Processing and properties of soda lime silica glass glass insulation soft magnetic composite

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Fe/SiO_2-Na_2O-CaO glass soft magnetic composite (SMC) was fabricated by hot-pressing with low compaction pressure (80 MPa). Sol–gel method was used to create SiO_2-Na_2O-CaO glass insulation on the surface of pure iron powders. Iron powder was well coated with glass substances, which have some micro-cracks. No significant deformation or grain size difference of iron particles was observed in Fe/SiO_2-Na_2O-CaO glass SMC in different directions. Continuous distribution of Si and Na elements between iron particles was revealed by EDS. Interface between iron particles and amorphous SiO_2-Na_2O-CaO glass layer was straight, clean and well bonded. Static magnetic properties of Fe/SiO_2-Na_2O-CaO glass SMC could be improved by annealing treatment, while the electrical resistivity (ρ) decreased slightly after annealing treatment. The eddy current loss increased more rapidly than the hysteresis loss with frequency for Fe/SiO_2-Na_2O-CaO glass SMC in present work. It should be due to the low electrical resistivity of composites. Therefore, the coating and fabrication parameters should be further optimized in future research.

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1. Introduction

Recently, soft magnetic composites (SMC) are widely investigated as potential materials for application in electric, computer and communication fields [1-3]. They possess several advantages over traditional soft magnetic materials including (i) isotropic magnetic property, (ii) relatively low total core loss at medium to high frequencies and (iii) reduction in size and weight [3,4]. Moreover, SMC three-dimensional magnetic devices with net shape and tight tolerance can be produced by various powder metallurgy compaction techniques, which satisfy the precision and miniaturization for special components [5].

Many researchers have tried to improve the magnetic properties performance of SMC, by selecting a suitable material and applying a suitable coating method [6-14]. The phosphating magnetic powders and organic resin adhesives (such as polypropylene [6], epoxy resins [7-9] and phenolic resins [10]) were often chosen to prepare SMC under high pressure at room temperature followed by annealing [6-13]. However, the phosphating layer has low temperature toleration [15] and cannot be heat treated at high temperature, which leads to the difficulty of releasing residual stress in preparing process. Inorganics and organics mixture coatings (such as phosphating-resin coatings [13], SiO_2-silicone resin coatings [14] and SiO_2-B_2O_3-PbO-phenolic resins [10]) and inorganics coatings with high thermal stability (such as SiO_2 [16], MgO [17] and CuO coatings [18]) were researched in recent years. Inorganics and organics mixture coatings cannot overcome the difficulty of high temperature heat treatment fundamentally. Though MgO, SiO_2 and CuO coatings are stable at high temperature, the coefficient of thermal expansion (CTE) of MgO, SiO_2 and CuO are quite different from iron, which will result in high internal stress on account of environment temperature change or self heating and affect the magnetic properties of SMC.

Unfortunately, the difference of coefficient of thermal expansion (CTE) between iron and these inorganic insulation layers will induce thermal stress during the serving process of SMC, which decreases the magnetic properties. Therefore, we explored the possibility to prepare Fe/SiO_2-Na_2O-CaO glass SMC by hot-pressing with low pressure (80 MPa) for decreasing residual stress. Adjusting the component of glass to match CTE of magnetic matrix will reduce the residual stress in hot-pressing process and the thermal stress in serving process.

Sol–gel method was used firstly to create SiO_2-Na_2O-CaO insulation on the surface of pure iron powders. The microstructure and magnetic properties of Fe/SiO_2-Na_2O-CaO glass SMC were investigated.

2. Experimental

Pure iron powders (ASC 100.29, Höganäs

(China).Ltd.) with average particle size of 88 μm were used as the raw material [19]. SiO_2 sols were prepared from tetraethyl orthosilicate (TEOS). The fundamental hydrolysation process of TEOS can be expressed using the following reactions [22]:

$$(C_2H_5O)_4Si + 4H_2O \rightarrow Si(OH)_4 + 4C_2H_5OH \quad (1)$$

$$Si(OH)_4 \rightarrow SiO_2 + 2H_2O \quad (2)$$

The detailed experimental procedures to get the SiO_2-Na_2O-CaO-coated iron particles are as follows: (i) iron particles were ultrasonic washed successively with acetone and ethanol two times to clean the surface of iron particles; (ii) ethanol and TEOS with volume ratio of 1:1 were mixed firstly and then mixed with the cleaned iron particles by a high-speed electrical stirrer with a speed of 800 rpm for 1 h at room temperature. (iii) $NaNO_3$ and $Ca(NO_3)_2 \cdot 4H_2O$ with weight ratio of 5.35:1 were mixed with deionized water, ethanol and hydrochloric acid to obtain a solution (pH = 1). (iv) the solution was added into the mixture of SiO_2 and iron particles, and then the suspension was stirred for 3 h at room temperature; (v) after stirring, the suspension was dried at 60 ºC for 3 h to obtain SiO_2-Na_2O-CaO-coated iron particles. The coated iron particles were then hot pressed at 750 ºC under a pressure of 80 MPa in Ar atmosphere (0.5 atm) for 30 min in graphite die. For comparison of annealing treatment effect, the obtained composites were annealed at 600 ºC for 1 h under vacuum (10^{-1}Pa).

The morphology of Fe/SiO_2-Na_2O-CaO glass SMC was revealed by ZEISS 40MAT optical microscope. The microstructure of the coated particles and Fe/SiO_2-Na_2O-CaO glass SMC was observed by a scanning electron microscope (SEM) with energy dispersive spectrometer (EDS). Further observation was carried out on a Philips CM-12 transmission electron microscope (TEM) with an accelerated voltage of 100~120 kV. The static magnetic properties of Fe/SiO_2-Na_2O-CaO glass SMC were measured at NIM-2000S DC autohysteresis loop tracer with field amplitude of 5000 A/m (National Institute of Metrology, China). The dynamic magnetic property was measured by an Agilent 4294A precision impedance analyzer and an ac B-H loop tracer (National Institute of Metrology, China). Magnetic field intensity was 0.5 T with frequency of 50Hz, 100Hz and 400 Hz, respectively. The specimen used for magnetic testing is ring-shape with external diameter 25mm, internal diameter 17mm and thickness 3mm.

3. Results and discussions

3.1 Microstructure

The representative SEM images of iron powders before and after coating treatment are shown in Fig. 1a and b, respectively. It is clear that the iron powders were coated with a layer, which presented some cracks. It is well known that existence of cracks is a common problem in sol-gel derived coatings, particularly at thickness above 1 μm, because of their inherent rigidity and high volumetric shrinkage during processing [20].

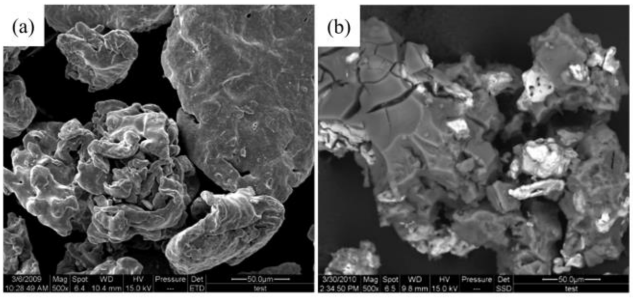

Fig. 1. Representative microstructure of iron powders (a) without and (b) with SiO_2-Na_2O-CaO coating.

Microstructure of Fe/SiO_2-Na_2O-CaO glass SMC at compressing surface and side surface are shown in Fig. 2a and b, respectively. No significant difference in particle size and deformation of iron powders was observed, which is different to SMC obtained by cold-pressing with high pressure [6-13]. The Fe/SiO_2-Na_2O-CaO glass SMC in present work were fabricated by at 700 ºC. The insulator has been softening and could be deformed under pressure. Moreover, the pressure adopted in present work (80 MPa) was relative lower than that of their process (usually above 800 MPa) [1-8]. Therefore, the iron particles showed little deformation after preparation. Thus, properties of Fe/SiO_2-Na_2O-CaO glass SMC in present work would be isotropic.

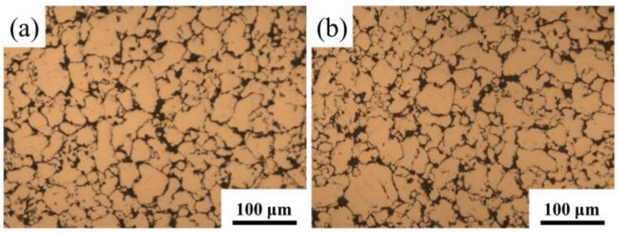

Fig. 2. Representative microstructure of Fe/SiO_2-Na_2O-CaO composites in (a) transverse and (b) longitudinal directions.

EDS analysis reveals the distribution of Si and Na elements between iron particles (Fig. 3). It should be noted that extensive O element was also detected on the polished surface of iron particles due to oxidation. Therefore, the distribution of O element was not shown in present work. Moreover, the distribution of Ca element was not clear due to its low content (the presence of Ca element was proved in Fig. 4). Therefore, the distribution of Ca element was also not shown in Fig. 3. However, it could be concluded that the iron particles have been are separated from each other by a continuous insulating layer (Fig. 3c and d). Furthermore, TEM observation revealed that interface between iron particles and amorphous layer (Fig. 4b) was well bonded (Fig. 4a). The interface was straight and clean, and no interfacial reaction product was observed (Fig. 4a). The presence of O, Si, Na and Ca in the amorphous layer was detected by EDS (Fig. 4c).

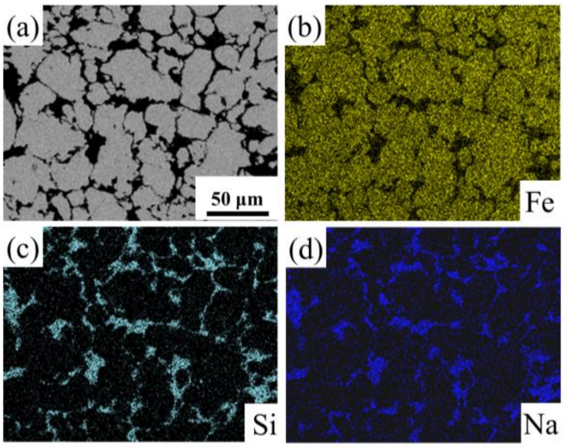

Fig. 3. Morphology and element distribution in Fe/SiO_2-Na_2O-CaO composites, (a) microstructure, (b) Fe, (c) Si, (d) Na.

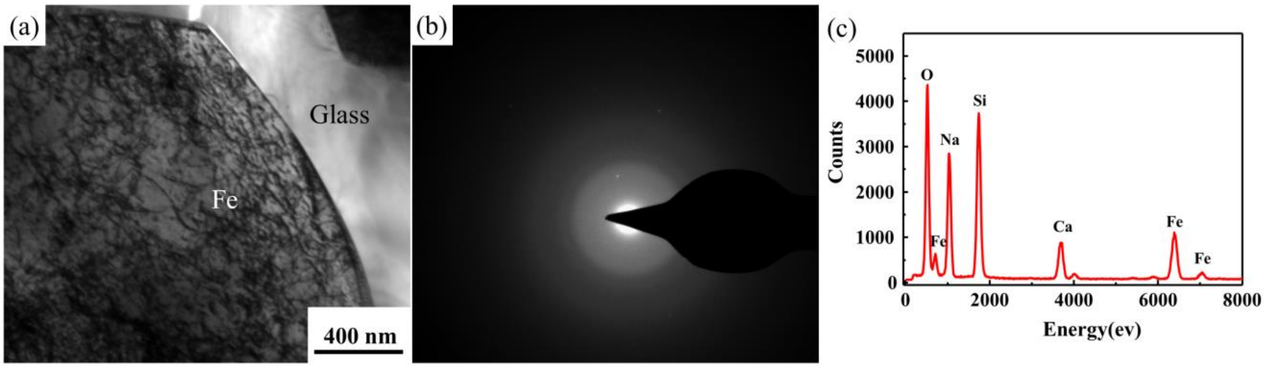

Fig. 4. TEM observation of Fe/SiO_2-Na_2O-CaO composites. (a) interface, (b) electron diffraction pattern and (c) EDS analysis of amorphous SiO_2-Na_2O-CaO.

3.2 Magnetic properties

Static magnetic properties of Fe/SiO_2-Na_2O-CaO glass SMC before and after annealing treatment are listed in Table 1. The initial permeability (μi) and maximum permeability (μm) were 105 and 423, respectively. Magnetic properties of Fe/SiO_2-Na_2O-CaO glass SMC could be improved by annealing treatment due to release of residual stress. It should be noted that the electrical resistivity decreased slightly after annealing treatment. Generally, electrical resistivity of SMC would decrease greatly due to decomposition of phosphate-coating at high temperature [15]. However, electrical resistivity of SMC with inorganic coatings would change little because of high thermal stability of inorganic coating.

Low core loss is required for soft magnetic materials for application in alternating electric field, such as motors and power transformers. The contribution to the total loss of soft magnets in ac applications is classified into three categories including hysteresis loss Ph, eddy current loss Pe and residual loss Pr. Furthermore, the residual loss can be ignored at a frequency below 1000 Hz. Therefore, the core loss could be expressed as following Eq. (1) [22]:

$$W = nB^{1.6} f + eB^2 f^2 \tag{1}$$

where W is the core loss, n is hysteresis loss coefficient, B is the maximum induction, f is the frequency and e is the eddy current loss coefficient.

The core loss (W) versus frequency (f) of Fe/SiO_2-Na_2O-CaO glass SMC at the induction (Bmax) level of 0.5 T is shown in Fig. 5. Bai Yang et al [16] reported that the core loss of SMC had a linear relationship with frequency in low frequency filed. However, the core loss of SMC in this paper was not proportional to frequency. Then, hysteresis loss coefficient (n) and eddy current loss coefficient (e) could be obtained by fitting the core loss (P) versus frequency (f). The hysteresis loss coefficient (n) and eddy current loss coefficient (e) of Fe/SiO_2-Na_2O-CaO glass SMC were 0.25 and 1.70×10-3, respectively. Afterward, hysteresis loss and eddy current loss could be separated, as shown in Fig. 5. The eddy current loss increased more rapidly than the hysteresis loss with frequency for Fe/SiO_2-Na_2O-CaO glass SMC in present work. Because the eddy current loss is in inverse proportion to the electrical resistivity. In order to decrease the eddy current loss at high frequency, it is necessary to increase the electrical resistivity of SMC [23]. Therefore, the coating and fabrication parameters should be further optimized in future research. Moreover, SiO_2-Na_2O-CaO glass coating exhibits insulating characteristics, which could decrease the eddy current loss of composites. Therefore, if well coated, the eddy current loss inside the iron particles may be responsible for the eddy current loss of Fe/SiO_2-Na_2O-CaO glass SMC. As suggested by Yang et al.[16-18], metallic powders with smaller size or higher intrinsic electrical resistivity should be chosen to decrease eddy current loss.

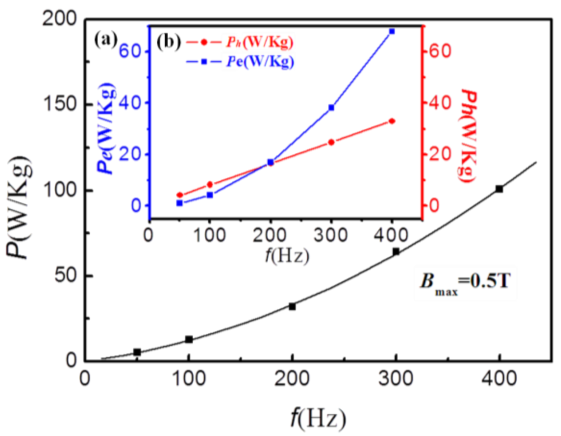

Fig. 5. Dynamic magnetic properties of Fe/SiO_2-Na_2O-CaO composites at the induction (B_{max}) level of 0.5 T. (a) The core loss (P) versus frequency (f) curve and (b) Hysteresis loss and eddy current loss of Fe/SiO_2-Na_2O-CaO composites at different frequencies.

4. Conclusions

Fe/SiO_2-Na_2O-CaO glass SMC was fabricated by hot-pressing with low compaction pressure (80 MPa). Sol–gel method was used to create SiO_2-Na_2O-CaO glass insulation on the surface of pure iron powders. Iron powder was coated with glass substances, which have some micro-cracks. No significant deformation or grain size difference of iron particles was observed in Fe/SiO_2-Na_2O-CaO glass SMC in transverse and longitudinal directions. Continuous distribution of Si and Na elements between iron particles was revealed by EDS. Interface between iron particles and amorphous SiO_2-Na_2O-CaO glass layer was well bonded. The interface was straight and clean, and no interfacial reaction product was observed. Static magnetic properties of Fe/SiO_2-Na_2O-CaO glass SMC could be improved by annealing treatment due to release of residual stress, while the electrical resistivity (ρ) decreased slightly after annealing treatment. The eddy current loss increased more rapidly than the hysteresis loss with frequency for Fe/SiO_2-Na_2O-CaO glass SMC in present work. It should be due to the low electrical resistivity of composites. Therefore, the coating and fabrication parameters should be further optimized in future research.

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