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Crystallization kinetics of high iron concentration amorphous alloys under high magnetic fields

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ABSTRACT

Crystallization kinetics of a $Fe_{83.3}Si_{4.2}B_{12.5}$ amorphous alloy and a $Fe_{83.3}Si_4B_8P_4Cu_{0.7}$ hetero-amorphous alloy under magnetic fields was investigated by the differential thermal analysis (DTA). In DTA for $Fe_{83.3}Si_{4.2}B_{12.5}$, the first crystallization peak, which indicates the precipitation of bcc-Fe shifts about 10 K toward the lower temperature side, whereas the second peak which indicates the precipitation of iron-compounds, shifts about 4 K toward the higher temperature side by applying a magnetic field of $B = 20$ T. On the other hand, in the case of $Fe_{83.3}Si_4B_8P_4Cu_{0.7}$, the first peak at $B = 20$ T does not change in comparison with that at a zero magnetic field, whereas the second peak shifts toward a higher temperature side as well as $Fe_{83.3}Si_{4.2}B_{12.5}$.

1. Introduction

Recently, the crystallization of amorphous alloys in a high magnetic field has been extensively investigated [1–6]. The aim of these research is a development of a new process for nano-crystalline alloys using a magnetic field, which is prepared by crystallization of amorphous alloys [7]. In general, the distribution of nano-crystals in an amorphous matrix provides an enhancement of their functionalities. In the case of magnetic amorphous alloys, it dominates their magnetic properties, such as the saturation magnetization and the coercivity. Especially, the coercivity of nano-crystalline alloys strongly depends on a grain size of their precipitated crystals [8]. Hence, in order to acquire the excellent soft magnetic nano-crystalline alloys, the accurate control of their crystallization reaction such as nucleation and grain growth processes, is required.

There are several reports that dealt with the crystallization kinetics of amorphous alloys in a high magnetic field [1,3,4]. It was reported that the acceleration of crystallization takes place under magnetic fields, and this magnetic field effect was discussed by taking into account the contribution of the magnetic energy to the nucleation [1,3,4]. According to their model, under the magnetic fields, the activation energy and the critical radius for the nucleation are decreased by contribution of Zeeman energy in the case of the crystallization of ferromagnetic crystal from paramagnetic amorphous phase. These effects mean the acceleration of the nucleation by applying magnetic fields. Thus, it can be expected that an increase of the nucleation rate under the high magnetic field. The increase of the nucleation rate leads to the increase of the number of ferromagnetic crystal grains, which gives the improvement of the saturation magnetization. On the other hand, in our recent work, we found a suppression of the growth process on the crystallization in $Fe_{79}Si_{12}B_9$ amorphous alloy, although this crystallization reaction is the precipitation of ferromagnetic bcc-Fe from the paramagnetic amorphous phase [9,10]. This magnetic field effect cannot be explained by the contribution of the Zeeman energy to the nucleation. We concluded that the origin of this magnetic field effect is the suppression of atomic diffusion by applying the magnetic field [10]. The suppression of crystal growth is useful for obtaining small size crystal grains, which results in a low coercivity. Therefore, the application of the magnetic field to the crystallization process has a possibility to produce nano-crystalline alloys with excellent soft magnetic properties.

In our previous study above-mentioned, we have investigated the crystallization kinetics of the basic iron-based amorphous alloys [9,10], which do not nano-crystallize. However, in order to apply the magnetic field effects to material processing for soft magnetic nano-crystalline alloys, an investigation of the magnetic

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field effect on the crystallization kinetics of high iron concentration compounds, which exhibit the nano-crystallization, is desirable. In this study, we have carried out differential thermal analysis (DTA) measurements to investigate the crystallization kinetics in high magnetic fields of a typical hetero-amorphous alloy composition $Fe_{83.3}Si_4B_8P_4Cu_{0.7}$ [11], which is a precursor material of a soft magnetic nano-crystalline alloy, and $Fe_{83.3}Si_{4.2}B_{12.5}$ amorphous alloy that is a basic composition of $Fe_{83.3}Si_4B_8P_4Cu_{0.7}$.

2. Experimental procedure

Sample ribbons were prepared by single-roll melt-spinning. The size of ribbons is 3 mm in width and 27–29 μm in thickness for $Fe_{83.3}Si_{4.2}B_{12.5}$ and 5.5 mm in width and 22–24 μm in thickness for $Fe_{83.3}Si_4B_8P_4Cu_{0.7}$. DTA measurements in magnetic fields [12,13] were carried out in a temperature range of R.T.–900 K in applied magnetic fields up to $B = 20$ T by using a cryocooled superconducting magnet with a 52 mm room temperature bore, that was developed in High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University [14]. Samples were heated by using an electrical furnace, which is installed in the experimental bore of a superconducting magnet. The furnace is made of Pt resistance wire heater combined with water-cooled jacket [15].

The sample and the reference material temperatures were measured using sheathed Pt–13%Rh thermocouples. The reference material is an Al_2O_3 powder. We confirmed that the influence of a magnetic field on the electromotive force of the thermocouple was negligibly small. The temperature accuracy of the experiments is within 1 K. The sample and the reference material were wrapped by an aluminum foil, and were then wrapped around the thermocouple tip. The direction of applied magnetic fields is parallel to the ribbon surface. The DTA heating rate is 10 K/min. The sample space was evacuated to $\sim 10^{-3}$ Pa by a turbo-molecular pump system during the measurements. Crystal phases of each sample were determined by X-ray diffraction of annealed samples.

3. Results and discussion

3.1. $Fe_{83.3}Si_{4.2}B_{12.5}$ amorphous alloy

Fig. 1 shows a DTA curve of $Fe_{83.3}Si_{4.2}B_{12.5}$ at 0 T. Two exothermic peaks due to the crystallization reactions are observed. The first peak is caused by a crystallization of bcc-Fe, and the second peak is due to the crystallization of iron-boron compounds such as Fe_2B and Fe_3B. The first and second crystallization temperatures T_{x1} and T_{x2}, which are determined by the onset of the DTA peaks, are $T_{x1} = 706$ K and $T_{x2} = 795$ K, and the peak temperatures are $T_{p1} = 730$ K and $T_{p2} = 809$ K. Fig. 2(a and b) show the magnetic field dependence of the first and second crystallization peaks of $Fe_{83.3}Si_{4.2}B_{12.5}$, respectively. As shown in Fig. 2(a), the first peak of $Fe_{83.3}Si_{4.2}B_{12.5}$ shifts toward the lower temperature side under high magnetic fields. This result is different from that of $Fe_{79}Si_{12}B_9$ [9] that shows the crystallization peaks shift toward the higher temperature region. In our previous work, we reported that the shift of the peaks toward the higher temperature indicates a

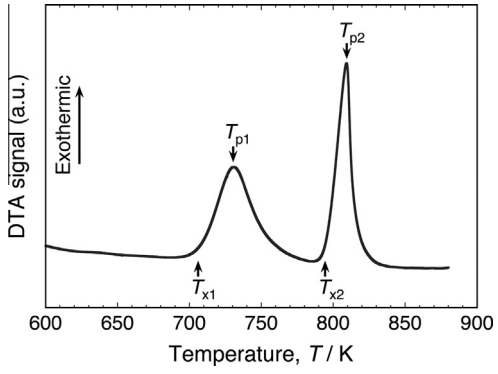

Fig. 1. DTA curve of a $Fe_{83.3}Si_{4.2}B_{12.5}$ amorphous alloy with heating rate 10 K/min in a zero magnetic field. T_{x1} and T_{x2} are the crystallization temperatures determined by the onset of the exothermic peak. T_{p1} and T_{p2} are peak temperatures.

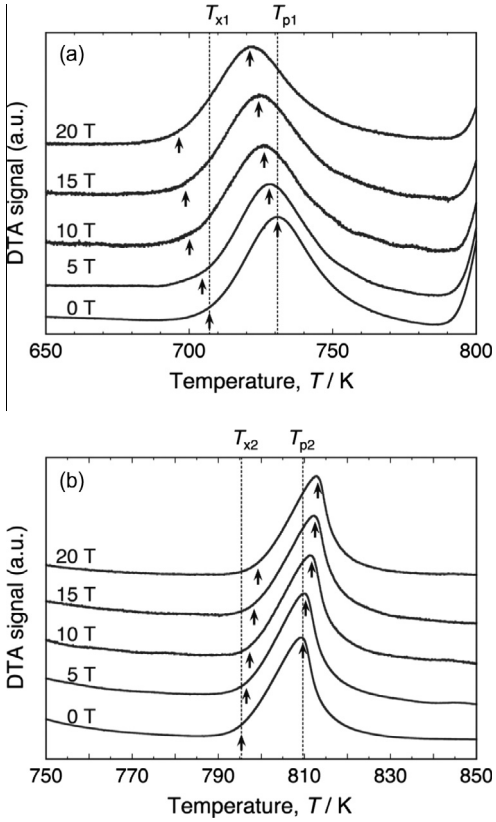

Fig. 2. Magnetic field dependence of (a) first and (b) second crystallization peaks of a $Fe_{83.3}Si_{4.2}B_{12.5}$ amorphous alloy.

suppression of the crystallization of $Fe_{79}Si_{12}B_9$ [9,10]. On the other hand, the shift toward the lower temperature side of the first peak in $Fe_{83.3}Si_{4.2}B_{12.5}$ suggests an acceleration of the crystallization under high magnetic fields, which is consistent with the expectation from the contribution of the gain of the Zeeman energy. It is considered that this difference of the magnetic field dependence of the first crystallization peak between $Fe_{83.3}Si_{4.2}B_{12.5}$ and $Fe_{79}Si_{12}B_9$ is caused by a difference of the effective magnetic moment between the crystal phases of $Fe_{83.3}Si_{4.2}B_{12.5}$ and $Fe_{79}Si_{12}B_9$. In both cases, bcc-Fe contained Si [bcc-Fe(Si)] precipitates on the first crystallization. However, Si concentration in bcc-Fe(Si) precipitated in $Fe_{79}Si_{12}B_9$ is higher than that of $Fe_{83.3}Si_{4.2}B_{12.5}$, because a larger amount of Si is contained in its alloy composition. Therefore, the effective magnetic moment of bcc-Fe(Si) precipitated in $Fe_{79}Si_{12}B_9$ is likely smaller than that of $Fe_{83.3}Si_{4.2}B_{12.5}$. In fact, the Curie temperature $T_{C,cry.} = 1026$ K of the crystal phase in $Fe_{83.3}Si_{4.2}B_{12.5}$, which is determined by our magnetization measurements (not shown here) is higher than $T_{C,cry.} = 946$ K of $Fe_{79}Si_{12}B_9$. The acceleration of the crystallization and the suppression of the crystal growth by applying the magnetic field, reported so far, are considered to have different origins. The acceleration occurs in the nucleation process due to a gain of the Zeeman energy, while the suppression is caused by the reduction of the atomic diffusion in the crystal growth process [10]. Thus, we speculate that the acceleration of the nucleation and the suppression of the crystal growth by applying magnetic fields compete on the crystallization process of bcc-Fe. When the crystal phase has a large magnetic moment, the contribution of the Zeeman energy becomes dominant. As a result, the acceleration behavior appears in $Fe_{83.3}Si_{4.2}B_{12.5}$. By contrast, when the magnetic moment of the crystal phase is small, the effect from the Zeeman energy smears out, and then the suppression behavior appears in $Fe_{79}Si_{12}B_9$. Thereby, even though