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Fe/SiO_2 Nanocomposite Soft Magnetic Materials

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ABSTRACT

In an effort to explore new highly resistive soft magnetic materials, Fe/SiO_2 nanocomposite materials have been synthesized using a wet chemical reaction approach in which the precursor complex was annealed at various temperatures. The crystallographic structure, nanostructure, morphology, and magnetic properties of the synthetic Fe/SiO_2 particles were studied by x-ray diffraction, transmission electron microscopy, and magnetic measurements. The experimental results show that for this approach, the α-Fe particles are coated with amorphous silica. The progress of the reaction, the purity of Fe/SiO_2 in the synthetic powder, and the Fe particle size are highly dependent on the annealing temperature. By adjusting the annealing temperature, the particle size can be controlled from approximately 20 nm to 70 nm. For the synthetic nanopowder obtained by H_2 reduction at 400 °C, there exists a superparamagnetic behavior below room temperature; while for the nanopowders obtained by reduction at higher temperatures, the ferromagnetic behavior is dominant. Based on these studies, optimum synthesis conditions for Fe/SiO_2 nanocomposites is determined.

INTRODUCTION

The application of soft magnetic materials in AC electrical and electronic devices can be divided into two categories. One is the low power application, such as magnetic cores in inductors. In this case, the material works in its initial (linear) magnetization region; the frequency range can be from 100 Hz to 10 GHz. The other is the high power application, such as magnetic cores of transformers or chokes. In this case, the material works in a high magnetization region (over 50% of its saturation magnetization). Both applications require soft magnetic core materials which possess high resistivity, high saturation magnetization, high Curie temperature, high initial permeability, low eddy current loss, low hysteresis loss, low residual loss, and low dielectric loss. The advances in electronic equipment technology are in the direction of increasing the operating frequency, which creates a large demand for high frequency magnetic core materials.

Originally, metallic alloys possessed the best soft magnetic properties among all of the soft magnetic materials, but their extremely low resistivity (10^{-6} Ω cm) make them inapplicable for slightly elevated frequency (> 50 kHz), even in thin ribbon form. To overcome this problem, two types of highly resistive magnetic materials have been developed: ferrites and powder materials. However, ferrites possess low saturation magnetization and low Curie temperature, while the demagnetizing factor makes the permeability of powder materials very low.

Nanocomposite processing has resulted in new opportunities to develop novel magnetic materials. For instance, in a metal/insulator nanocomposite, resistivity, which is the major concern at high frequencies, can be dramatically increased, leading to significantly reduced eddy current loss. Meanwhile, the exchange coupling between neighboring magnetic nanoparticles can overcome the anisotropy and demagnetizing effects, resulting in better soft magnetic properties than conventional materials [1]. Based on this idea, nanocomposite thin films with significantly improved high frequency properties have been developed [2,3]. However, the thin film techniques cannot be used to produce bulk components. In an earlier report, a study was presented which concerned Co/SiO_2 nanocomposite high frequency soft magnetic materials. These materials were fabricated using chemical synthesis and powder processing, suitable for mass production of bulk size magnetic components [4]. In addition, the Fe/SiO_2 nanocomposite system has also been developed. Here, the structural and magnetic property studies on the synthetic Fe/SiO_2 nanoparticles are presented.

EXPERIMENTAL

Fe/SiO_2 nanocomposites were synthesized using a wet chemical solution technique [5]. Solutions of Fe- and Si-containing precursors were prepared separately in alcohol with controlled pH, reaction time, and temperature, to form precomposite precipitates in solution. The precipitated nanoparticle suspensions were dried in an oven to obtain a precomposite powder, which was then transferred to a high-temperature crucible in an environmental furnace. The precomposite powder was then thermochemically converted into a metal-ceramic Fe/SiO_2 nanocomposite powder in the presence of a reducing agent at temperatures from 400 °C to 900 °C. The nanocomposites were passivated in oil to prevent oxidation. The oil was finally washed away by an organic solvent.

Characterization of the crystal structure and particle size of the synthetic powders were carried out using x-ray diffraction (XRD) with Cu $K\alpha_1$ radiation. The nanostructure of the Fe/SiO_2 particles was also examined using high-resolution transmission electron microscopy (HRTEM) and Mossbauer effect (ME) experiments. Static magnetic properties of the synthetic $NiFe_2O_4$ nanoparticles were measured using a Quantum Design SQUID magnetometer at temperatures between 10 K and 300K.

RESULTS AND DISCUSSION

Structure

In the synthesis procedures of the Fe/SiO_2 nanoparticles, the H_2 reduction treatment of the precursor is the most critical step. In a previous paper, it was mentioned that the precursor consisted of ferrihydrite ($5Fe_2O_3 \bullet 9H_2O$) and Fe_2SiO_4 [6]. By annealing the precursor in H_2 at different temperatures, they gradually convert into bcc α-Fe. We refer the reader to Ref. [6] for a detailed discussion of the ferrihydrite-to-Fe conversion. Here, we focus on the crystal structure and morphology of these $(Fe)_v/(SiO_2)_{1-v}$ nanoparticles, where v represents volume percentage. Figure 1 shows the XRD patterns of the synthetic $(Fe)_{50}/(SiO_2)_{50}$ powder samples obtained by reducing the precursor in hydrogen at 400, 600, and 900 °C for 3 hours. For comparison, the figure also includes the XRD pattern of a bulk-size conventional Fe sample. The XRD results indicate that all of the synthetic $(Fe)_{50}/(SiO_2)_{50}$ powders have the same bcc structure. Chemical

analysis was performed for the nanoparticles as synthesized, and the content for SiO_2 is 48.7% in volume for $(Fe)_{50}/(SiO_2)_{50}$ sample. As shown in the figure, there is no trace of any SiO_2 XRD lines, implying that the SiO_2 is in an amorphous state. From the linewidth of the main peak at 44.6° the mean particle size for the Fe nanoparticles was calculated according to the Scherrer equation [6]. Our results show that the Fe particle size increases with increasing H_2 reduction temperature in general; however, other processing parameters affect the particle size as well. For example, at the same H reduction temperature of 600 °C, the resultant Fe particle size can vary between 20 nm to 70 nm.

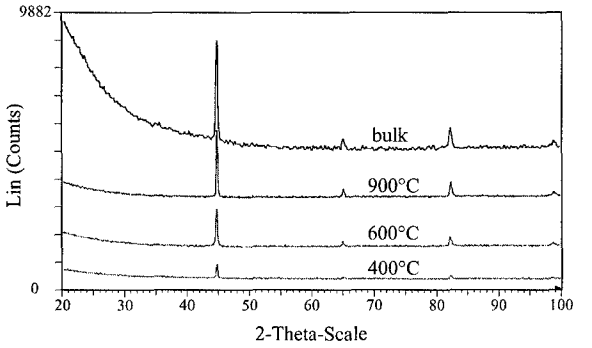

Figure 1. XRD patterns for Fe/SiO_2 nanoparticles and microsized (bulk) Fe particle.

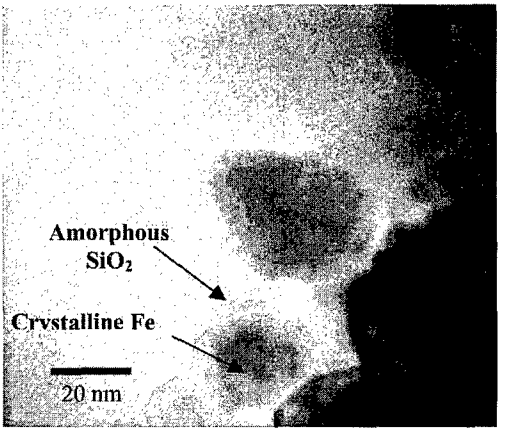

Figure 2. A typical TEM image showing the morphology of Fe/SiO_2 nanoparticles.

High-resolution transmission electron microscopy experiments were used to study the nanostructure of the synthetic Fe/SiO_2. Bright field images, electron diffraction, and lattice images were carried out. A typical HRTEM image showing the morphologies of the $Fe_{50}/(SiO_2)_{50}$ nanoparticle materials is presented in Figure 2. This image shows a nanoparticle

where the inner Fe core and the outer SiO_2 cover are distinguished. Transmission electron microscopy observations clearly show that the outer SiO_2 cover is amorphous. Another interesting result obtained from TEM is that the outer cover and the inner Fe core are in close contact all around the interface.

Static magnetic properties

Figure 3 shows typical magnetization curves measured at 10 K and 300 K for $Fe_{50}/(SiO_2)_{50}$ powder sample reduced in H_2 at 700°C. It can be seen from the figure that the magnetization curve is close to saturation in a field of 6 kOe. The saturation magnetization of the powder is obtained to be 7095 G. Figure 4 shows the saturation magnetization as a function of the H_2 reduction temperature. Starting from 400°C, reducing at an elevated temperature makes the ferrihydrite and Fe_2SiO_4 phases more completely converted to bcc α-Fe, thus the saturation magnetization increases with increasing reduction temperature.

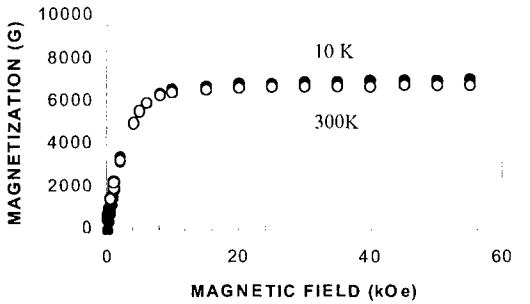

Figure 3. Magnetization curves of the 400°C reduced $Fe_{50}/(SiO_2)_{50}$ nanopowder sample measured at 10 K and 300 K.

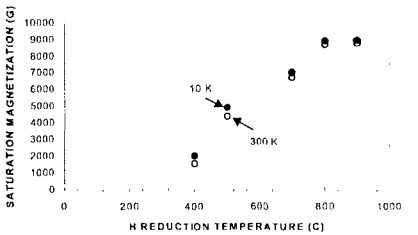

Figure 4. The variation of saturation magnetization with H_2 reduction temperature for $Fe_{50}/(SiO_2)_{50}$ nanopowder samples measured at 10 K and 300 K.

As shown in Figure 3, the magnetization curves measured at 10 K and 300 K for the 700°C annealed $Fe_{50}/(SiO_2)_{50}$ sample are very close. This is due to the high Curie temperature of α-Fe

(> 700 °C). However, for the 400°C annealed sample, its magnetization curves shown in Figure 5 are quite different; a significant decrease in saturation magnetization occurs going from 10 K to 300 K. In order to gain additional information, a low field measurement of the magnetization temperature dependence was carried out. In this measurement, the sample was cooled from room temperature to 4.2 K in a zero magnetic field (ZFC), and then a 100 Oe field was applied. The magnetization variation was measured with increasing temperature up to 360 K, followed by decreasing temperature (FC) to 10 K. The results are plotted in Figure 6. The curve shows a superparamagnetic relaxation behavior with a rather broad blocking temperature distribution.

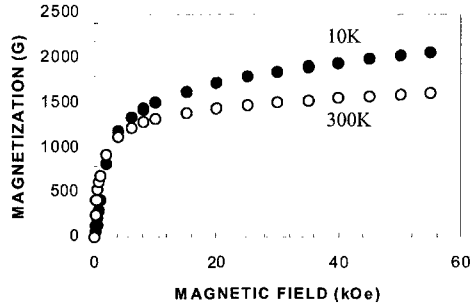

Figure 5. Magnetization curves of the 700°C reduced $Fe_{50}/(SiO_2)_{50}$ nanopowder sample measured at 10 K and 300 K.

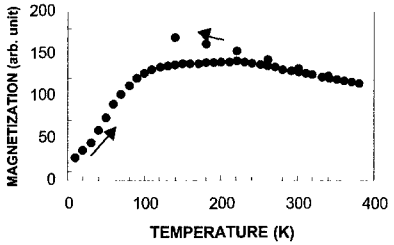

Figure 6. Magnetization as a function of temperature for the $Fe_{50}/(SiO_2)_{50}$ nanopowder reduced in H_2 at 400 °C. The arrows indicate the direction of the temperature variation.

CONCLUSION

A wet chemical approach is developed for synthesizing Fe/SiO_2 nanoparticles. By reducing the precursor with a controlled H_2 atmosphere, temperature and time period, the conversion into α-Fe can be completed and the particle size can be controlled. In this Fe/SiO_2 nanocomposite, the SiO_2 is in an amorphous state which plays as a network coating the Fe nanoparticles.

ACKNOWLEDGMENTS

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