Enhance the Magnetic Properties of Fe-Si-Al-Cr Flaky Particles by Annealing

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Abstract— Fe-Si-Al-Cr flaky particles were used as microwave absorbing material. Iron-Silicon-Aluminum-Chromium ingots with nominal compositions $Fe_{73}Si_{15}Al_{10}Cr_2$ were produced by alloying the pure elements in an induction melting furnace under argon atmosphere at 0.03 MPa, and then the ingots were crushed into irregular shaped particles. As-crushed alloy had been milled by planetary miller for 32 h. In order to increase the effective permeability, the material was annealed under different conditions. The morphology of the powders was characterized by scanning electron microscope (SEM). The phases of powders were estimated by X-ray diffract ometer (XRD). The microwave permeability dispersion spectra of annealed powders within a frequency range of 0.5–8 GHz was measured. The SEM results showed that particles had been successfully transformed into the flaky shape. The average diameter of flakes is about 43 µm. The average aspect ratio (width/thickness) of flakes is about 28. The XRD results showed that the DO3 ordered phase coexisted with α-Fe(Si,Al,Cr) matrix phase by annealing above 500°C. When the annealing temperature increased from room temperature to 800°C, the grain size grown from 10.4 nm to its maximum 29.8 nm and part of α-Fe(Si,Al,Cr) matrix phase translated into DO3. The imaginary part of the effective permeability increased with the annealing temperature, and decreased when the temperature was above 500°C. The resonance frequency decreased with the annealing temperature, and increased when the temperature was above 400°C. The effective permeability of the powders, obtained by 500°C annealing, had a maximum value of 6.0 at 2.5 GHz.

1. INTRODUCTION

Electromagnetic wave absorbers in the gigahertz range for mobile communications have become widely used. Soft magnetic metal powders, such as Fe-Si-Al alloysor nanocrystalline metal alloys, have attracted considerable attention for shielding against electromagnetic interference (EMI) radiation.

In order to increase the bandwidth of electromagnetic wave absorption andre duce the thickness of an absorber, high permeability values are required, especially high imaginary parts of permeability [1]. The microwave permeability of milled Fe-Si-Al flaky particles has been found to be much larger than those of unmilled particles with irregular shape [2]. It has been proven that magnetic particles with flaky shapes can effectively improve the absorption properties [3]. Tianyu Ma et al. have fabricated thin $Fe_{85.8}Si_{9.6}Al_{5.6}$ (wt.%) nanocrystalline flakes from melt-spun ribbons by a high-energy planetary ball-milling method. They found that the D03 superlattice recovered after annealing for 30 min at 300°C and over, accompanied by modest crystalline growth, causing a clear enhancement in magnetic properties [4]. It has been proven that the addition of Cr in the alloys can improve the effectively permeability of materials [5]. But they did not consider the effect of annealing on the microwave magnetic properties of Fe-Si-Al-Cr flaky particles. Therefore, in this paper, $Fe_{73}Si_{15}Al_{10}Cr_2$ precursor was prepared by ball-milled and then annealed under different temperature. The effects of annealing temperature on the microstructure and magnetic properties of the as-prepared powders were investigated.

2. EXPERIMENTAL PROCEDURES

Stoichiometric $Fe_{73}Si_{15}Al_{10}Cr_2$ compounds were prepared by induction melting under argon atmosphere, the constituting elements with a purity higher than 99.9%. The ingots were crushed into irregular shaped particles with the diameter was hundreds of microns. As-crushed alloy had been milled by planetary miller for 32 h. The as-prepared flakes were annealed in vacuum furnace at different treatment temperatures (from 300°C to 800°C) for 1 hour.

The morphology of the powders was characterized by scanning electron microscope (SEM, JEOL TSM-7600F). The phases of powders were estimated by X-ray diffractometer (XRD, SHIMADZU XRD-7000) with Cu K-radiation. Complex permittivity and complex permeability measurements, in the frequency range between 0.5 and 18 GHz, were carried out by to roidal samples (mass ratio of powders and paraffin was 4 : 1) of Ø7 mm × Ø3.04 mm × 2.5 mm using microwave vector network analyzer (Agilent 8720ET).

3. RESULTS AND DISCUSSION

After these particles have been milled for 32 h, they have been successfully transformed into the flaky shape, as shown in Figure 1. The average diameter of flakes is about 43 µm. The average aspect ratio (width/thickness) of flakes is about 28.

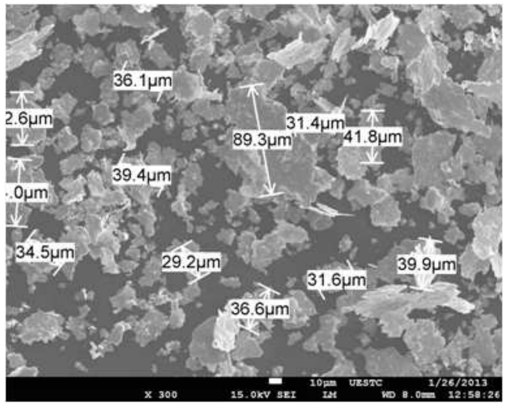

Figure 1: SEM micrographics of various powders milled for 32 h FeSiAlCr.

Figure 2 shows the XRD patterns of different samples. $Fe_{75}Si_{13}Al_{10}Cr_2$ has two kinds of crystal structures with ferromagnetic properties: The ordered DO3 (superlattice) structure and the disordered α-Fe(Si,Al,Cr) structure. Three prominent diffraction peaks in all samples are associated with the (200), (400) and (422) planes of α-Fe(Si,Al,Cr) solid-solution. Two additional peaks at angle about 28° and 31° observed in the ingot sample correspond to a DO3 ordered phase. For the α-Fe(Si,Al,Cr) phase with B. C. Clattice structure, Fe, Si, Al and Cr atoms occupy the lattice sites randomly in the way of continuous substitutional solid solutions. For the DO3 phase, Al and Si atoms dominantly occupy the body-centered site of its unit cell (i.e., superlattice), which is composed of eight B.C.C unit cells. After milling for 32 h, the diffraction peaks of DO3 phase fully disappears, XRD of original powder shows the disordered α-Fe(Si,Al,Cr) structure. The XRD results showed that the DO3 ordered phase coexisted with α-Fe(Si,Al,Cr) matrix phase by annealing above 500°C. When the annealing temperature increased from room temperature to 800°C, three prominent peaks are relatively narrow, attributing to the variation of average grain size and internal strain which can be estimated by Williamson-Hall method [6], as shown in Table 1.

Figure 3 reflects the effective permeability and permittivity of the alloy powders annealed un-

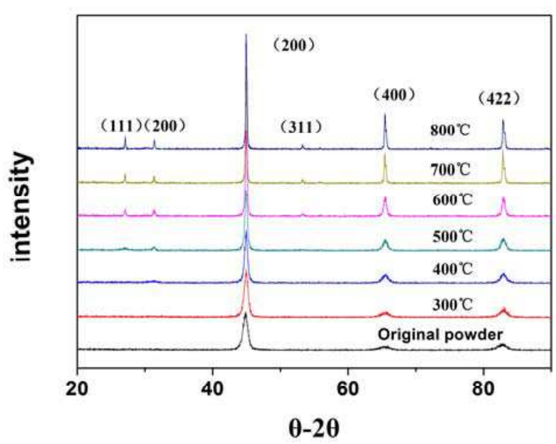

Figure 2: XRD patterns of the ingot simple, the powders milled for 32 h, and then annealed in vacuum furnace at 300°C, 400°C, 500°C, 600°C, 700°C, 800°C.

Table 1: Grain size D (nm), internal strain ε (%) with different annealed temperature.

Annealed temperature (°C)	D (nm)	ε(%)
as-milled	10.4	0.501
300	12.2	0.478
400	16.6	0.461
500	19.6	0.345
600	27.6	0.166
700	28.7	0.108
800	29.8	0.068

der different temperature. Both the real part and imaginary part decreases during the annealing temperature increased from room temperature to 800°C. After annealed, the real part of the permeability of the sample with an increasing temperature first increases, then descending. The real part of the permeability of the powders obtained by 500°C annealing has a maximum value of 9.0 at 1.05 GHz. The imaginary part of the effective permeability also presents a first increase and then decreasing trend. The imaginary part of the effective permeability increased with the annealing temperature, and decreased when the temperature was above 500°C. The effective permeability of the powders, obtained by 500°C annealing, had a maximum value of 6.0 at 2.5 GHz. First, the resonance frequency moves to the low frequency with the annealing temperature, the minimum value of resonance frequency is 1.2 GHz, obtained by 400°C annealing. Then the resonance frequency increased when the temperature was above 400°C.

For cubic crystal ($K_1 > 0$), we can get the resonance frequency f_r by the following formula: $f_r = \gamma H_{\mathit{eff}}$. H_{eff} is magnetic anisotropy field, γ is gyromagnetic ratio. Average grain size grown up from 10.4 nm to 29.8 nm, leading to an increase magnetic anisotropy H_{eff}. Internal strain has an decrease from 0.501% to 0.068%, leading to an decrease of H_{eff}. When the temperature is under 500°C, the decrease of internal strain contribute more to the change of H_{eff} than the grown up of grain size; When the teperature is above 500°C, it is opposite. As a result, f_r presents a first decrease and then increasing trend when the annealing temperature increased from room temperature to 800°C. Similar to H_{eff}, M_s the also presents a first decrease and then increasing

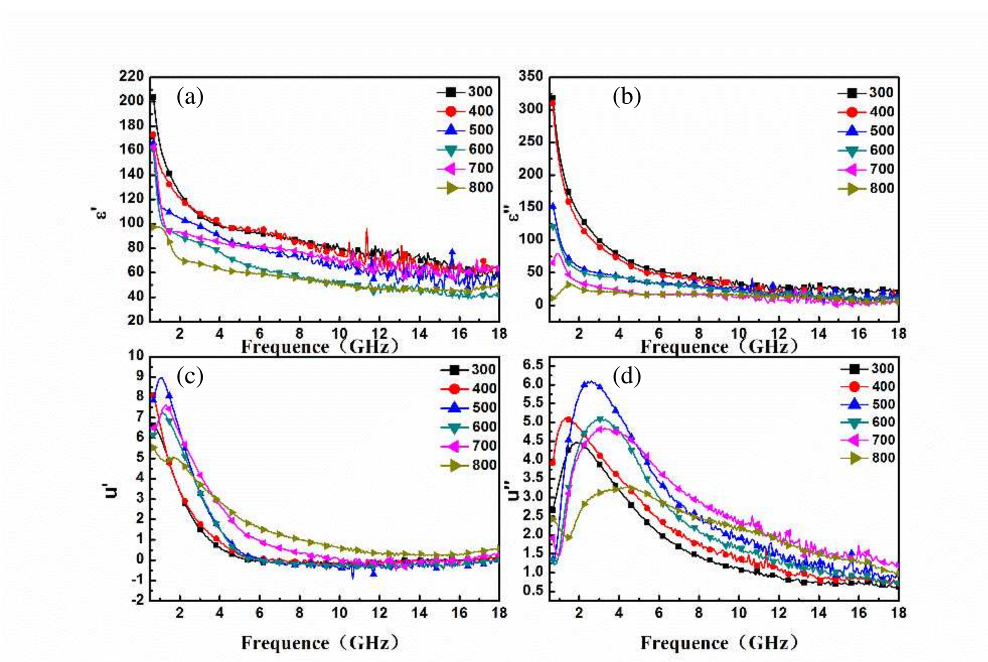

Figure 3: Effect of different components on the microwave permeability and permittivity. (a) The real part of permittivity; (b) The imaginary part of permittivity; (c) Thereal part of permeability; (d) The imaginary part of permeability.

trend.

Permeability values of traditional microwave magneticmaterials are small in quasimicrowave frequencies of gigahertz due to Snoek's limit:

$$(\mu_r - 1)f_r = 2/3\gamma M_s \tag{1}$$

Fe-Si-Al-Cr flaky particles were annealed under different temperature. The shape-dependent Snoek's law as given below indicates that higher permeability can be obtained in flaky particles:

$$(\mu_r - 1)f_r^2 = (\gamma/2\pi)^2 4\pi M_s(H_k + 4\pi M_s D_z) \tag{2}$$

where D_z is the demagnetization factor of the direction normal to the particle plane. D_z value does not change after annealing. According to the change of M_s and f_r, μ_r presents a first decrease and then increasing trend. μ' had a maximum value of 9.0 at 1 GHz, μ'' had a maximum value of 6.0 at 2.5 GHz, obtained by 500°C annealing.

4. CONCLUSION

Flake-shaped particles were annealed under different temperature. The XRD results show that there are two major phases exist: the ordered phase (DO3) and the disordered α-Fe(Si,Al,Cr) structure. When the annealing temperature increased from room temperature to 800°C, the grain size grown from 10.4 nm to its maximum 29.8 nm, the internal strain increases from 0.079% to 0.485%, part of α-Fe(Si,Al,Cr) matrix phase translated into DO3. The effective permeability of the powders, obtained by 500°C annealing, had a maximum value, $\mu' = 9.0$ at 1.2 GHz and $\mu'' = 6.0$ at 2.5 GHz.

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