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Original Research Paper

Intergranular insulated Fe/SiO_2 soft magnetic composite for decreased core loss

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ABSTRACT

An intergranular insulated Fe/SiO_2 soft magnetic composite has been designed to reduce the core loss and improve the magnetic properties of cores by a modified Stöber method combined with the spark plasma sintering. Most of conductive Fe particles could be coated by insulated SiO_2 using the modified Stöber method. In the spark plasma sintering, the high compact and intergranular insulated cores were obtained quickly. The Fe/SiO_2 composite core displayed much higher electrical resistivity, lower core loss, better frequency stability of permeability and higher quality factor than that of the raw Fe core without insulated SiO_2 layers. The model introduced here provides a promising method to reduce the core loss and improve the magnetic properties for soft magnetic composite materials, which can reach larger energy conversion efficiency for electric–magnetic switching device.

1. Introduction

Due to their ubiquity, magnetic materials have already played a significant role in improving the efficiency and performance of devices in electric power generation, conditioning, conversion, transportation, and other energy-use sectors of the economy [1]. Improved and innovated soft magnetic composites, in which ferromagnetic powders are separated by insulated layers, have received considerable attention recently and have a substantial impact on electric–magnetic switching device for their high electrical resistivity, three-dimensional isotropic ferromagnetic behavior, very low eddy current loss and relatively low total core loss at medium and high frequencies [2–6]. Generally, about 9% of electrical energy is lost during electromagnetic transmission and distribution due to eddy current loss [1]. So the reduction of electrical losses needs to be addressed. In maximizing the energy efficiency for power conversion, a key challenge is to prepare high-performance soft magnetic composite with lower core losses and better magnetic properties.

Pittini-Yamada et al. [7] put forward a percolation model for soft magnetic composite containing magnetic and insulated phases (as shown in Fig. 1, type 1). In this case, in order to enhance the electrical resistivity as well as keep good magnetic properties, the insulated phase is applied for soft magnetic composite. But the insulated phases are still too much in such conventional soft magnetic composite, which cause a serious decline in their magnetic properties (such as permeability). Furthermore, the organic insulated phases like phosphating-resin cannot withstand heat treatment at a temperature above 500 °C in conventional soft magnetic composite [8]. Thus the density and compactness of conventional soft magnetic composite are remains to be improved, so as to improve their mechanical and magnetic properties. To decrease the core loss, improve the compactness and magnetic properties, a new model is designed for soft magnetic composite in this paper (as shown in Fig. 1, type 2). In this model, the magnetic phases are separated effectively by a homogeneous insulated layer and an intergranular insulated soft magnetic composite with high compactness, excellent magnetic properties and low core loss is obtained.

In order to build such structure in soft magnetic composite, a modified Stöber process [9] was used to generate silica coatings on the surface of Fe particles by using a silane-coupling agent as modification additives [10]. Then the spark plasma sintering

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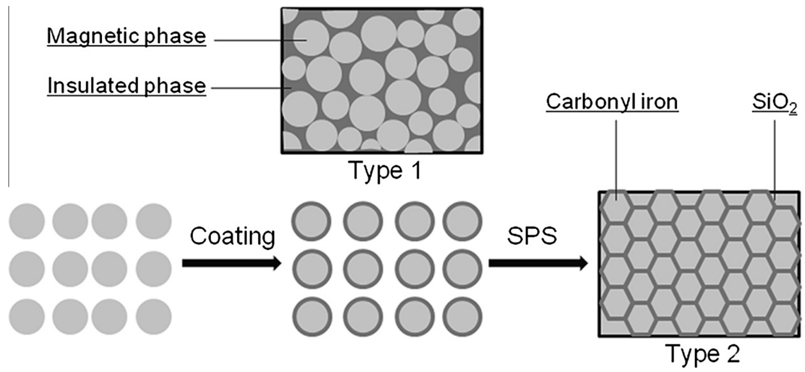

Fig. 1. The schematic graph of soft magnetic composite materials containing magnetic and insulated phases. Type 1 corresponds to the conventional model for soft magnetic composite. Type 2 corresponds to the model of soft magnetic composite in this work.

(SPS) technique [11–15] is employed for consolidating Fe/SiO_2 composite compacts. The insulated SiO_2 layers not only eliminate the electrical conducting path to suppress the eddy current inside cores effectively, but also confine the eddy current inside the particles. Thus the lower core loss can be reached.

2. Experimental

2.1. Materials and reagents

The starting material is spherical or quasi-spherical carbonyl iron powder with an average particle size of <10 μm. Tetraethyl orthosilicate (TEOS, 98 wt.%), 3-triethoxysilypropylamine (APTES, 99 wt.%), aqueous ammonia (28 wt.%, lab grade) and absolute ethanol were purchased from Tianli Chemical Reagent Company, Tianjin, China. Distilled water (H_2O) was prepared by Automatic Double Pure Water Distillatory (SZ-93A, China) in laboratory. All chemicals used were of analytical grade and were used as received without further purification.

2.2. Preparation of Fe/SiO_2 core–shell particles

In a typical synthetic procedure, 50 g carbonyl iron powders were dispersed in 500 ml absolute ethanol by mechanical stirring for 10 min, and then 3.0 g APTES and 10 ml deionized water were added to the mixture under mechanical stirring for 1 h at 50 °C. And 15 ml TEOS and 2 ml aqueous ammonia (25 wt.%) were injected simultaneously into the mixture by injection pump (LSP01-1A) at a constant speed at 60 °C for 20 h afterward. And then the mixture was stirred for another 2 h. Finally, the suspension was filtered, washed with absolute ethanol several times, and dried at 60 °C for 24 h.

2.3. Preparation of Fe/SiO_2 composite core

The Fe/SiO_2 core–shell particles obtained were sintered at 1050 °C under a uniaxial pressure of 30 MPa into a cylindrical shaped with 20.3 mm outer diameter, 12.7 mm inner diameter and 6.35 mm height by SPS process. The heating rate was fixed at 50 °C min^{-1}. In all the cases, the holding time at the sintering temperature was 10 min. Finally, the Fe/SiO_2 composite core was obtained after removing residual stress by annealing at 800 °C for 2 h under pure argon atmosphere. To investigate the effect of SiO_2 insulated layer on the electrical and magnetic properties, the raw Fe core without SiO_2 insulated layer were also prepared under the same conditions.

2.4. Characterization

X-Ray diffraction (XRD) were analyzed on an X-Pert Philips diffractometer with Cu Kα radiation (λ = 1.5418 Å). The morphology and local chemical homogeneity of the as-prepared samples were observed by scanning electron microscopy (SEM) (Nova400) equipped with an energy dispersive X-ray spectrometer (EDS) (IE350PentaFETX-3). Fourier transform infrared spectra (FTIR) were recorded in the wave number range from 400 cm^{-1} to 4000 cm^{-1} at room temperature. The resistivity was obtained by a four probes method. Core loss at high-frequency was measured by an auto testing system for magnetic materials (MATS-2010SA). The quality factor (Q) and inductance (L) were measured under a constant voltage of 0.3 V in a frequency range from 50 Hz to 1000 kHz by impedance Analyzer (HIOKI 3532-50 LCR HiTESTER). The effective permeability (μ_{eff}) was calculated using the following formula (1) [16,17]:

$$\mu_{eff} = \frac{\bar{l}L}{\mu_0 N^2 A} \tag{1}$$

where $\bar{l}$ represents the average length of the magnetic circuit, N stands for the number of turns of inductor, A refers to the effective area of magnetic circuit, μ_0 means the permeability of vacuum and it is $4\pi \times 10^{-7}$ H m^{-1}.

3. Results and discussion

3.1. Microstructure of Fe/SiO_2 core–shell particles

Fig. 2(a)–(d) shows the SEM images of Fe particles before and after chemical coating process. Most of Fe microspheres have a diameter of less than 10 μm and their surface are smooth. After chemical coating process (Fig. 2(c) and (d)), their surface became rough and was coated with something. The magnified image of selected region in Fig. 2(c) indicates clearly that the Fe particles are coated by some nano-scaled particles as illustrated in Fig. 2(d).

The chemical compositions of Fe particles before and after chemical coating are characterized by EDS as depicted in Fig. 3 (a). Only elements Fe and O exist on the surface of Fe particles before chemical coating process, but another Si signal is also present after chemical coating process. The existence of trace amounts of element O before chemical coating process is owing to the high oxidation activity of iron. The increase of O concentration from 1.83 wt.% to 8.77 wt.% and the appearance of Si signal should be attributed to the chemical coating, which implies that the coating layer may be SiO_2.