The Effects of High Magnetic Field Annealing on the Structural Relaxation of Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_6$B$_{23}$ Bulk Metallic Glass

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Fe-based magnetic metallic amorphous and nanocomposites have excellent soft magnetic properties including greater magnetization and magnetic permeability compared with crystalline alloys, especially at high operation frequency and temperature. The high magnetic field (HMF) up to 12T is introduced to the heat annealing of Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_6$B$_{23}$ bulk metallic glass (BMG). The 12T HMF annealing shows the effect of improve the thermal stability of amorphous state during annealing in the BMG’s supercooled liquid region as compared with the annealing without magnetic field. The HMF annealing inhibits the brittleness of BMG during the structural relaxation, due to the increment of the activation energy under the HMF. The HMF annealing also results in squared hysteresis loops after the structural relaxation and lower coercivity.

Keywords: metallic glasses, soft magnetic property, high magnetic field, structural relaxation

1. Introduction

In the past decades, Fe-based bulk metallic glasses (BMGs) with a high glass forming ability (GFA) have become a very hot research topic not only because of their soft-magnetic properties but also the high fracture strength and corrosion resistance. In particular, these BMGs can be obtained directly either from the liquid or the deformation within the supercooled liquid region in the final shape suitable for various applications in different devices, such as magnetic sensors, magnetic valves, and magnetic clutches etc. Soft magnetic materials with low core losses, high magnetization, and low cost are the key components for improving the intrinsic and extrinsic soft magnetic properties have focused on tailoring the composition, controlling the microstructure with varied heat treatment environments. The low temperature annealing of metallic glass causes changes in most physics properties. This is attributed to atomic rearrangement in the amorphous state. Very recently, high magnetic field (HMF) up to above 10T has been successfully applied to materials design and productions. The finding has demonstrated that the magnetic field is a powerful tool to affect the crystallization process of metallic glass and texture formation of the crystallized phases. The high magnetic field (HMF) up to 12T has been successfully applied to materials design and productions. The finding has demonstrated that the magnetic field is a powerful tool to affect the crystallization process of metallic glass and texture formation of the crystallized phases. In this work, a HMF up to 12T is introduced to the low temperature structural relaxation process of high-boron Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_6$B$_{23}$ BMG. The results show that HMF can improve the thermal stability of amorphous state during annealing in the BMG’s supercooled liquid region as compared with the annealing without magnetic field. The HMF annealing inhibits the brittleness of BMG during the structural relaxation, due to the increment of the activation energy under the HMF. The HMF annealing also results in lower coercivity and squared hysteresis loops of the metallic glass after structural relaxation.

2. Experimental

Elemental pieces with a purity better than 99.9 wt.% were used as starting materials. The master alloy ingots of Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_6$B$_{23}$ with the nominal composition (in at.%) were prepared by arc melting under a Ti-gettered argon atmosphere. Bulk rods of 1.0 mm in diameter were fabricated using suction casting in a copper mold. The amorphous nature as well as homogeneity of the rod was ascertained using X-ray diffraction. The differential scanning calorimeter (DSC) measurements were performed under a purified Ar atmosphere in a TA Q100 at a heating rate of 2, 5, 10, 20 and 40 °C/min for both the as-cast and annealed samples. The morphology of the annealed rod was observed by using a scanning electron microscope (SEM). The magnetic field annealing was performed in a vacuum furnace, where a superconducting magnet (JMTD-12T100, JASTEC, Japan) was used to generate a magnetic field with a maximum magnetic flux density up to 12T at the center of a bore (100 mm in diameter). The 20-mm-length isothermal region of the furnace has an accuracy of around ±3 °C. The stable homogeneous magnetic fields were used in this work. The axis of the rod was parallel to the direction of the magnetic field. As the magnetic field was applied, the samples were heated to a given temperature at a heating rate of 5 °C/min, kept for 60 minutes, and then cooled down to the temperature below 100 °C in the furnace. The average cooling rate is...
around 6 °C/min. Vickers microhardness was measured in a MVR-HS hardness tester (Kawasaki, Japan) using a load of 300 N and 20 s hold time. The microhardness value given is the average of 20 individual measurements. Vibrating sample magnetometer (VSM, Lakeshore 7407) was used to measure the magnetic properties of the samples.

3. Results and Discussions

Figure 1 shows the DSC curves of the as-cast and annealed rods for Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_{6}$B$_{23}$ BMG. Characteristic temperatures associated with the glass transition temperature ($T_g$) and the onset temperature of primary crystallization ($T_{x1}$) are labeled in Figure 1. To simulate the heating rate for the magnetic field annealing, a heating rate of 5 °C/min was used for the as-cast rod in the work. At the heating rate of 5 °C/min, the $T_g$ and $T_{x1}$ of the as-cast rod are revealed to be 555 °C and 600 °C, respectively. Therefore, the structural relaxation annealing temperatures of this BMG are selected to be 500 °C and 550 °C, respectively.

The DSC curves of the samples both annealed at 500 °C for 60 min with or without a 12 T magnetic field are shown in Figure 1, which exhibit the significant endothermic characteristics of a glass transition followed by an exothermic peak almost with the same exothermic heat amount of the as-cast rod indicating the normally full amorphous structure after the low temperature annealing. From the XRD curves, no peak correspond to the crystalline phase is visible, which are not shown here. In contrast, there is no obvious $T_g$ observed in the DSC curve of the sample annealed at 550 °C without the magnetic field and a following smaller exothermic heat amount which indicates a partial crystallization already occurred in the precursor sample. The endothermic enthalpy of sample annealed at 550 °C with 12T high magnetic field is higher than that without magnetic field, which showed that the HMF improved the thermal stability of amorphous state of Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_{6}$B$_{23}$ BMG during the structural relaxation. Due to the partial crystallization when annealed at 550 °C, the structural relaxation discussed in this work focused on the rod annealed at 500 °C only.

The continuous DSC traces of the as-cast and annealed rods at 500 °C with heating rate of 2, 5, 10, 20 and 40 °C/min are done which are not shown here. The $T_g$, $T_{x1}$, and $T_p$ (peak temperature of primary crystallization) of the rods are shifted to a higher temperature by increasing heating rate. The Kissinger plots for the primary crystallization reactions for the three rods are shown in Figure 2. The activation energy $E_a$ and $E_p$ are deduced from the slope of -ln(R/T$^2$) versus 1/T, where T stands for the $T_g$, $T_{x1}$, and $T_p$ for the heating rate. The $E_a$ and $E_p$ for the as-cast rod are 508.2 and 528.5 kJ/mol, respectively. However, the $E_a$ and $E_p$ increased dramatically after annealed at 500 °C with 12T high magnetic field, which are 586.8 and 541.4 kJ/mol, respectively, and the $E_p$ is lower than $E_a$, which is different from as-cast rod and annealed without magnetic field.

From the DSC curves in Figure 1, the improved thermal stability of Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_{6}$B$_{23}$ BMG during HMF annealing assumed ascribing to the increased $E_a$.

The indentation of the BMG rods after structural relaxation is also investigated. In comparison, the indentation results of BMG rods annealed at higher temperature are also presented in Figure 3a. It is clearly seen that the hardness is higher in the rod annealed without magnetic field compared that with 12T HMF. For the rods (A and B as shown in Figure 3a after structural relaxation, the indentation images are also show in Figure 3b and 3c, respectively. Shear bands around the indentation are clearly seen in the rod annealed with 12T, which showing the higher plasticity, in contrast with that annealed without magnetic field.

The effect of the field annealing on the hysteresis loops of the Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_{6}$B$_{23}$ BMG is demonstrated in Figure 4. It is evident that the HMF annealing increase the saturation magnetization compared with those of annealing without magnetic field and as-cast rods. The different magnetization process indicates that the domains rotate easily by external field due to low anisotropy. While, saturation field is lowest in the rod annealed under 12T, indicating the lowest anisotropy. Therefore, the coercivity filed is also deduced from 30.5 A/m in the rod annealed without external field to 10.6 A/m in the rod annealed under 12 T.

![Figure 1](image1.png)

Figure 1. DSC traces for the Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_{6}$B$_{23}$ BMG after annealing for 1h at different states at the heating rate of 20 °C/min.

![Figure 2](image2.png)

Figure 2. Kissinger plots of the start and peak temperatures for the primary crystallization for Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_{6}$B$_{23}$ BMG.
4. Conclusions

(1) HMF can improve the thermal stability of amorphous state during structural relaxation annealing in the BMG’s supercooled liquid region as compared with the annealing without magnetic field for Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_6$B$_{23}$ bulk metallic glass.

(2) HMF can effectively suppress the brittleness of Fe$_{71}$(Nb$_{0.8}$Zr$_{0.2}$)$_6$B$_{23}$ bulk metallic glass during structural relaxation.

(3) HMF can reduce the coercivity and saturation magnetization, which is thought due to the reduced anisotropy by high magnetic field annealing.

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References


