass structure by the structural relaxation was quantitatively investigated by the atomic level stress model [4]. Following this model, the $\Delta G(r)$ for structural relaxation was expressed as,

$$\Delta G(r) \approx -\frac{1}{2} \gamma^2 \partial^2 G_0 / \partial r^2 \left(\left\langle p^2 \right\rangle_{as} - \left\langle p^2 \right\rangle_{relax} \right),$$

where $\langle p^2 \rangle$ is the fluctuation of the atomic level hydrostatic pressure, and γ^2 is the *r*-dependent parameter approximately equivalent to a constant over first peak of G(r). The GO(r) is the reduced radial distribution function from the region with almost 0 pressure, and it is actually approximated by the G(r) in as-cast state. In case of structural relaxation, the $\Delta G(r)$ changed in phase to a minus second derivative of $G_{as}(r)$, $-\partial^2 G_0 / \partial r^2$, and consequently a positive change in $\langle p^2 \rangle_{as} - \langle p^2 \rangle_{relax}$ was obtained. The result was interpreted as the decrease of topological disorder, i.e. the fluctuation of atomic level stress. Figure 3 plots $-\partial^2 G_0 / \partial r^2$ against the distance, r,



Fig. 3: $-\partial^2 G_0 / \partial r^2$ of as-cast G(r)

where the curve was smoothed by cubic spline functions after the subtractive calculation. We can easily see an anti-phase behavior of $-\partial^2 G_0/\partial r^2$ against $\Delta G(r)$, meaning that $\langle p^2 \rangle_{as} - \langle p^2 \rangle_{cw}$ becomes a negative value, and it is an opposite sign to the structural relaxation. Thus we conclude that the volume fluctuation, which is characterized by the atomic-level stress, increases after plastically deforming BMG. The result consists with the volume and enthalpy change of cold-worked Zr₅₅Cu₃₀Ni₅Al₁₀ BMG [5].

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