JOURNAL OF MATERIALS SCIENCE 25 (1990) 3159–3161

Effects of annealing on magnetic properties of $Fe_{78}B_{13}Si_9$

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We have annealed amorphous $Fe_{78}B_{13}Si_9$ samples at temperatures T_a = 355 to 460° C for time t_a = 10 to 60 min, in an atmosphere with purging of nitrogen gas. After thermal treatment, we examined the hysteresis loop, DSC curve, and the X-ray diffractogram of each sample. The coercivity H_c, the peak induction B_{max}, and the loss W_h were analysed to study the affects of annealing. Stress-relief and partial crystallization are two important parameters in determining optimal annealing conditions.

1. Introduction

Metallic glasses obtained by the melt-spinning method are useful in many industrial applications. In particular, the commercialized $Fe_{78}B_{13}Si_9$ glass (2605S2) is characterized by high magnetization $M_s \simeq 1.5$ T and high magnetostriction $\lambda_s \simeq 3 \times 10^{-5}$ from other glasses [1]. Large residual stresses formed during casting, mean that proper annealing is usually needed for this kind of glass to improve its magnetic properties such as peak induction B_{max}, coercivity H_c, and the loss W_h. However, since the $Fe_{78}B_{13}Si_9$ glass is in a metastable state, the annealing conditions including the annealing temperature T_a, and the annealing time t_a should be properly defined so that no excessive crystallization occurs. Generally, crystallization is deleterious except for some applications in the high frequency range [2]. Also, as discussed in this paper, by annealing the $Fe_{78}B_{13}Si_9$ glass in a protected atmosphere, partial crystallization would have started for $T_a > 400$° C, irrespective of the annealing time chosen. In this paper, the affects of annealing on low frequency (60 Hz) properties will mainly be discussed.

2. Experimental procedures

The $Fe_{78}B_{13}Si_9$ glass was purchased from the Allied corporation, USA. Our samples were cut from a master ribbon with dimensions of 1.5 × 150 × 0.022 mm^3. Depending on the annealing conditions as shown in Table I, we have labelled the samples from 0 to 24. The heat treatment was performed in the atmosphere with purging of purified nitrogen gas. The flow rate was set at 50 ml min^{-1}. The heating rate was kept around 5° C min^{-1}. The sample is set in a bed made of stainless steel. By automatic temperature control, T_a was stabilized to within 1° C of the programmed temperature. After a preset annealing time t_a, the sample was immediately extracted from the furnace, and cooled rapidly to room temperature [3].

After various annealings, samples 0 to 24 were obtained for the hysteresis measurements at 60 Hz. A *BH* loop tracer was set up for this purpose [4]. The maximum external field used was 6 Oe. The data of peak induction, coercivity, and loss were gathered together and analysed by an IBM PC.

For auxiliary measurements, we have cut small pieces of 0 to 24 samples and run their DSC data in a DuPont 9900 system. The heating rate used was also set at 5° C min^{-1}. Sample 2 to 4, 13, and 24 ($T_a > 385$° C) were analysed with Cu Kα X-ray to determine the crystallized phase [5]. To further determine the thickness of the crystallized layer, each sample was etched about 1 μm each side in a polishing solution of 30% HNO_3, 10% H_2SO_4, 10% H_3PO_4, 50% CH_3COOH, and exposed to the same X-ray for comparison. The thickness of the sample was determined by the density and mass method [6].

3. Results and discussion

All the samples, except sample 1, show the characteristics of two-peak DSC curves. As in Fig. 1, the first crystallization peak appears at T_{x1} = 527.9° C, and the second peak at T_{x2} = 542.1° C. However, it is clear that for sample 1, the first peak has disappeared. Hence, although it is difficult to associate the first peak totally with the α-ion crystallization, we may still conclude that for T_a = 460° C sample 1 has crystallized substantially. This sets the upper limit of any useful annealing condition for $Fe_{78}B_{13}Si_9$ glass.

To make certain of the crystallized phase and its distributed thickness in the annealed $Fe_{78}B_{13}Si_9$ glass, we scanned several samples (as discussed in Section 2) in a X-ray diffractometer. Sample 24 (T_a = 385° C) did not show any obvious pattern of crystallization. However, all the rest showed the similar results to that of sample 13, shown in Fig. 2. For the unetched sample 13, the observed peak at $2\theta \simeq 44.8°$ is associated with α-Fe or α-(FeSi) [5]. After etching, the peak disappeared. This may indicate that partial

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TABLE I Annealing conditions of samples

Sample	T_a (°C)	t_a (min)
0	as-cast	
1	460	10
2	445	10
3	430	10
4	415	10
5	415	20
6	415	30
7	430	20
8	400	20
9	400	30
10	400	40
11	385	60
12	385	40
13	400	10
14	385	30
15	385	20
16	370	60
17	370	40
18	370	30
19	370	20
20	355	60
21	355	20
22	355	40
23	355	30
24	385	10

crystallization has occurred in surface layers of about 0.5 to 1 μm thick in these samples. Although we did not check other samples, it is believed that for a longer annealing time, the degree of crystallization or the layer thickness increases further. These affects will be seen and discussed in the context of the magnetic properties.

Fig. 3 shows the coercivities H_c of samples 0 to 24 measured from the maximum field to the 1 Oe *BH* loop. For the as-cast sample $H_c \simeq 0.17$ Oe. H_c reaches a minimum ($\simeq 0.075$ Oe) for the batch of $T_a = 370°$ C samples. Hence, many of the stresses have been removed in samples for these kinds of annealing. By raising T_a further, H_c increases. Moreover, as $T_a > 400°$ C, H_c increases distinctly with annealing time t_a even for a relatively short anneal which indicates that crystallites enhance pinning. To explore more of the magnetic properties, we plotted the peak induction B_{max} of each sample for the 6 Oe *BH* loop as in Fig. 4. The horizontal axis of Fig. 4 was so placed that both T_a and t_a increase from left to right. It is then obvious that by annealing at $T_a = 355°$ C and $t_a = 40$ min, B_{max} reaches the highest

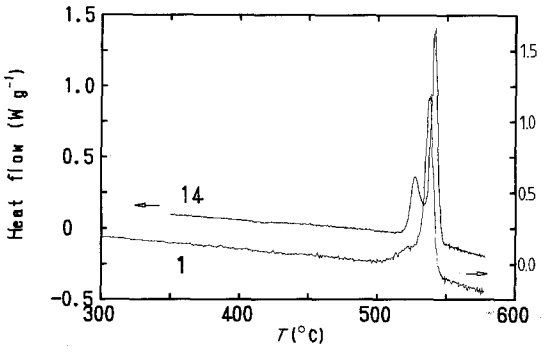

Figure 1 Differential scanning calorimeter recordings of samples 14 and 1.

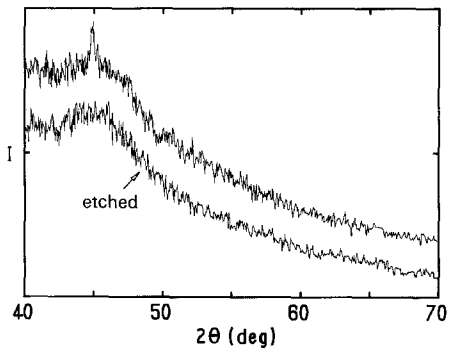

Figure 2 X-ray diffractograms of sample 13 (unetched) and 13 (etched) samples.

value of 1.47 T. This also shows that $T_a = 355$ to 370° C annealing is sufficient for stress relief in the sample. Continuing to raise T_a and to prolong t_a will result in a slight decrease of B_{max} until $T_a \simeq 400°$ C. For $T_a > 400°$ C, partial crystallization affects B_{max} in two ways: (a) a tensile stress in the crystalline surface layer will force magnetization to lie in the surface plane, and α-Fe crystallites will enhance the magnetization (per unit volume), (b) a compressive stress in the amorphous layer will tilt the magnetization out of the plane of layer [7]. Hence, by combining these two affects, it is not difficult to explain the observed decreasing and then increasing behaviours of B_{max} for $T_a > 400°$ C.

Finally, for discussion of loss W_h, Fig. 5 shows W_h at $B_{max} = 1.2$ T for all samples. The horizontal axis is similar to that of Fig. 4. The loss in the as-cast sample (0) is too large ($\simeq 1.5$ W kg^{-1}) to be shown in the figure. From Fig. 5, it is clear that there exists a minimum of W_h for all the thermally treated samples. This minimum lies closely in the batch of $T_a = 385°$ C, $t_a > 20$ min samples. Thus, to obtain an optimal sample from annealing for low frequency uses, it is better to release the stress in the sample completely rather than to partially crystallize it.

4. Conclusions

We have discussed the affects of annealing ($T_a = 355$ to 460° C, and $t_a = 10$ to 60 min) on the magnetic properties of $Fe_{78}B_{13}Si_9$. For T_a in the range of 370 to 385° C, and t_a in the range of 20 to 60 min, it is sufficient to stress-relieve the sample, and to optimize its magnetic properties. To anneal at higher T_a or longer t_a ($T_a > 400°$ C) will crystallize the sample. We have

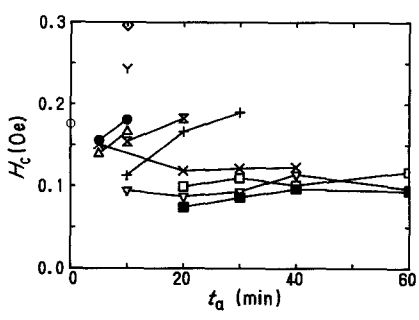

Figure 3 The coercive force H_c ($H_{max} = 1$ Oe) of samples 0 to 24. (□ 355° C, ■ 370° C, ▽ 385° C, △ 390° C, × 400° C, ● 410° C, + 415° C, ⧖ 430° C, Y 445° C, ◇ 460° C, ○ cast).

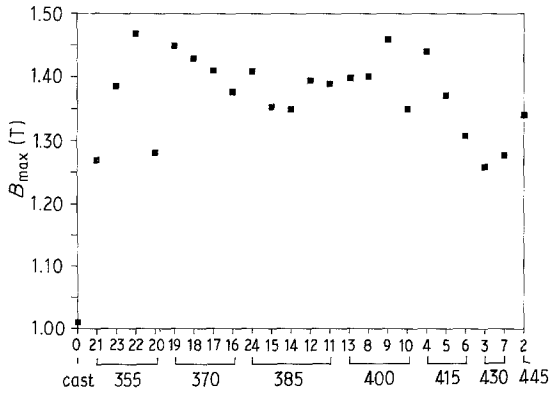

Figure 4 The peak induction B_{max} (H_{max} = 6 Oe) of samples 0 to 24.

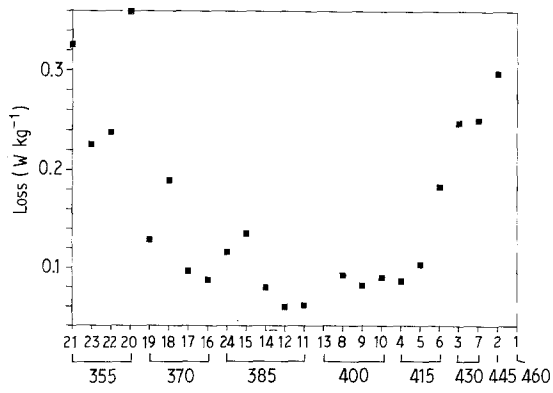

Figure 5 The loss W_h at B_{max} = 1.2 T for samples 0 to 24.

found the crystallized phase to be α-Fe related. Our conclusions have been based on the condition of anneals in a protecting atmosphere. If the oxidizing environment during anneals had been changed, the result would have been different. It is still not clear which mechanism triggers the surface crystallization, and therefore changes the upper limit for the optimal anneal [8]. Evidence has shown that by annealing $Fe_{78}B_{13}Si_9$ in vacuum, the optimal condition may be shifted to 400 to 410° C [9].

Acknowledgements

We are grateful for partial financial support from the China Steel Corporation under the project TRC-77-32.

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Received 27 February
and accepted 30 August 1989