Properties of FeSiAl-based soft magnetic composites with AlN/Al₂O₃ and hybrid phosphate–silane insulation coatings

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Abstract
FeSiAl soft magnetic composites (SMCs), which are widely used in electromagnetic applications, can be described as FeSiAl particles surrounded by electrical insulating layers. In general, good insulation is required to minimize eddy currents within SMCs in high frequency applications. In this work, FeSiAl-based SMCs with uniform single insulation layer of AlN/Al₂O₃ were prepared by high-temperature selective nitridation and oxidation. For comparison, SMCs with double layers of phosphate-resin or phosphate-silane were produced by traditional chemical process. Their magnetic properties were systematically studied. The results revealed that the core insulated by AlN/Al₂O₃ with high electrical resistivity had much better stability in the real part of permeability \( \mu_0 \) than double layer insulated cores with respect to frequency in the range of 100 kHz to 28 MHz. In addition, the FeSiAl-AlN/Al₂O₃ core had higher quality factor Q at higher frequency than other cores. The investigation of the effects of annealing process up to 600 °C on the cores indicated that the magnetic properties of FeSiAl-AlN/Al₂O₃ cores were strongly improved in terms of \( \mu_0 \), Q, total core loss \( P_{tot} \) and coercivity \( H_c \) mainly on account of the hysteresis loss reduction by stress relaxation and defects elimination. By contrast, annealing treatment had less positive effects on the properties of the cores with double insulating layer. In addition, the microstructure, composition and thermal stability of the AlN/Al₂O₃ insulated particles were characterized by Scanning Electron Microscopy, Transmission Electron Microscopy, X-Ray Diffraction, X-ray Photoelectron Spectroscopy and Derivative Thermogravimetric Analysis, respectively.

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1. Introduction

FeSiAl (also known as sendust) soft magnetic composites (SMCs) are composed of FeSiAl magnetic particles coated with electrical insulating material. They are widely used in electromagnetic applications due to their excellent properties such as three-dimensional magnetic isotropy, high magnetic permeability, low coercivity, high resistivity and relatively low core loss [1–9]. Among them, core loss, which consists of hysteresis loss, eddy current loss and residual loss, is the most important parameter for high-frequency applications in pulse transformers, electric motors, sensors and filter inductors, etc. [3–7,10–16] The hysteresis loss is largely affected by the internal stress introduced by compacting process. In general, higher pressure leads to higher dislocation density and imperfections, giving rise to higher hysteresis loss. Correspondingly, high temperature heat treatment is an effective way to minimize the deleterious effect of compaction process on the magnetic properties of SMCs [7,17–19]. For another, it is well acknowledged that the eddy current loss of SMCs consists of intraparticle and interparticle parts. The former is typical for the magnetic powders constituted SMCs with well insulation layers while the later occurs for cores with non-insulated or incomplete insulated powders [11]. The combination of both kinds of eddy current loss usually occurs. The paths of inter-particle eddy currents are concentric curves lying in the cross-section perpendicular to the magnetic induction while that for the intra-particle eddy currents are lying inside each particle [11]. It can be derived that the inter-particle eddy current loss can be minimized by effective insulating treatment of the magnetic powders surfaces [4–6,11,20]. Therefore, it is essential to prepare homogeneous insulating layers with good thermal stability up to 800 °C on the FeSiAl magnetic particles.

Generally speaking, there are two basic types of insulating coatings applied for SMCs, organic and inorganic coatings. Organic
coatings, including epoxy resin and phenolic resin, normally exhibit poor thermal stability and will decompose or even burn during annealing process, leading to the deterioration of the magnetic properties of the SMCs [19,21]. Among various inorganic coatings, phosphates produced by using orthophosphoric acid have attracted much attention for their simple preparation procedures, high electrical resistivity and good adhesiveness to the magnetic particles [7,22–25]. However, they have limitations in terms of thermal stability. To solve this, SMCs with double insulating layers consisting of phosphate and silicate or silicone resin were developed [17,19,26]. It was found that such SMCs present better magnetic properties after annealing treatment. In addition, oxides such as MgO [20,27], SiO2 [5,28–31], Al2O3 [4,6,18,32,33] and MnO2 [34] produced using wet chemistry methods were also employed as insulation coatings for SMCs. There is no doubt that all of them could withstand high temperature annealing without any deterioration. However, the complicated preparation technology and weak adhesion to the magnetic powders could not be ignored.

In this paper, a novel kind of FeSiAl SMCs with AlN and Al2O3 insulation layers that exhibit excellent thermal stability and high electrical resistivity were produced by simple high-temperature selective nitridation and oxidation method. For comparison, traditional FeSiAl SMCs with phosphate-resin and phosphate-silane double insulation coatings were also prepared. The effects of annealing treatment at different temperatures on their magnetic properties were investigated systematically.

2. Experiment

2.1. Preparation of the FeSiAl SMCs

The raw FeSiAl powders with the chemical composition of 85 wt % Fe–9 wt % Si–6 wt % Al were provided by Yahoo Materials & Technology Co., Ltd. in China. The powders were prepared by means of water atomization, and the mean powder size was approximately 30 µm. The selective nitridation of the surfaces of FeSiAl powders were achieved in a traditional tube furnace after exposing at 1100 °C for 30 min under high purity nitrogen atmosphere, where the partial pressure of nitrogen and oxygen were about 1 atm and 10⁻⁶ atm, respectively. According to the thermodynamic calculation results of the free energy changes of the potential generated in oxides and nitriles, it is expected that the formation of Al2O3 and AlN as a result of selective oxidation and nitridation of Al are most likely to happen under such circumstance, while the iron oxides are less likely to be formed [10]. To be more specific, the non-standard Gibbs free energy changes for the formation of Al2O3, SiO2 and Fe2O3 by oxidation under such circumstance are −663.12, −286.15 and −155.77, respectively, while the ones for the formation of AlN, Si3N4 and Fe3N by nitridation are −336.982, −145.611 and −125.014, respectively [10]. Before the nitridation process, the FeSiAl powders were placed in the furnace tube, and nitrogen with a flow rate to prevent further uncontrollable reaction of FeSiAl powders.

For comparison purposes, the other two different kinds of FeSiAl powders with double insulating layers were prepared. The common step for the two corresponding surface-modified methods was traditional phosphating. The specific process was as follows: the FeSiAl powders were degreased in acetone and then dried, followed by stirring in the solution of orthophosphoric acid diluted in ethanol with three different concentrations of 0.010 g/ml, 0.015 g/ml, and 0.020 g/ml at room temperature for 30 min for passivation; then the powders were rinsed in acetone and dried. The concentration of FeSiAl powders in the solution was 2.5 g/ml. Following the phosphate treatment, the passivated FeSiAl powders prepared with each of the three concentrations of orthophosphoric acid were separated into two parts, respectively. One part was coated with 0.7 wt% epoxy resin, and the other coated with 0.7 wt% silane coupling agent. The specific process was as follows: the epoxy resin and silane coupling agent were dissolved in acetone, respectively; then each of the separated parts of FeSiAl powders were stirred in the solutions, respectively. The mixing process was continuously carried out until the solvent was evaporated totally.

All of the different kinds of treated FeSiAl powders mentioned above were then compacted into toroidal cores with outer diameter of 18 mm and inner diameter of 8 mm at 1200 MPa by use of zinc stearate as a die wall lubricant. The powder cores with double insulation layers of phosphate and resin were then cured at 120 °C for 1 h, and the ones with single insulation layer of nitriles and double insulation layers of phosphate and silicate were then subjected to annealing in the tube furnace at 400 °C, 500 °C, 600 °C and 700 °C for 1 h using argon as protective atmosphere.

2.2. Characterization

The phase identifications of the samples were examined by X-ray powder diffraction (XRD, DX–2000 Model using Cu-Kα radiation). The microstructures of the samples were characterized by scanning electron microscopy (SEM, Hitachi S–4800) attached with energy dispersive spectroscopy (EDS) and transmission electron microscopy (TEM, PHILIPS CM200). The components of the products were analyzed by X-ray photoelectron spectroscopy (XPS, Kratos XSAM800). The thermal stability of the FeSiAl powders before and after nitridation treatment was analyzed by derivative thermogravimetric analysis (DTG, Shimadzu DTG–60). The complex permeability and total power loss of the FeSiAl SMCs were measured using LCR meter (Agilent 4285) in the frequency range of 100 kHz–28 MHz and B–H analyzer (SY–8232), respectively. The hysteresis loops of the samples were measured using vibrating sample magnetometer (VSM, Lakeshore 7410).

3. Results and discussion

In Fig. 1(a)–(d), the SEM images and EDS results of the FeSiAl powders before and after treatment under high purity nitrogen atmosphere at 1100 °C for 30 min are presented. It is shown that the surfaces of the FeSiAl powders without coating treatment are smooth, while the treated ones are covered with a layer of relatively homogeneous flocculent materials. Based on the results of EDS measurement, it can be observed that compared to the raw FeSiAl powders, the intensity of the Al peak is much higher while that of Fe peaks are much lower for the treated powders. In addition, it should be noted that the EDS measurement could not provide the accurate conditions of O and N elements. In Fig. 1(e), the TEM image of the treated FeSiAl powder is presented. It can be seen that the FeSiAl particle is tightly covered by a complete and uniform shell composed of fine floccule and the interface between the particle and the shell is quite distinct. Considering the analyzed depth of EDS, it can be deduced from the above analysis that the treated FeSiAl powders have been coated with complete and homogeneous layers mainly composed of Al element.

Shown in Fig. 2(a) are the XRD patterns for the FeSiAl powders before and after high temperature treatment under nitrogen atmosphere. Based on the analysis of XRD measurement results, it is demonstrated that the raw FeSiAl powders have two phases of Al0.3Fe3Si0.7 and Fe5Si. And compared with raw powders, besides the peaks of Al0.3Fe3Si0.7 and Fe5Si, the treated ones have three
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