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Preparation and Characterization of Fe/SiO_2 Powder Composites Using Impregnation Method

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Fe/SiO_2 powder composite materials based on irregularly and/or spherically shaped iron powder particles with an addition of SiO_2 nanopowder were prepared in two ways, (i) by mixing the Fe/SiO_2 powder with 1.0 wt.% of Shellac dissolved in ethanol and (ii) by vacuum/pressure impregnation of low-temperature sintered Fe/SiO_2 components with shellac dissolved in ethanol and with thermoplast SL450. SiO_2 was implemented either as nanopowder or by sol-gel coating. Vacuum/pressure impregnation (VPI) of pre-sintered samples was performed in a steel container. The influence of iron particle shape and processing conditions on the electro-insulating layer was microscopically evaluated and correlated with the values of the electrical resistivity and coercivity. It has been found that the continuity, distribution and thickness of insulating phase is strongly controlled by the shape of iron particles. Using the VPI procedure, the irregular surface of iron particles may cause discontinuities of insulating layer, while the spherical iron particles are well covered with continuous evenly distributed insulating layer.

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1. Introduction

One of the modern trends of applying powder metallurgy (PM) products, are the soft magnetic composites (SMCs) suitable for many soft magnetic applications, e.g. [1], mostly composed of pure iron powder particles insulated from each other by organic or inorganic material, which insulates and binds ferromagnetic particles and produces a high electrical resistivity. Different thermosets are applied as organic insulation, e.g. [2], while as inorganic insulation mostly the $FePO_4$, MgO and SiO_2 are applied, e.g. [3-6]. This paper deals with the effect of the shape of iron particles on continuity and distribution of insulating layer in the Fe/SiO_2 composites prepared by VPI with shellac and/or SL450 thermoset, as well as by mixing the Fe/SiO_2 powder with shellac. Microscopical observations were correlated with electrical resistivity and coercivity obtained.

2. Experimental

The commercial Ancorsteel 1000C iron powder (Hoeganaes Corp., USA) in as-received state with irregularly shaped particles and in as-milled/annealed state (Pallmann mill; 11000 rpm; 710 °C / 15 min) with spherical particles was used. As electro-insulating material the shellac dissolved in ethanol (14 g/100 ml) and SL450 thermoset were applied. SiO_2 was implemented as a nanopowder (fumed Silica, Sigma-Aldrich) using dry mixing in Turbula mixer. Fe/SiO_2 powder was cold pressed into cylindrical samples $\Phi 10\times 5$ mm^3, with density of 6.9-7.2 g·cm^{-3}, and subsequently sintered at 850 °C / 15 min / N_2-10% H_2. The VPI was performed using a vacuum of $\sim 10^{-2}$ kPa and pressure of ~ 500 kPa both acting for 15 min. The formulation and processing conditions are given in Table I. Microscopic observation was done using light and SE microscopy (Olympus GX71, Jeol-JSM-7000F+EDX INCA analyser). The measurement of coercivity was performed using the Foerster Koerzimat HCJ 1.097. Electrical resistivity was measured using the Van der Pauw method [7].

TABLE I

Formulation and processing of composites.

Sign.	Formulation	Fe shape, size	Preparation
1M	Fe+s	irregular 30-160 μm	mixed with 1wt.% shellac* compacted, cured at 100 °C / 20 min / air
2M	Fe+0.4%SiO_2+s		
1VPI	Fe/s		compacted pre-sintered VPI with shellac* cured at 100 °C / 20 min / air
2VPI	Fe+0.4%SiO_2/s		
3VPI	Fe+0.4%SiO_2/s	spherical 100-160 μm	
4VPI	Fe+0.4%SiO_2/S		as 3VPI, with SL450, cured at 350 °C / 1 h / air

s – shellac, S – SL450, *shellac dissolved in ethanol.

3. Results and discussion

Comparing data in Table II for mixed composites 1M and 2M, both based on irregular iron particles, a small positive effect of SiO_2 nanopowder on electrical resistivity is evident, however the difference in relatively high coercivity is low. The electrical resistivity of the 1VPI and 2VPI composites showed significant decrease, while the coercivity is about three times lower than that of the

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mixed variants. This is a consequence of metallic necks formation during pre-sintering stage in places of "protrusions" on surface of irregular iron particles, Fig. 1a,b. The microstructure consists of iron particle agglomerates. SiO_2 nanoparticles in composites 2M as well as in 2VPI tend to locate preferentially in concave areas of irregular iron particle surfaces and therefore they do not fulfill the expected role of effective "spacers" between adjacent iron particles.

TABLE II

Density, electrical resistivity and coercivity.

Sign.	Density ($g\cdot cm^{-3}$)	El. resistivity ($\mu\Omega\cdot m$)	Coercivity (A/m)
1M	7.02	1760	408
2M	6.87	3900	437
1VPI	7.20	0.119	133
2VPI	7.10	0.365	177
3VPI	7.03	776	178
4VPI	6.93	19500	174

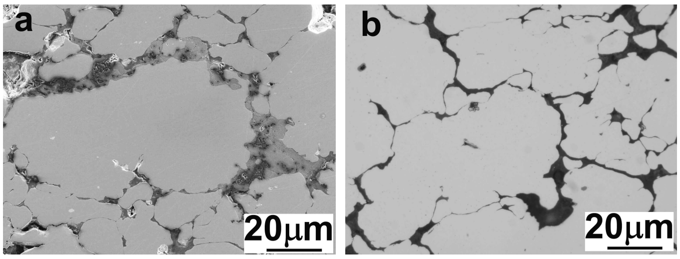

Fig. 1. Microstructure of composite 2M (a) and 2VPI (b).

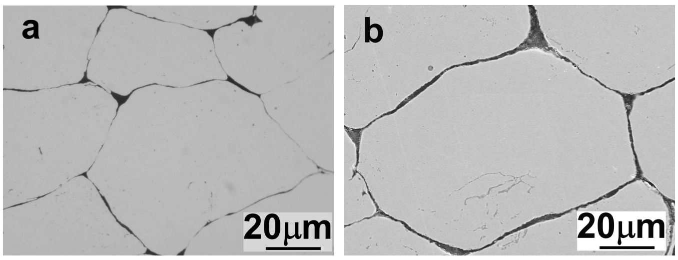

Fig. 2. Microstructure of composite 3VPI (a) and 4VPI (b).

From the comparison of the data in Table II for the 3VPI and 4VPI composites, both based on spherical iron particles, it is seen that electrical resistivity of the composite 4VPI impregnated with SL450 is about 25 times higher than that of composite 3VPI, while relatively low coercivity obtained for both 3VPI and 4VPI composites, is comparable with value for composite 2VPI. From the analysis of the microstructure, Fig. 2, it is seen that the difference in electrical resistivity results from different insulating layer thickness, which is significantly larger for the composite 4VPI.

However, the insulating layer for both composites is continuous and well isolates the iron particles. SEM analysis confirmed that no metallic necks were created at pre-sintering. In terms of the insulating layer thickness it is evident that shellac dissolved in ethanol is preferable for VPI as the SL450 thermoset.

4. Conclusion

The effect of the shape of iron particles on the continuity, thickness and distribution of insulating layer in the Fe/SiO_2 composites, prepared by non conventional new developed vacuum/pressure impregnation of pre-sintered components with shellac and with SL450 thermoset, was evaluated and correlated with the electrical resistivity and coercivity. The findings are summarized as follows:

The continuity and distribution of electro-insulating layer in VPI composites strongly depends on the shape of iron particles.

Using irregularly shaped particles with protrusions on the surface results in metallic necks formation and consequently a breach of continuity of the insulating layer.

Spherical shape and smooth surface of iron particles enabled an even distribution of SiO_2 nanoparticles that created effective "spacers" between adjacent iron particles. Thus effective "barriers" appear, preventing the metallic necks formation and allowing excellent filling-up the space between ferromagnetic particles.

In terms of the insulating layer thickness it is evident that shellac dissolved in ethanol is preferable for VPI as the SL450 thermoset.

Creating a continuous insulating SiO_2/shellac layer, well isolating iron particles has led to acceptable values of electrical resistivity and coercivity, 776 $\mu\Omega\cdot m$ and 178 A/m, which we hope will result in good ac soft magnetic properties at frequencies above 1 kHz.

Acknowledgments

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