A Study of Magnetic Properties in $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}$ $B_{7.6}$ by Magnetic Annealing

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The crystallographic and high frequency characteristics of $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}$ $B_{7.6}$ soft magnetic alloys were investigated under the magnetic field annealing. The crystallization fraction of annealed samples with longitudinal magnetic field is higher than that of samples without magnetic field. When the transverse magnetic field is applied, the crystallization fraction does not increases but decreases until 500℃. It is found that for all samples, the saturation induction are all same with 1.3 T. The coercive field of as-cast sample is 1.06 A/cm, but in annealed samples it decrease from 0.56 to 0.1 A/cm with increasing annealing temperature from 400 to 550℃. The squareness of annealed samples under transverse magnetic field has a small value than that of both without field and with longitudinal field annealing. It is noted that the magnetic field annealing with transverse direction to amorphous $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}B_{7.6}$ profoundly influenced on the Mössbauer spectra in contrast to that with longitudinal direction and without magnetic field.

Keywords : Nanocrystalline, Amorphous, Magnetic Annealing, Mössbauer

1. INTRODUCTION

Amorphous magnetic alloys have been extensively investigated for the past decade and are used for many applications[1-4].

For example, due to their excellent soft magnetic properties with the high magnetic flux density and permeability[1],[5],[6], Fe-based soft magnetic alloys have been applied to magnetic components.

Nanocrystalline Fe-Cu-Nb-Si-B alloys reveal a homogeneous ultra-fine grain structure of bcc FeSi with grain sizes typically 10-15nm and random texture[7]. Owing to the small grain size the local magnetocrystlline anisotropy is randomly averaged out by exchange interaction[8-9] and the structural phases present lead to low or vanishing saturation magnetostriction[7],[10]. This leads to superior soft magnetic . properties comparable to those of permalloys or near zero-magnetostrictive Co-based amorphous alloys.

As in other soft magnetic materials, magnetic field annealing induces an uniaxial anisotropy, the easy axis being parallel to the magnetic field applied during the heat treatment[11]. This allows the shape of the hysteresis loop to be varied according to the demands of various applications

The induced anisotropy energy, K depends both on the annealing conditions and the alloy composition. The influence of annealing conditions has been recently discussed for nanocrystalline by Yoshizawa and Yamauchi[12].

In this paper crystallographic and magnetic properties of nanocrystalline alloys with the composition $Fe_{73.9}Cu_{1.0}Nb_{3.0}Si_{13.5}B_9$ according to the annealing temperatures and methods with and without magnetic field are investigated.

2. EXPERIMENTAL

Amorphous ribbons of composition $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}B_{7.6}$ were prepared by single roller method. The ribbons, typically 3 mm wide and 25

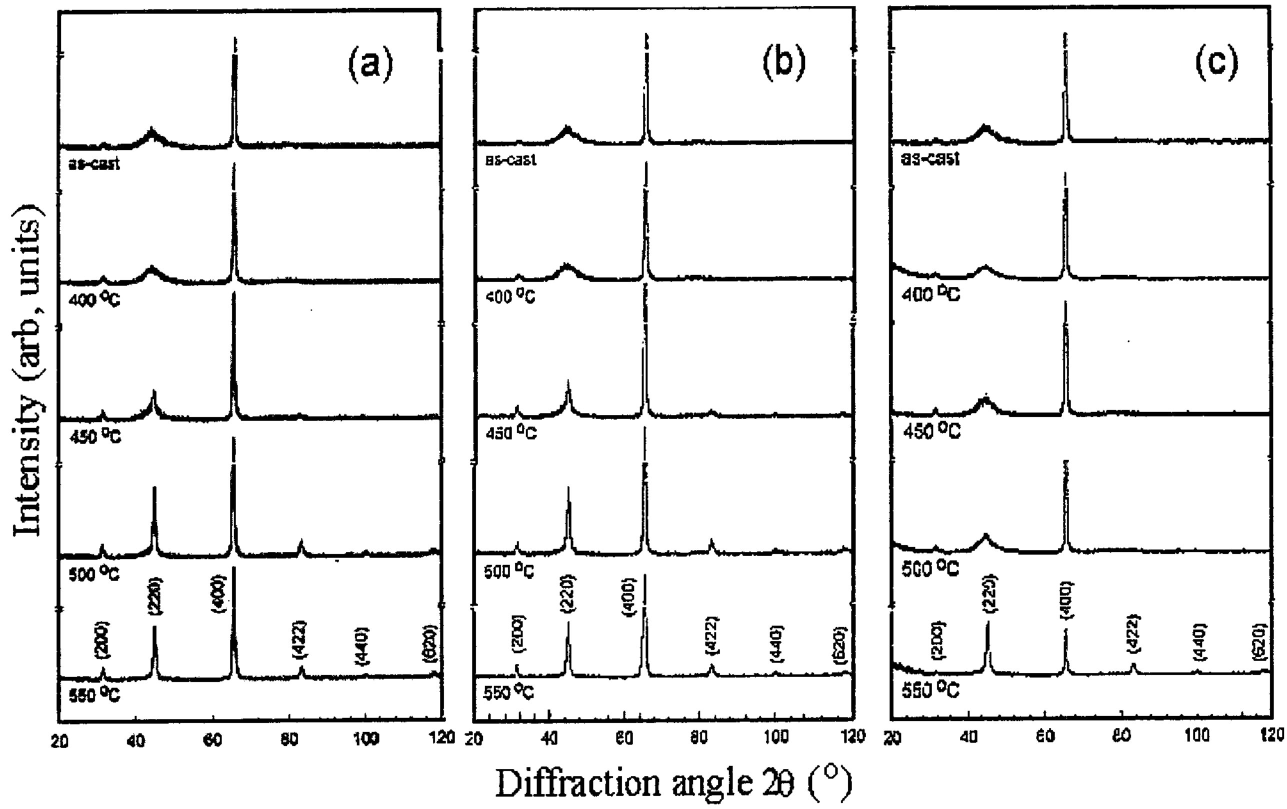

Fig. 1. X-ray-diffraction patterns for annealed samples for 1 hour between 400℃ and 550℃ (a) without external field, (b) with externally longitudinal magnetic field, (c) with externally transverse magnetic field.

μm thick, were wounded to troidal cores with 17 mm inner and 23 mm outer diameter, and annealed in protecting nitrogen atmosphere with the longitudinal field or the transverse magnetic field and without magnetic field for 1 hour at 400℃, 450℃, 500℃ and 550℃, respectively.

To obtain the structural information of as-cast and the annealed samples, x-ray diffractometer(12 kW, Rotaflex D/MAX-RB wide-angle goniometer) with CuKα was used, and the accelerating voltage and current were set to 50 kV and 100 mA respectively. Using θ - 2θ reflection method with rotating sample, we measured x-ray intensity as a function of reflection angle, 2θ, between 20° and 120°.

In order to measure coercive force, remanance and saturation magnetic induction, we used a single strip B-H loop tracer operating up to 100 kHz magnetizing frequency. The B-H signals were digitized using 12-bit resolution of digital storage oscilloscope (Nicolet Pro32) and the digitized data were transferred to PC via IEEE-488 bus.

3. RESULTS AND DISCUSSION

Fig. 1 shows x-ray-diffraction patterns for annealed samples for 1 hour between 400℃ and 550℃ (a) without external field, (b) with externally longitudinal magnetic field, (c) with externally transverse magnetic field.

As-cast ribbon with which already imbedded nanocrystalline Fe-Si phase on the surface have a preferred orientation with(400) plane to the surface and also with the [011] direction parallel to the ribbon length.

X-ray-diffraction patterns for annealed samples at 400℃ are similar to those for as-cast samples, but over 450℃ it is found that a crystalline Fe-Si phase with DO_3 near θ = 44.5° is detected and the other several diffraction patterns occur in Fig. 1 (a).

In Fig. 1 (b) when the longitudinal magnetic field is applied, the x-ray-diffraction patterns for annealed samples are nearly like those for annealed samples without magnetic field, but the crystalline Fe-Si phase of annealed samples with longitudinal magnetic field increased in comparison with that of without magnetic field.

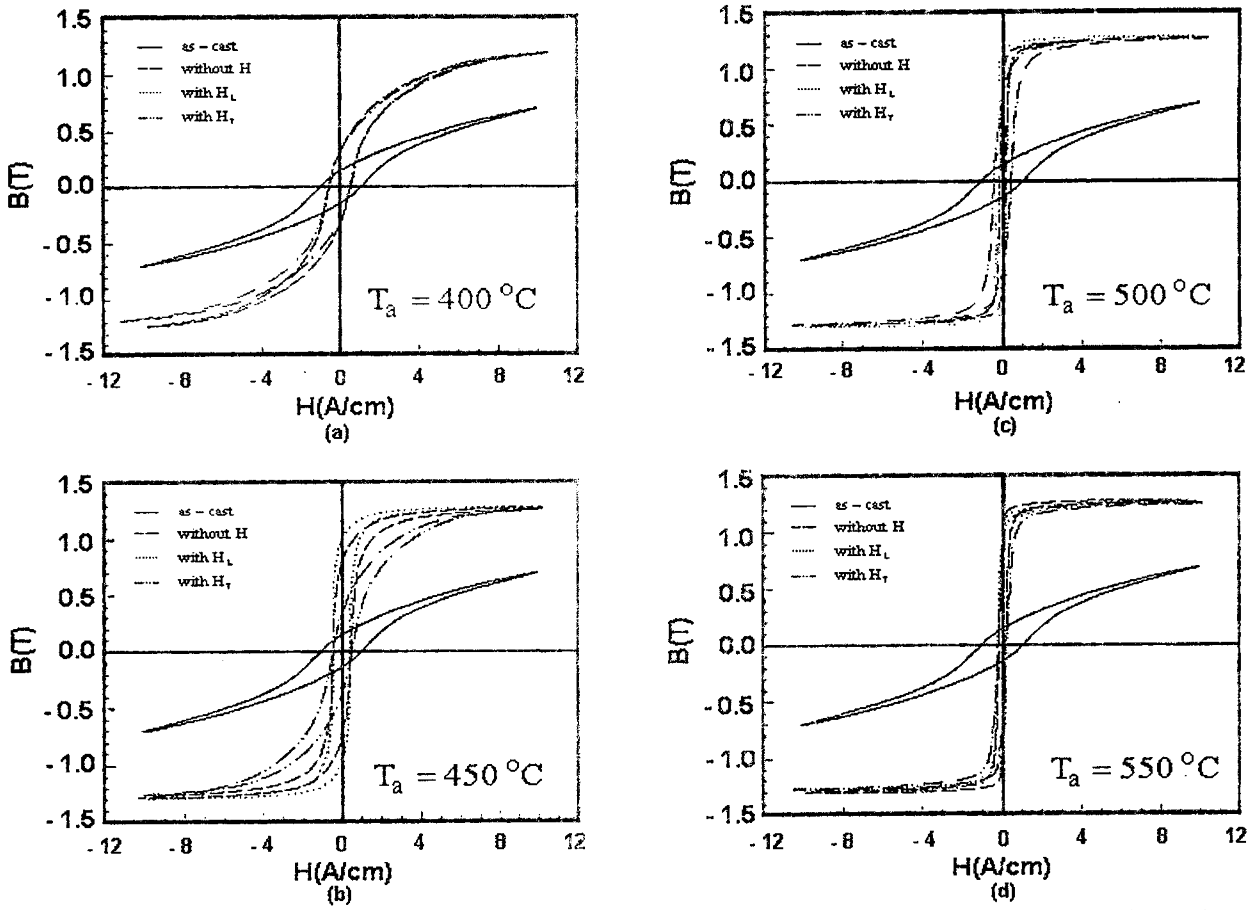

Fig. 2. The magnetic hysteresis curves under the magnetizing frequency of 10 kHz of annealed samples with and without external magnetic field at (a) 400℃ (b) 450℃ (c) 500℃ (d) 550℃, respectively.

With increasing annealing temperature the crystallization fraction of annealed samples with longitudinal magnetic field is higher than that of samples without magnetic field. Fig. 1 (c) shows that in the case of external transverse magnetic field annealing the crystalline Fe-Si phase (200) scarcely emerges on the x-ray-diffraction patterns for annealed samples at 500℃.

When the transverse magnetic field is applied, the crystallization fraction does not increases but decreases until 500℃.

The formation of nanocrystalline phase does not appear on the surface layer until 550℃ annealing temperature under the transverse field. However, the crystallization of internal regions can be confirmed by x-ray diffraction measurement via tilting the sample.

Fig. 2 shows the magnetic hysteresis curves under the magnetizing frequency of 10 kHz of annealed samples with and without external magnetic field at (a)400℃ (b)450℃ (c)500℃ (d)550℃, respectively.

The B-H loops of annealed sample at 400℃ were constant and irrelevant to the direction of the induced external magnetic field in Fig. 2 (a). This result is consistent with the x-ray diffraction patterns for the samples annealed at 400℃ that the crystallization fraction of samples annealed at 400℃ was unchanged regardless of the induced external magnetic field. However, it is found that with increasing annealing temperature the B-H loops of annealed samples at 450, 500 and 550℃ are dependent upon the direction of the induced external magnetic field in Fig. 2 (b) - (d).

It is also found that for all samples, the saturation induction are all the same with 1.3 T which is two times as that of as-cast sample. It is because as-cast sample dose not reach the magnetic saturation under low frequency magnetic field.

The coercive force of as-cast sample is 1.06 A/cm, but in annealed samples it decrease from 0.56 to 0.1 A/cm with increasing annealing temperature from 400 to 550℃. This indicates that the formation of nanocrystalline grew increasingly with increasing annealing temperature and Fe-B system with high crystalline magnetic anisotropy disturbed the movement of the magnetic domain wall.

The squareness of annealed samples under transverse magnetic field has a small value than that

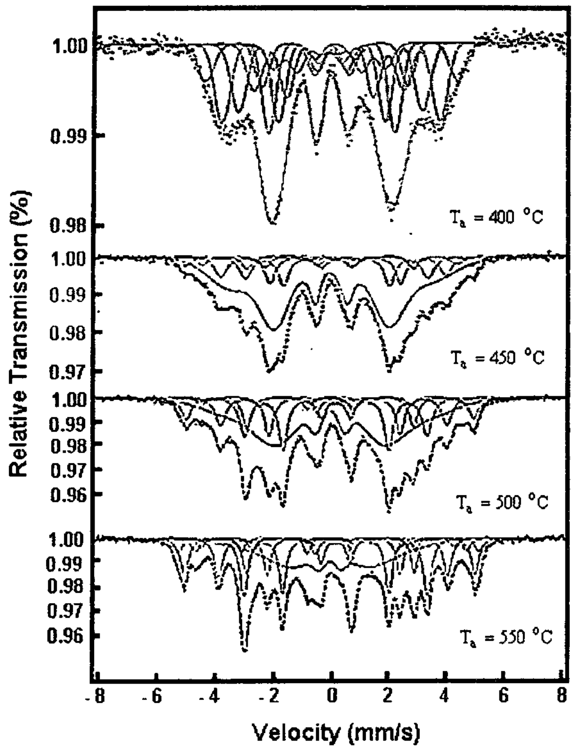

Fig. 3. Mössbauer spectra taken at room temperature for annealed $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}B_{7.6}$ from 400℃ to 550℃ with externally longitudinal magnetic field for one hour.

of both without field and with longitudinal field annealing[13].

Fig. 3 shows Mössbauer spectra taken at room temperature for annealed $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}$ $B_{7.6}$ from 400℃ to 550℃ with externally longitudinal magnetic field for one hour. Those spectra for the samples consist of six-well defined but broad lines.

Five sets of six lines indicate the discrete hyperfine field, respectively. However, as annealing temperature is raised above 500℃, sharp lines of crystalline phase appear and Mössbauer spectrum for the completely crystallized sample consist of five sextets. Least-square fitting has been carried out using Gaussian line shapes for amorphous shape and Lorentzian line shapes for the crystalline phases.

The reason why Gaussian line shapes were based instead of the usual Lorentzian line shapes was based on the result of Morrish et al.[14] that when the broadened absorption lines of amorphous solids were separated by an external magnetic field, Gaussian line shapes gave better fits.

Table I shows room-temperature Mössbauer parameters for annealed $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}$ $B_{7.6}$ as a function of the annealing temperature.

It is found that there exists no differences in Mössbauer spectra between the annealed samples for one hour without magnetic field and with externally longitudinal magnetic field from 400℃ to 550℃. But, Mössbauer spectra for the annealed samples with externally transverse magnetic field are the same as those for as-cast sample.

It is evident that the magnetic annealing with longitudinal direction to amorphous $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}B_{7.6}$ hardly affect the Mössbauer spectra in comparison with the annealing without magnetic field.

However, it is noted that the magnetic field annealing with transverse direction to amorphous $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}$ $B_{7.6}$ profoundly influenced on the Mössbauer spectra in contrast to that with longitudinal direction.

Conclusively, the magnetic annealing with transverse direction to amorphous $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si_{14.0}$ $B_{7.6}$ disturbs the formation of nanocrystalline phase.

4. CONCLUSION

1. The crystallization fraction of annealed samples with longitudinal magnetic field is higher than that of samples without magnetic field.
2. It is found that for all samples, the saturation induction are all same with 1.3 T.
3. The coercive field of as-cast sample is 1.06 A/cm, but in annealed sample it decrease from 0.56 to 0.1 A/cm with increasing annealing temperature from 400
4. The squareness of annealed sample under transverse magnetic field has a small value than that of both without field and with longitudinal field annealing.
5. It is noted that the magnetic field annealing with transverse direction to amorphous $Fe_{73.9}Cu_{1.0}Nb_{3.5}$ $Si_{14.0}$ $B_{7.6}$ profoundly influenced on the Mössbauer spectra in contrast to that with longitudinal direction and without magnetic field.

Table I . Room-temperature Mössbauer parameters for annealed $Fe_{73.9}Cu_{1.0}Nb_{3.5}Si1_{14.0}$ $B_{7.6}$ with externally longitudinal magnetic field for one hour at 500 and 550℃.

Annealing T(°C)	Line	H_{hf}(kOe)	ε(mm/s)	δ(mm/s)
	1	320.0	0.00	0.03
	2	309.2	0.00	0.08
500	3	280.9	-0.04	0.20
	4	243.3	-0.01	0.22
	5	196.0	0.00	0.27
	1	322.5	0.00	0.03
	2	312.3	0.00	0.09
550	3	291.0	0.00	0.13
	4	246.0	0.00	0.20
	5	197.1	0.00	2.27

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