PREPARATION AND PROPERTIES OF SOFT MAGNETIC COMPOSITES BASED ON CHEMICALLY TREATED IRON PARTICLES

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1. Introduction

Soft magnetic composites (SMC) form a group of materials that are appropriate for various AC and DC applications, such as transformers and electro motors where they are used as core with three-dimensional isotropic ferromagnetic behaviour. They are also used in electromagnetic circuits, sensors and electromagnetic control devices [1]. SMC can be described as ferromagnetic powder particles coated by insulating layer, since the ferrous powder requires sufficient electrical resistance to prevent eddy current loss at higher frequencies by preventing metal on metal contact points. This is the reason why the coating has to evenly and coherently cover the entire particle [2]. Thus treated powder is then submitted to compressing process which leads to formation of compact. SMCs are further known for they relatively low total core losses at medium and high frequencies, low coercivity and enhanced magnetic permeability. Common phenomenon is the high hysteresis losses at low frequencies resulting from the high internal stress generated during the compaction which leads to increased density of defects, mostly dislocations. These defects are obstacles that hinder the movement of magnetic domain walls and that leads to increased hysteresis loss and coercivity. To eliminate this effect, heat treatment at high temperature or hot temperature compacting is recommended [3, 4]. Based on this fact, the magnetic properties of such composites are significantly influenced by the nature of the insulating layer. Researchers are trying to improve the magnetic properties of SMC by focusing on the selection of suitable materials for coating and also on suitable coating methods. SMCs made of iron particles coated by organic resins were developed for low core losses applications [5, 6]. Unfortunately as mentioned above, due to the high residual stress generated during the compression, SMCs have to be annealed at high temperature. For iron, the recommended temperature of annealing is over 600 °C, which is a problem, due the organic coatings starting to decompose at temperatures above 500 °C [7, 8]. Another alternative is a hybrid, organic-inorganic coating (such as silicone resin with SiO_2) but similarly like pure organic coatings, they also can´t withstand higher temperatures [8]. This is a reason why is nowadays a trend of developing new inorganic coatings, for example Al_2O_3 [9], CuO [10], $FePO_4$ [11]. Inorganic oxides, such as SiO_2 can withstand high temperatures, so that makes them predestinated for high temperature compacting.

This paper focuses on two selected coating methods on pure iron particles by inorganic substances. By the Stöber method [12] the SiO_2 coating was created, and by common phosphating treatment [13] the $FePO_4$ coating was prepared. Coated particles were then further examined by scanning electron microscopy (SEM) with the energy dispersive X-ray analyzer (EDX) and were compared with each other. For more precise verification of coating process effectiveness by SiO_2 was those compacted via high temperature pressing and the electrical resistivity was measured.

2. Experimental

a) Stöber method

The ABC 100.30 pure iron powder which was provided by Höganäs AB Sweden and sieved to particle size of <63 μm was chosen as a ferromagnetic material. For Stöber coating method we used tetraethyl ortosilicate (TEOS, 98 wt.%), ammonium hydroxide (NH_4OH), isopropyl alcohol and deionized water as received without further purification to provide SiO_2 coating. 500 mg Fe powder was added to solution mixture of 4 ml deionized water and 20 ml isopropyl alcohol. Then for adjusting the pH value 0.5 ml NH_4OH was added. Subsequently 2 ml TEOS was added into mixture dropwise. To determine the optimal length of the process, the Fe particles were subject to synthesis for 5 and 17 hours. The coated powder was hot compacted into a cylindrical die at 400 °C to get a ring shaped compact with a diameter of 24 mm. The compaction of powders was conducted with a commercially purchased die wall lubricant Loctite 8191.

b) Phosphating method

Phosphating is commonly used for passivation of metals (especially iron) for instance in automotive industry. For coating we used an acetone as a solvent, orthophosphoric acid as a phosphating agent and above mentioned pure Fe powder as a substrate. Before the process, the powder was not treated because acetone has degreasing character. According to literature [13,14] the optimal concentration of othophosphoric acid is 0.01 g per 1 ml acetone and 2.5 g Fe powder per 1 ml acetone. The solution was then stirred for 1 hour in 250 ml glass cell. At the liquid and solid state interface, two processes are present. One is the anodic dissolution of Fe powder followed by creation of Fe^{2+} cations and cathodic reduction of hydrogen. The presence of Fe^{2+} cations and PO_4^{3-} anions near the surface in sufficient concentration allows to formation of protective coating in form of $FePO_4$. Chemical formula of coating creation:

$$3Fe + 2H_3PO_4 \rightarrow Fe_3(PO_4)_2 + 3H_2 \quad (1)$$

After drying, the treated Fe changed a colour to characteristic blue, which is a sign that $FePO_4$ coating was formed on a surface.

The coated particles were then embedded in a special resin. After grinding and polishing, the samples were consequently investigated by SEM. To confirm the presence of Si phase EDX analysis was performed.

3. Results and discussion

The SEM examination of SiO_2 coated Fe particles after 5 hours of synthesis, are shown in Fig. 1a, b). After examination of the coated Fe we can see, that the created layer of SiO_2 is discontinuous and does not have a uniform thickness, Fig. 1c). It is because the SiO_2 spherical nanoparticles are formed first and during the process they decompose and create a layer on Fe surface. Obviously, 5 hours of synthesis is not enough to complete conversion of spherical particles into layer. In Fig. 1, b) it can be seen that in the formed layer between two iron particles are still present incompletely transformed SiO_2 particles. In Fig. 1d), is a detailed look on all three phases, from left: SiO_2 nanoparticles, SiO_2 coating and iron. In Fig. 1 e, f) the EDX analysis of both nanoparticles and layer, which proved that they consist from Si and O is shown.

The SEM examinations of Fe particles coated by SiO_2 after 17 hours of synthesis (Fig. 2 a, b) revealed, that the coating is more continuous than after 5 hours, but with still some incompletely transformed nanoparticles. Rarely there was found some spherical SiO_2 nanoparticles on the surface of Fe powder, but most of them transformed to layer. The EDX analysis, Fig. 2 c) proved the presence of Si and O in the layer.

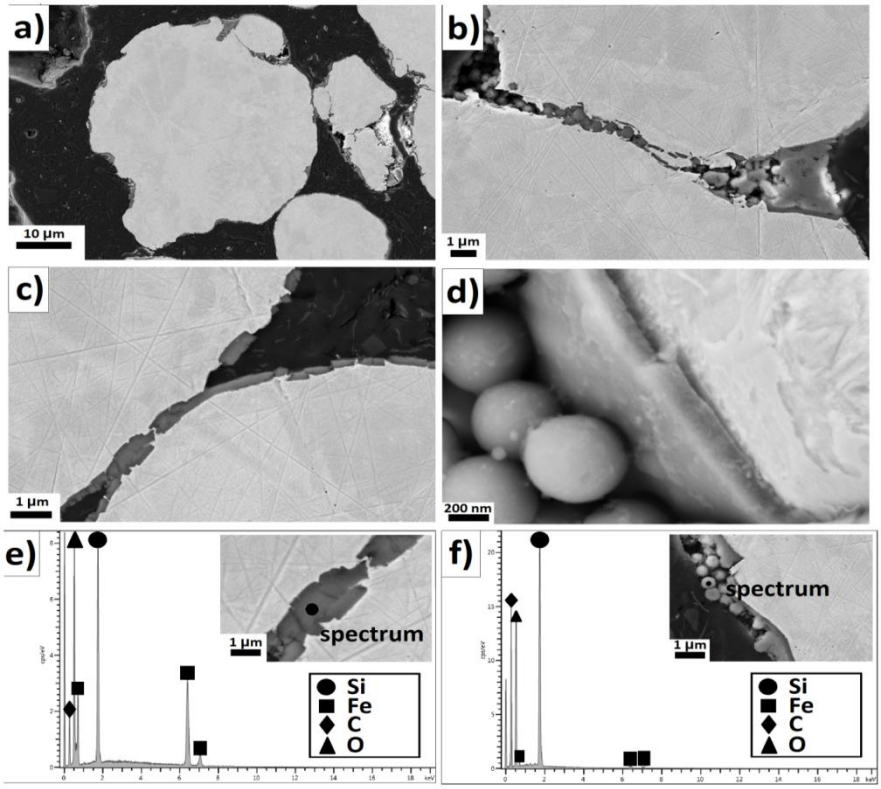

Fig.1: a) cross section view of coated Fe particles after 5 hours of synthesis (bright phase) embedded in resin (dark phase), b) and c) detailed views on formed coating between two particles which is visibly discontinuous, d) detailed look on two visible phases located on the surface of iron, SiO_2 spherical nanoparticles and SiO_2 layer, e) EDX analysis from SiO_2 nanoparticle formed on a surface of Fe grain, f)EDX analysis of coating formed on the interface of two particle.

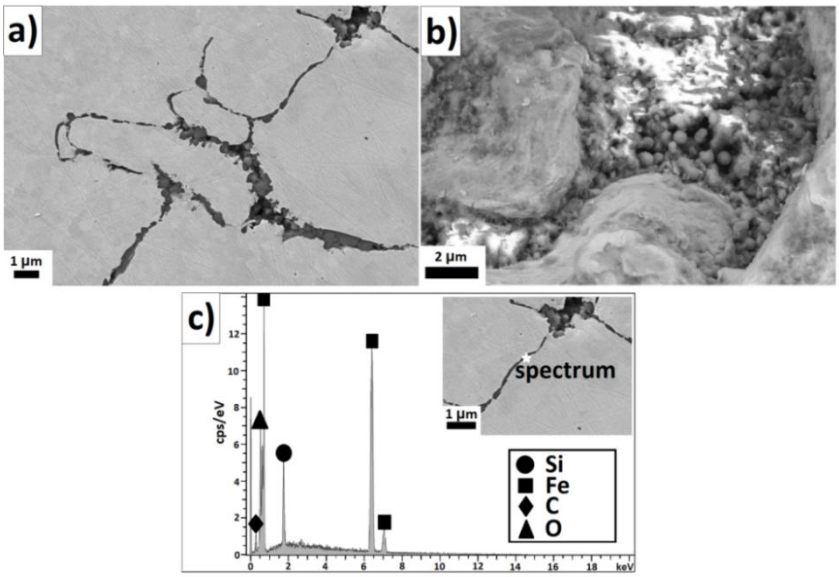

Fig.2: a) cross-section view of coated Fe particles (bright phase) after 17 hours of synthesis, with visible coating layer (dark phase) between the particles, b) SEM image of Fe particles (surface view) with SiO_2 nanoparticles, rarely spherical shape, c)EDX analysis of coating formed on the interface of two particle.

After compacting the Fe/SiO_2 composite, the specific electrical resistivity was measured on the ring shaped sample by four point method. The resistivity of sample was 2.6×10^{-7} Ω·m, which is one order higher than pure iron. This suggests that the Fe particles were insufficiently cowered by the coating, thus conductive points occurred between them.

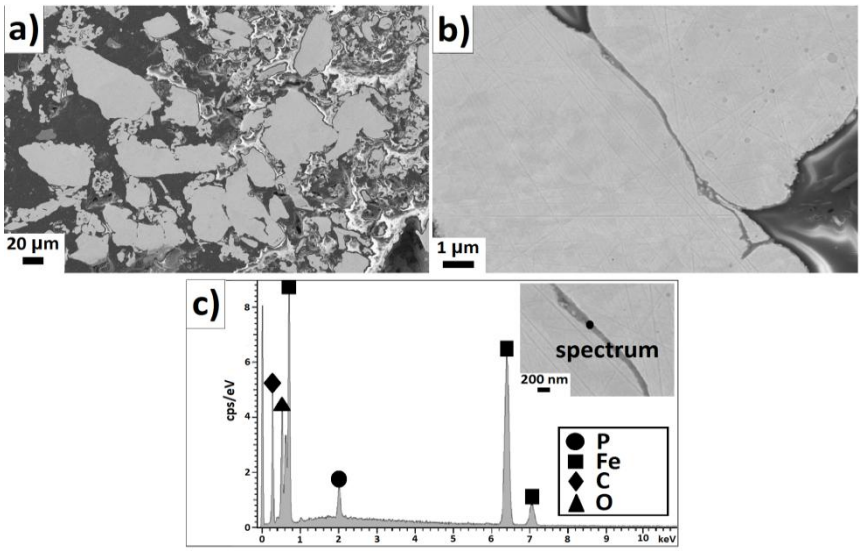

Fig.1:*a) cross section view of coated Fe particles (bright phase) embedded in resin (dark phase), b) detailed look on a interface between two particles with visible coating between them, c) EDX analysis of formed coating proved the presence of phosphorus.*

The SEM examination of $FePO_4$ treated Fe particles are shown in Fig. 3. It can be seen, that on the interface of two particles a coating is formed thus the metal on metal contact is prevented. EDX analysis confirmed that the coating mostly consists from Fe, P and O. Thus we can conclude that the formed coating is $FePO_4$, which is in good accordance with references [13, 14].

4. Conclusion

Two types of composite powders coated by two different inorganic compounds, namely SiO_2 and $FePO_4$ were compared. The Fe/SiO_2 composite was prepared by Stöber method and the $Fe/FePO_4$ by phosphating. Prepared samples were characterized by SEM and EDX analysis by four point method. Although the production of $Fe/FePO_4$ composite powder is simple and low cost and the formed layer is consistent and homogenous, this material is not suitable for hot pressing or annealing at high temperatures. On the contrary, the SiO_2 layer on Fe particles shows to be appropriate for high temperature pressing and to be promising candidate for high frequency applications.

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