Soft magnetic properties of amorphous $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloy treated by pulsed magnetic field and annealing*

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The crystallization, microstructure, and soft magnetic properties of $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloy are studied. Amorphous $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloys are first treated by a pulsed magnetic field with a medium frequency, and then annealed at 100 °C–400 °C for 30 min in a vacuum. The rise in temperature during the treatment by a pulsed magnetic field is measured by a non-contact infrared thermometer. The soft magnetic properties of specimens are measured by a vibrating sample magnetometer (VSM). The microstructure changes of specimens are observed by a Mössbauer spectroscopy and transmission electron microscope (TEM). The results show the medium-frequency pulsating magnetic field will promote nanocrystallization of the amorphous alloy with a lower temperature rise. The nanocrystalline phase is α-Fe(Co) with bcc crystal structure, and the grain size is about 10 nm. After vacuum annealing at 100 °C for 30 min, scattering nanocrystalline phases become more uniform, the coercive force and the saturation magnetization of the specimens are 41.98 A/m and 185.15 emu/g.

Keywords: amorphous alloys, pulsed magnetic field, vacuum annealing, nanocrystallization

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1. Introduction

Since Birringer *et al.*[1] presented nano-material, the nano-materials have been developing rapidly.[2] Nanocrystalline soft magnetic alloy is characterized by higher saturation magnetic induction, low coercivity, high permeability, and low high-frequency power loss. Its applications cover all fields of using soft magnetic materials, such as work transformers, pulse transformers, high frequency transformers, mutual inductors, and magnetic switches. Thus, the processing and synthesis techniques of nanocrystalline alloy are important research topics in the nano-material area. Various methods have been used for preparing the nanocrystalline alloy. Relatively, the crystallization of an amorphous alloy is used commonly, which includes conventional annealing,[3,4] the flash annealing,[5,6] treatment by radio-frequency magnetic field[7,8] or ultrashort pulse current.[9,10] Conventional annealing is mostly used, but it may lead to material embrittlement at high temperature. In recent years, some research groups have studied the effect of a pulsed magnetic field on Fe-based amorphous alloys.[11−13] Chao *et al.*[14] have reported on nanocrystallization of $Fe_{78}Si_9B_{13}$ amorphous alloy and have obtained two-phase nanocrystalline magnetic alloys with better soft magnetic properties. To improve the soft magnetic properties and restrain embrittlement at high temperature annealing of amorphous alloy, in this article we study the soft magnetic properties of amorphous $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloy treated by a pulsed magnetic field and annealing.

2. Experiment

Ingots of $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloys were prepared by arc melting in an argon atmosphere. Amorphous ribbons 2 mm in width and 25 μm in thickness were prepared by using the single-roller melt-spinning technique. The amorphous alloy was treated by a pulsed magnetic field in program-controlled pulsed magnetic field equipment. The experimental conditions were as follows: pulsed magnetic field intensity was 250 Oe (1 Oe=79.5775 A/m), impulse frequency was 1000 Hz and process-time was 10 min. The temperature rise in

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the treating process was measured by a sensitive digital thermometer. After the treatment of the pulsed magnetic field, a series of 30-min vacuum annealing treatments was carried out at a temperature ranging from 100 °C to 400 °C. Table 1 shows the condition of treatment, and each group has 10 specimens. The vibrating sample magnetometer (VSM) was used to measure the saturation magnetic induction (M_s) and coercivity (H_c). The microstructural changes of some specimens were analysed by a Mössbauer spectrometer and transmission electron microscopy (TEM). The Mössbauer spectrum was obtained by a constant acceleration Mössbauer spectrometer controlled by computer. The source of γ-ray was ^{57}Co/Pd. The experimental data were fitted by MSU8 software, and the accuracy of fitting was $\chi^2 \to 1.0$.

Table 1. Parameters of annealing treatment after the pulse magnetic field treating.

Sample No.	Annealing temperature/°C	Annealing time/min
1	100	30
2	200	30
3	300	30
4	400	30

3. Results and discussion

Figures 1(a) and 1(b) show the Mössbauer spectra of specimens before and after the treatment by pulsed magnetic field with medium frequency respectively. The experimental condition is as follows: pulsed magnetic field intensity is 250 Oe, impulse frequency is 1000 Hz, and process-time is 10 min.

In Fig. 1(a), the spectra show broadened and overlapped sextet, assigned to the structural disorder of an amorphous state. In Fig. 1(b), the spectra also show broadened and overlapped sextet, which show the main characteristics of ferromagnetic amorphous remain in the samples after the treatment. However, according to the fitting result of the Mössbauer spectra shown in Fig. 1(b), initial crystal is found, and the crystal precipitated phase is α–Fe (Co) with content of 17.073%. It should be considered that the initial nanocrystal is produced in the process of the treatment by pulsed magnetic field. The temperature increase of amorphous $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloy in the treating process is measured by a non-contact infrared thermometer and it is less than 10 °C. Therefore, the treatment of a pulsed magnetic field with medium frequency promotes the nanocrystallization of Fe-based amorphous alloy under low temperature conditions.

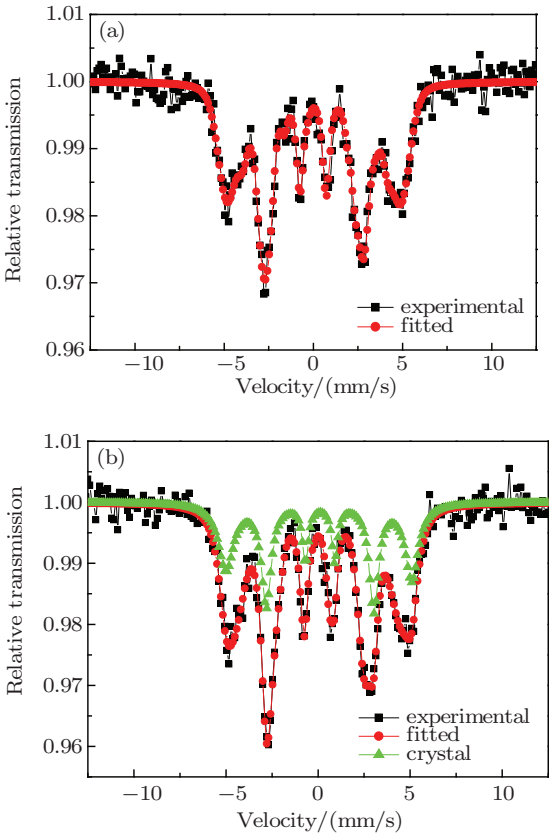


Fig. 1. (colour online) Mössbauer spectra of amorphous alloy specimens before (a) and after (b) treatment of a pulsed magnetic field with medium frequency.

Figures 2(a) and 2(b) show the TEM image and selected area electron diffraction (SAED) image of the amorphous alloy respectively. The SAED image shows the typical feature of amorphous structure morphology, that the diffraction pattern is composed of a wide halo and blurring circles. Figure 3 shows TEM micrograph (a) and SAED image (b) of amorphous alloy after treatment by a pulsed magnetic field with medium frequency. In TEM micrograph (Fig. 3(a)), there are some small white precipitates appearing in amorphous phases, which are the crystal precipitated phases. Thus, the amorphous changes to a two-phase nanocrystalline alloy after it has been treated by the pulsed magnetic field with medium frequency. The nanocrystalline phase is bcc-Fe (Co) with an average grain size of about 10 nm and it is embedded in the remaining amorphous matrix. Figures 4(a) and 4(b) display the TEM micrograph and SAED of the nanocrystalline alloy after annealing at 100 °C for 30 min, respectively. The crystal precipitated phases are more uniformly distributed than the scenario in Fig. 3(a).

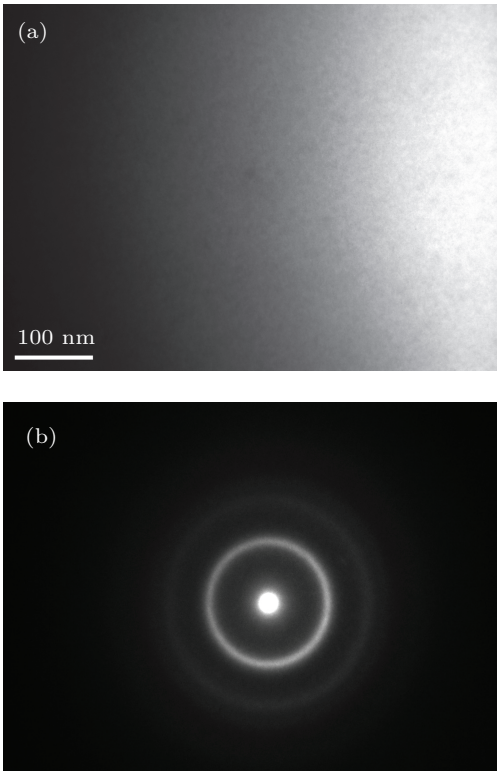


Fig. 2. TEM micrograph (a) and SAED (b) of amorphous alloy.

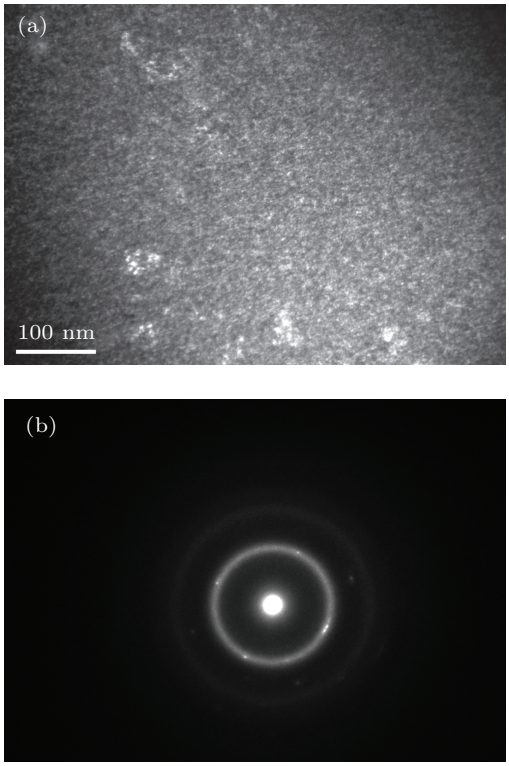


Fig. 3. TEM micrograph (a) and SAED (b) of amorphous alloy after treatment by pulsed magnetic field with medium frequency.

The vibrating sample magnetometer (VSM) is used to measure the saturation magnetic induction and the coercive force of the specimens. Table 2 shows the VSM measured results. It can be seen from Table 2 that No. 1 (annealed at 100 °C/30 min) specimen displays more excellent properties as a soft magnetic alloy. The coercive force and the saturation magnetization of the sample are 41.98 A/m and 185.15 emu/g. The coercive force increases and the saturation magnetization reduces with the increase of annealing temperature.

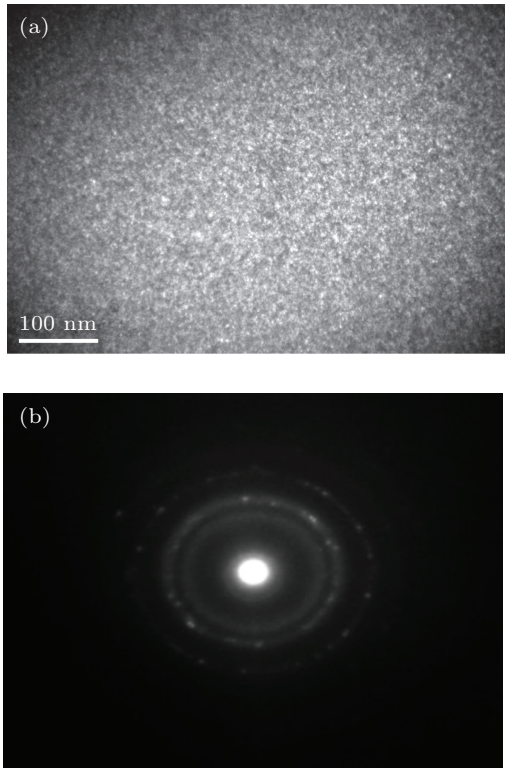


Fig. 4. TEM micrograph (a) and SAED (b) of nanocrystalline alloy after annealing at 100 °C for 30 min.

Table 2. Values of coercive force and saturation magnetization of alloy specimens.

Samples	H_c/(A/m)	M_s/(emu/g)
Amorphous	44.50	147.87
Nanocrystalline	47.81	143.66
1	41.98	185.15
2	56.82	159.93
3	66.26	171.71
4	72.11	172.04

To sum up, nanocrystallization of amorphous $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloy is promoted by the treatment of a pulsed magnetic field with medium frequency, and the soft magnetic properties of specimens are much improved after annealing at 100 °C/30 min compared with before annealing. As is well known, the melt-spinning technique is adopted mostly to prepare the

amorphous alloy and the preparation of the normal nucleation and growth of grain is restrained. So, the amorphous alloy is in a thermodynamically metastable state at room temperature. If the external treatment condition is changed, there appears a naturally translating tendency from low-energy metastable state to stable crystalline state. The pulsed magnetic field is a physical field with high energy. The activating force is exactly provided for amorphous alloy transition when a pulsed magnetic field is used to treat the amorphous $Fe_{52}Co_{34}Hf_7B_6Cu_1$ alloy, thus causing atomic periodic oscillation and leading to the crystallization of the amorphous alloy. However, atoms are difficult to produce a long-range diffusion in a short time and at low temperature in the process of crystallization, therefore, there appear many substructures, internal stress, and defects in boundary components and alloy. Then, annealing may lead to the structure relaxation, the residual stress release and the structure homogenization as shown in Fig. 3, the crystallization phases present a dispersive distribution after annealing at 100 °C for 30 min. Crystal orientation has no regular arrangement and the angles between crystal grain easy magnetization directions are big, so the exchange coupling effect of crystal grain leads to the reduction of the effective anisotropy, and therefore, reduction of the coercive force.

The saturation magnetization of specimen (M_s) is determined by the crystal phases and non-crystal phases: $M_s = V_1 M_{s_1} + V_2 M_{s_2}$. M_{s_1} and M_{s_2} represent saturation magnetizations of crystal phase and non-crystal phase respectively, V_1 and V_2 denote the volume fractions of crystal phase and non-crystal phase respectively. For the same material, the saturation magnetization of crystal is bigger than that of non-crystal. In contrast with the pulsed magnetic field with medium frequency, the volume fraction of crystal phase (V_1) does not change too much after annealing at 100 °C for 30 min. But the dispersive distribution of crystallization phases leads to saturation magnetization rising.

4. Conclusions

(i) The treatment of a medium-frequency pulsating magnetic field will expedite the nanocrystallization of the amorphous alloy at low temperature ($\Delta t \leq 10$ °C). The nanocrystalline phases are α–Fe (Co) with bcc crystal structure, and the grain size is about 10 nm on average.

(ii) After vacuum annealing at 100 °C for 30 min, the distribution of nanocrystalline phases becomes more uniform, and the soft magnetic properties of specimens are much improved compared with those before annealing.

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