Preparation of soft magnetic composite from Fe-6.9wt%Si by different heat treatment strategies.

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Abstract. Present study investigated the effect of isothermal heat treatment strategies between 800 °C and 1150 °C on the magnetic properties of toroidal samples made from Fe-6.9wt%Si powder. The samples were prepared by classical powder metallurgy method since the classical sheet forming methods no longer work with the high silicon content. Our results presented here are part of a series of comparative experiments where we study the effectiveness of the insulating layers created during and before the compacting of soft magnetic composites (SMCs). Our goal was to create a soft magnetic composite made of ferromagnetic and inorganic insulating material with a frequency limit already in the megahertz range and a Snoek limit of few gigahertz. In the case of samples made from Fe-6.9wt%Si powder, the computed tomography results showed that significant porosity is to be expected after pressing. Its positive effect occurred during the heat treatment in the atmospheric agent, where silicon is precipitated and deposited on the surface of the particle. This coating is an electrically insulating layer at the grain boundaries. Depending on the heat treatment strategy, 1 or 2 ferromagnetic phases were observed. The frequency limit approached the target values, but due to the low value of static permeability, the Snoek limit did not reach the gigahertz range. However, there is a significant improvement in magnetic properties compared to the heat-treated samples in a protective gas.

1. Introduction

According to today’s demands of high efficient, energy-saving power electronics (motors, transformers), the conversion and transfer of electromagnetic energy are possible only with the use of advanced soft-magnetic materials because they have high saturation magnetization and low magnetic loss [1]. They also have unique magnetic properties, including stable magnetic permeability at higher frequencies and magnetic anisotropy. Fe-Si is a typical type of soft magnetic alloys that has excellent soft magnetic properties but still exhibits a drastic increase in eddy current loss when excitation frequency is above 400 Hz [2]. Soft magnetic composites (SMCs) consist of a core-shell structure where the core is made up of a ferromagnetic particle and an electrically insulating layer that completely covers it. They are typically manufactured using classical powder metallurgy techniques, but recently, the 3D printed parts are beginning to appear. The main steps in classical powder metallurgy production are powder production by atomization or milling, powder pressing and final heat treatment. In order to reduce the stress anisotropy, which is caused by the high forces awaking during pressing, various heat treatment methods are used. During 3D printing, the amount of heat transmitted by the laser generates significant residual stress in the structure, which can also be eliminated by heat treatment.
In this study, the most widely used magnetic material in the industrial application, Fe-Si alloy was used, within it, Fe-6.9wt%Si with outstanding properties [3]. Differently, to the usual procedure where the coating with insulating layer precedes the compaction, our aim is to create an oxide insulating layer around the ferromagnetic particles by heat treatment after the powder compression. The advantage of this inorganic coating is that it remains stable at higher temperature ranges as opposed to organic insulators. This is needed both for special applications and for miniaturization of power electronic equipment. This can be done by increasing the frequency limit of the iron core without loss of performance. In addition, if we can minimize dissipation losses, we can improve the energy balance of the unit by either eliminating cooling or reducing performance. Expressed in terms of the energy stored in the coil (\(L\) – inductance, \(I_{\text{max}}\) – maximum current flowing in the coil) and the magnetic field (where \(\mu_0\) is vacuum permeability), we obtain the ratio of effective permeability (\(\mu_{\text{eff}}\)) to volume \((V)\), which shows that maximal permeability and volume reduction give maximum saturation induction \((B_{\text{max}})\).

\[
\frac{\mu_{\text{eff}}}{V} = \frac{B_{\text{max}}^2}{\mu_0 \cdot I_{\text{max}}^2 \cdot L}
\]

We also aimed to achieve some gigahertz values of Snoek limit, which is a number for the characterization of soft magnetic composites. In this case, the complex permeability spectrum was obtained by multiplying the static permeability and the frequency limit. This value includes the physical characteristics that are important to our development.

2. Material and Methods

2.1. Material
Iron-silicon alloys are one of the most prominent known soft magnetic materials in the fields of power electronics, telecommunications, military industry, and vehicle manufacturing. When the alloy reached 6.5-6.9wt%Si content (high silicon electrical steel) it has excellent soft magnetic properties such as high permeability, low magnetic loss and near-zero magnetostriction. Above 3wt% Si, classical rolling technologies no longer work because the plastic deformation of the material is close to zero at room temperature [4]. In this case, the Fe-6.9wt%Si bulk material was produced by a home built cold-crucible induction melting equipment. The ingot of about 20 grams was powdered by grinding with a SPEX 8000 M ball mill machine. The resulting wide-size distribution of powder was separated on a sieve shaker. We were used to the smaller powder particles than 74 \(\mu\)m to the final sample preparation. After the production of powder raw material, the morphology and chemical composition of the particles were investigated. Scanning electron microscopy (SEM) images of Fe-6,9wt%Si powder are illustrated in Figure 1.
2.2. Methods
Toroidal samples (D = ~12 mm, d = ~7 mm; h = ~2.5 mm) were prepared in two steps. In the first step, the powder was compressed using a conventional hydraulic press at 1.34 GPa. In the second step, after the pressure was released, the samples were subjected to oxidation heat treatment in a chamber furnace. This procedure was always started in a heated furnace, maintaining the isotherm conditions. Heat treatment temperatures were 800 °C; 900 °C; 1000 °C; 1100 °C; 1150 °C. The annealing was repeated in the air atmosphere and in the argon shielding gas. We also examined the effect of the duration of heat treatment on the magnetic properties of the SMC. Table 1 summarizes the different heat treatment strategies.

| Heat treatment time | 800 °C | 900 °C | 1000 °C | 1100 °C | 1150 °C |
|--------------------|--------|--------|---------|---------|---------|
| atmosphere         | 3 h air| 3 h air| 3 h air | 3 h air | 1 h air  |
|                    |        |        |         |         | 3 h Ar   |
|                    |        |        |         |         | 3 h air  |

Heat treatment at 800 °C and 900 °C for 3 hours were performed in an air atmosphere, however, the samples did not sinter properly due to the relatively low temperature and were broken during handling.

Among the magnetic properties, we analyzed the complex permeability spectrum in the frequency range of 0.1 – 30 000 kHz, from which the product of the static permeability and the frequency limit gives the so-called Snoek limit. The Snoek limit can be considered as a figure of merit for comparing the SMCs from a magnetic point of view. Saturation magnetization was not part of our current measurement series.
3. Results and discussion

Scanning electron microscopy was used to verify the effect of heat treatment strategies. The particle morphology, coating, composition and the changes of composition were also examined. A core-shell structure was obtained which core is Si-poor Fe. However, silicon and oxygen-rich coating appears on the grain boundaries and acts as an electrically insulating material. This silicon diffuses from the pre-alloyed powder particles to the surface of the grains (Fig. 2, 2. point). In addition to the silica layer on the porous sites and on the surface of the toroidal sample, a significant volume fraction of iron oxide coating is also observed. A sample (1100 °C, 3h) which was prepared by the same conditions, except the heat treatment atmosphere, it was argon, did not show any precipitation or composition change.

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![Image](image_url)

**Figure 2.** Scanning electron microscopy images of compacted and heat treated Fe-6.9wt%Si sample (t = 1 h, T = 1150 °C on air). Point 1 indicates the core and point 2 indicates the location of the point analysis of the shell structure. The figure on the right shows the elemental mapping results.

The magnetic permeability spectra of toroidal samples were measured using the Agilent RLC meter type 4294 A measuring the series equivalent inductance ($L_s$) and resistance ($R_s$) data. The different exciting field amplitudes current in a wide frequency range between 100 Hz and 30 MHz [5]. The sample treated in argon shielding gas had a relatively high static permeability ($\mu_s = 954$), but it’s limit frequency was only $f_{lim} = 1$ kHz. The Snoek limit obtained by multiplying these values is only 954 kHz since SMC had only one component, which had not electrically insulating layer that separates the particles.

![Image](image_url)

**Figure 3.** The heat treatment temperature (left) and time-dependence (right) of complex permeability spectra.
On the other hand, SEM images of air-annealed samples showed a composite of at least 2 components. In this case, it is clear from the permeability spectra in Figure 3, that the temperature values of the heat treatment strategies have no effect on the frequency limit. The reason for this is that in all cases a sufficient size of the insulation layer is formed. However, the static permeability values decreased from the previous 954 value to 20-55 as a result of heat treatment in air. This is presumably due to the broadening of the B-H curve. Due to the appearance of insulating layers, which resulted in a drastic reduction in iron loss, at the same time increased the internal demagnetization (N) factor \(\mu_{\text{eff}} = \frac{1}{\mu_0 N}\), causing a significant decrease in effective permeability.

![Figure 4. Snoek limit evolution as a function of heat treatment temperature and period of time in air atmosphere](image)

Figure 4 shows the evolution of the Snoek limit in each case. In contrast to the 954 kHz sample treated with argon, the best sample treated with air was found to be 1.1 GHz. It should be mentioned that at 1150 °C, two magnetic “phases” can be observed in our sample after 1h, 3h, and 5h heat treatment. This is well illustrated by the local maxima of the imaginary part of the permeability spectrum. The 1.1 GHz Snoek limit came from the second higher limit frequency. If these values are not taken into account, the sample heat-treated at 1000 °C for up to 3h proved to be the best (Snoek limit = 0.74 GHz). It is also clear from this index that the heat treatment in the air has a positive effect on the magnetic properties of the samples.

4. Conclusions

After these experiments, it turned out that self-sustaining toroidal samples can be sintered by heat treating in the air at 1150 °C. The silicon content of the particles is reduced and a silicon oxide insulating coating is formed at the interface of the particles. It can be converted in situ to soft magnetic composites. Developing a uniform and thin coating and controlling the Si content of the particles is a rather complex task. SEM images revealed that the internal oxidation at the grain boundaries was not complete and consequently, there were regions in the sample where the powder particles were not electrically insulated from each other. This heterogeneity causes the presence of two magnetic “phases” with different insulations and different demagnetizing factors and different frequency limits. The samples treated at 1150 °C proved to be the best soft magnetic iron cores based on their Snoek limit value. With respect to the frequency limit, the temperature of the heat treatment has no significant effect, in each case approximately a value of 25 MHz was obtained. By increasing the duration of the heat treatment (see Fig. 3 right), the ratio of the first magnetic “phase” with a lower limit frequency is significantly reduced. Even with a further heat-treated sample, this magnetic phase can be completely eliminated.
In the future new ways of internal oxidation will be experimented under flowing $O_2+N_2$ and $O_2+Ar$ and $O_2+CO_2$ gas mixtures preventing the “burning” of the samples during the long duration heat treatment necessary for homogeneous internal oxidation of each powder particles. In further experiments, we will sinter a pre-oxidized Fe-Si powder, the oxide layer of which is chemically formed under controlled conditions by the Stöber method [6]. New types of alloys and insulating materials will be introduced in the series of experiments, which will be compared with 3D printed toroidal samples.

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