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Understanding the enhanced microwave permeability of Fe–Si–Al particles by Mössbauer spectroscopy

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ABSTRACT

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. Transmission Mössbauer spectra are found significantly different for particles with different shapes. More than 8 absorption peaks have been found for the unmilled particles while there are only 6 absorption peaks for the flaky particles. The unmilled particles are found with the ordered $D0_3$ type superlattice structure. Therefore the Mössbauer spectra can be fitted with 5 sextets representing 5 different Fe-site environments. However, the flaky particles are with the disorder α-Fe(Si,Al) structure, and their Mössbauer spectra can be fitted with a model with the distribution of hyperfine magnetic field and isomer shift. It also has been revealed by Mössbauer spectroscopy that the flaky particles have stronger tendency to possess the planar magnetic anisotropy, and their average magnetic moment is found to be about $1.63\mu_B$.

1. Introduction

The ternary Fe–Si–Al alloys with the compositions of $Fe_{(88-81)}Si_{(8-11)}Al_{(4-8)}$ (wt%) are well known for their excellent soft magnetic properties, and are usually named as “Sendust” alloys. Recently, there is an increasing interest in them due to their promising applications in suppressing the unwanted electromagnetic radiations from electronic devices working in the quasi-microwave band [1–3]. In order to increase the bandwidth of electromagnetic wave absorption and reduce the thickness of an absorber, high permeability values are required, especially high imaginary parts of permeability [4]. Walser et al. have theoretically proposed that the permeability of particles can be enhanced by transforming spheres into flakes, thin films, oblate particles or wires [5]. Composites with the flaky particles as fillers have been fabricated as electromagnetic wave absorbers. It has been proven that magnetic particles with flaky shapes can effectively improve the absorption properties [6,7]. Currently, it is still necessary to understand the enhanced permeability on the atomic scale, which will be helpful to further increase high frequency permeability. Mössbauer spectroscopy is a powerful tool to study magnetic properties on the atomic scale with extremely high energy resolution. In this contribution, we present the shape effects of Fe–Si–Al particles on their microwave permeability and Mössbauer spectra using the ^{57}Co(Rh) source Mössbauer spectroscopy, and uncover the possible variation of Fe atom environments due to the high energy ball milling.

2. Experimental details

The $Fe_{84.94}Si_{9.68}Al_{5.38}$ alloy particles with irregular shapes (called sample A) were provided by a Chinese vendor. The alloy was initially prepared by melting the starting materials (Al, Fe and Si with high purities) in a hydrogen reduction furnace. The obtained ingot was then crashed and milled into particles with irregular shape. These particles were further milled into the flake shape for 30 h (called sample B) by a planetary ball miller. During the milling, some ethanol has been added as the milling dispersant. The morphologies of particles were observed by scanning electron microscopy (SEM). The x-ray diffraction (XRD) patterns were collected with Cu K_α radiation to identify the phase variations resulting from the ball milling processes. The magnetic hysteresis loops were taken to study the effects of particle shapes on the static magnetic properties. To measure the microwave permeability dispersion spectra, both samples A and B were made of toroidal shape (the inner diameter, outer diameter and the thickness are 3, 7, and 3 mm, respectively). For each sample, the weight ratio of the particles over the wax was kept as a constant of 4:1, and they have been mixed homogeneously. The complex relative permeability was measured on a vector network analyzer (Agilent 8720 ET) within a frequency range of 0.5–10 GHz. Transmission Mössbauer spectroscopy with ^{57}Co radiation source in the rhodium (Rh) matrix is employed to study the effects of

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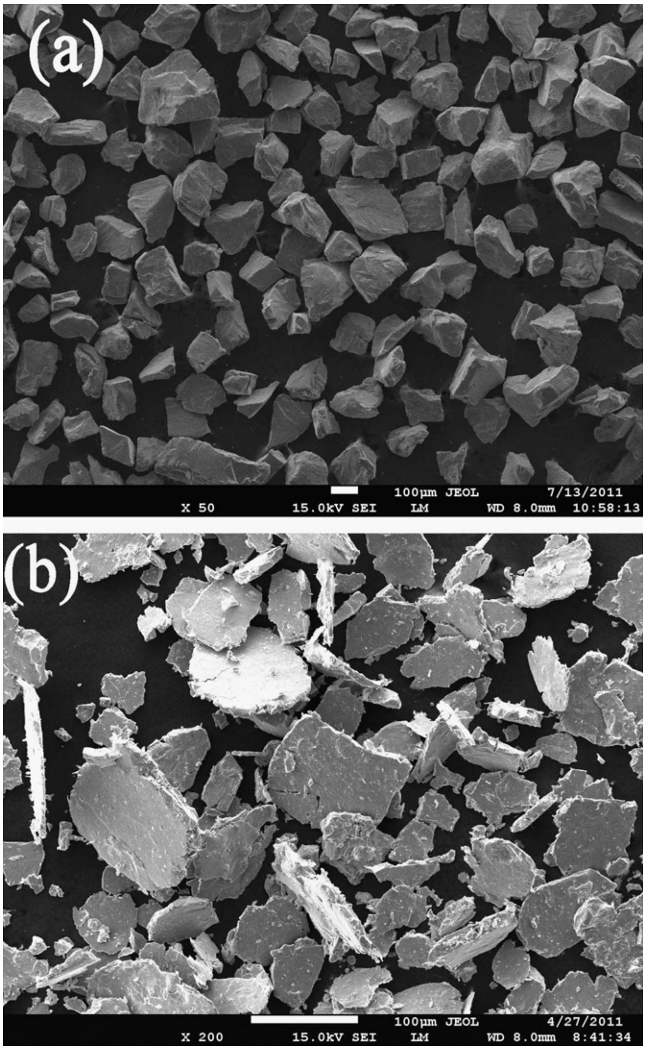

Fig. 1. SEM images of Fe–Si–Al alloy particles: (a) unmilled particles and (b) particles being milled for 30 h.

particle shapes on the γ-ray absorption spectra. The velocity of radiation source is calibrated with a standard α-Fe foil. The Mössbauer spectra have been fitted using a software called WinNormos-for-Igor®.

3. Results and discussions

The shape of unmilled Fe–Si–Al particles is irregular, as shown in Fig. 1(a). After these particles have been milled for 30 h, they have been successfully transformed into the flaky shape, see Fig. 1(b). The average thickness of flakes is about 5.16 μm. The average aspect ratio (width/thickness) of flakes is about 22. It is well known that the "Sendust" alloys have two kinds of crystal structures with ferromagnetic properties: the ordered $D0_3$ (superlattice) structure and the disordered α-Fe(Si,Al) structure. XRD patterns shown in Fig. 2 indicate that the attrition processes have changed the $D0_3$ phase into the α-Fe(Si,Al) phase. The diffraction peak around 31° in Fig. 2(b) is indicative of the formation of $D0_3$ superlattice in sample A [8,9]. For the α-Fe(Si,Al) phase with b.c.c lattice structure, Fe, Si and Al atoms occupy the lattice sites randomly in the way of continuous substitutional solid solutions. However, for the $D0_3$ 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. These different site occupancies will play critical roles in analyzing the Mössbauer spectra later.

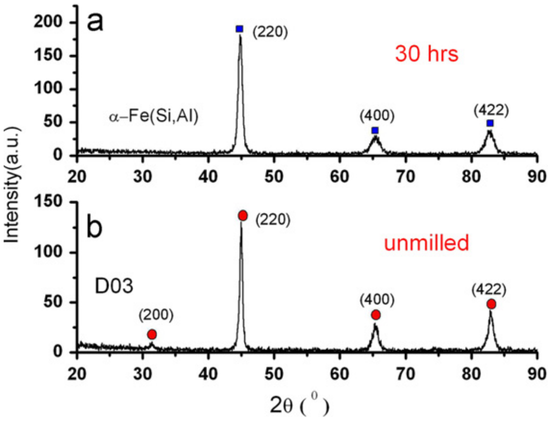

Fig. 2. XRD patterns of samples A and B.

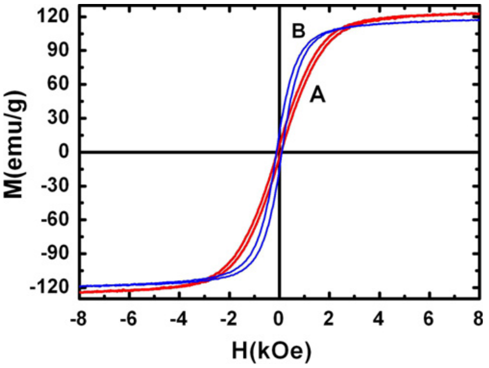

Fig. 3. Magnetic hysteresis loops of samples A and B.

The magnetic hysteresis loops of Fe–Si–Al are shown in Fig. 3 for the irregular shape particles (curve "A") and flaky particles (curve "B"). The saturation magnetization (M_s) is 124 emu/g and 120 emu/g for samples "A" and "B" respectively. The coercivity is about 72 Oe and 100 Oe for sample "A" and "B", respectively. Clearly, the unmilled particles have slightly "soft" magnetic properties. The larger coercivity of sample B could be also due to the residual internal stress arising from the high energy ball milling.

The high frequency measurements show that the particles with flaky shape exhibit significant enhancement in microwave permeability, see Fig. 4(a) and (b). At 0.5 GHz, the real part of permeability of Fe–Si–Al flakes is about 4.4. For the unmilled irregular shape particles, it is only about 1.3. Within 0.5–7 GHz, the μ' values of sample B are obviously larger than those of sample A. As for the imaginary parts of permeability (μ''), the flaky particles of Fe–Si–Al have larger values within the whole measurement frequency range, see Fig. 4(b). There are some reasons for the increased permeability. Firstly, the eddy current effect can be greatly suppressed by transforming the irregular shape particles into the flakes. Secondly, the shape-dependent Snoek's law as given below indicates that higher permeability can be obtained in flaky particles: $(\mu_s-1)f_r^2=(\gamma/2\pi)^2 4\pi M_s[(H_k+4\pi M_s D_z)]$, where D_z is the demagnetization factor of the direction normal to the particle plane. Similar results have already been found in magnetic thin films which have much more larger permeability than bulk materials due to their even larger D_z value [10]. It should be pointed out that "f_r" which appeared on the left of above equation is the intrinsic resonance frequency, not the effective resonance