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Imaging of Magnetic Domain Structure in FeSi/Mn_{0.8}Zn_{0.2}Fe₂O₄ Composite using Magnetic Force Microscopy

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Soft magnetic composite was prepared by coating the commercial FeSi powder by Mn_{0.8}Zn_{0.2}Fe₂O₄ (MnZn) ferrite, which serves as a potential dielectric phase. MnZn ferrite was prepared by the sol-gel method followed by the auto-self combustion process. The spinel structure of MnZn ferrite was analyzed by X-ray diffraction technique. The composite material was prepared by uniaxial compaction technique and sintered either by conventional or unconventional microwave method. The microwave sintering was applied in order to reduce a grain growth and decrease the overall sintering time. The microstructure of FeSi powder is formed by grains of different diameter. Magnetic force microscopy and scanning electron microscopy were used for an investigation of the correlation between the grain size, grain boundaries and magnetic domains. Magnetic force microscopy visualization of magnetic domains in the prepared soft magnetic composite brings insight into how the magnetically active coating (MnZn ferrite) influences the soft magnetic (FeSi) powder under the influence of the external magnetic field.

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1. Introduction

Soft magnetic composites (SMCs), which are of technological relevance in electromagnetic applications, can be described as ferromagnetic powder particles surrounded by an electrically insulating film. A design of the novel microcomposite material composed of commercial spherical FeSi particles produced by Höganäs Corporation [1] and Mn_{0.8}Zn_{0.2}Fe₂O₄ (MnZn) ferrite was reported together with a basic characterization of electrical and magnetic properties [2]. The sol-gel auto-combustion method was used for a preparation of MnZn ferrite, which has a spinel-type crystal structure as verified by X-ray diffraction (XRD) analysis.

The atomization is a standard thermal treatment process for the FeSi powder production, which consequently leads to an internal recrystallization of the original grains. Each FeSi particle consists of several grains. Each separate grain has a random orientation of its easy magnetization axis and is sufficiently large to split into several magnetic domains. On the other hand, the sub-micrometer sized MnZn ferrite consist of mono-domain structures located in between the large spherical FeSi particles.

The present work takes advantage of the magnetic force microscopy (MFM) for imaging of the magnetic domains in the final composite sample as well as to understand

ability to conduct a magnetic interaction between FeSi particles through a thin MnZn ferrite layer. The preparation of sample for a visualization of domain structure using MFM techniques is the hardest problem because polishing introduces an internal stress. The focused ion beam (FIB) technology represents the most nondestructive way of polishing FeSi/Mn_{0.8}Zn_{0.2}Fe₂O₄ in comparison with mechanical, electrolytic or mechanochemical procedures. The polishing of composite samples by means of FIB allows tracking of the magnetic domain structure close to its natural state in comparison with the various kinds of domain structures examined by aforementioned alternative polishing methods. The unconventional, low cost and low consuming time sintering process was used in order to reduce the grain growth. The microwave heating has also attracted considerable scientific interest as a technique for ceramic manufacturing.

The fundamental difference between microwave and conventional processing is in the heating mechanism and can be utilized for selective heating of materials. The molecular structure affects the ability of the microwave to interact with materials and transfer energy. When materials in contact have different dielectric properties, microwave selectively couples with the higher loss material. This phenomenon of selective heating has many applications. The present work is focused on a preparation of FeSi/Mn_{0.8}Zn_{0.2}Fe₂O₄ composite heat treated by microwave sintering method. The visualisation of magnetic domain structure by means of MFM method was used for evaluation of magnetic activity of dielectric ferrite layer between FeSi particles.

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2. Experimental

The commercial powder of spherical FeSi particles with size about $150\ \mu\text{m}$ in the diameter was used as the base ferromagnetic material. The analytical pure chemicals $\text{Mn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (99%, Acros Organic), $\text{Zn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (98%, Acros Organic), $\text{Fe}(\text{NO}_3)_3$ (99%, Acros Organic) and $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ (99.8% CentralChem) was used to synthesize $\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrite. The equimolar ratio of metal nitrates and citric acid were dissolved in a minimum amount of distilled water under continuous stirring. The pH=6 was adjusted by NH_4OH (26% aq., Sigma Aldrich). 5 g of the prepared gel was mixed with 20 g of FeSi particles and heated at $200\ ^\circ\text{C}$ for 12 h in laboratory dryer. The exothermic reaction and self-propagating combustion process initiated after the creation of xerogel. The 4 wt% of ferrite was created in the final composite powder agglomerate samples. The coated powder was compacted at 600 MPa in cylindrical die. The sintering process was realized in air atmosphere using the laboratory chamber at $780\ ^\circ\text{C}$ or single mode microwave sintering device (microwave generator 750 W) in air. Cylindrical cavity of diameter 28 mm and of height 80 mm was used. The microstructure and morphology of all the samples were examined by scanning electron microscopy (SEM, JEOL JSM-7000F) equipped with the energy dispersive X-ray analyzer (EDX). The structural analysis was carried out using XRD (PhilipsX' PertPro, $\text{Cu}\ K_\alpha$ radiation). AFM/MFM measurements were carried out at room temperature using Agilent 5500 AFM. Raw data obtained from the microscope were processed in Gwyddion open source software (<http://gwyddion.net>). Surface of specimens observed was polished by Ga ions using FIB LYRA3 FEG/SEM (Tescan, CZ).

3. Results

A comparison of the XRD pattern of the synthesized MnZn ferrite with the PDF-4 database (1-089-7550) demonstrates a presence of the spinel-type structure only (see Fig. 1). The computation of crystal parameters from the diffraction pattern by the Rietveld method with structural model based on ICSD (28516) determines the lattice parameter $a = 0.8445\ \text{nm}$ and crystalline size about 20 nm from the Scherrer formula. The shape of peaks and broadening of bottom of peaks indicate the presence of highly disordered (amorphous) phase. The crystalline phase in the sample can be indexed as the face centered cubic (fcc) structure corresponding to the inverse spinel structure. Figure 2 shows the SEM image and EDX analysis employed in order to verify the chemical composition of the final sintered samples. Figure 2a shows the SEM image of $\text{FeSi}/\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ composite, in which the spherical FeSi particles are linked together through the MnZn ferrite. The very fine polishing of sample by means of FIB technology was used for further MFM visualization. The individual grains with clearly visible boundaries are evident from SEM image. FeSi particles are separated from each other by the MnZn ferrite coating. The chemical composition and distribution

of oxygen and metal elements visualised in Fig. 2b–f clearly confirm this statement.

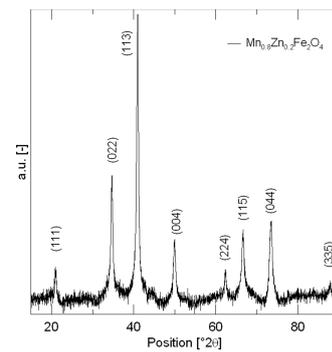


Fig. 1. XRD of spinel $\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$.

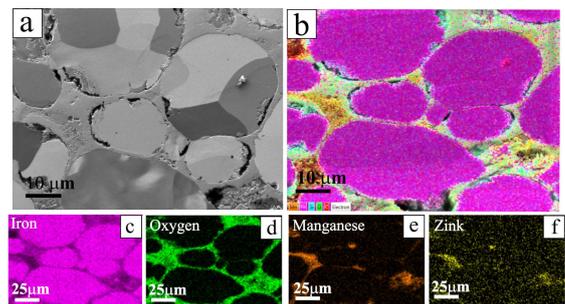


Fig. 2. SEM and EDX analyses of $\text{FeSi}/\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ composite sample, with visualization of oxygen and metal elements in ferrite.

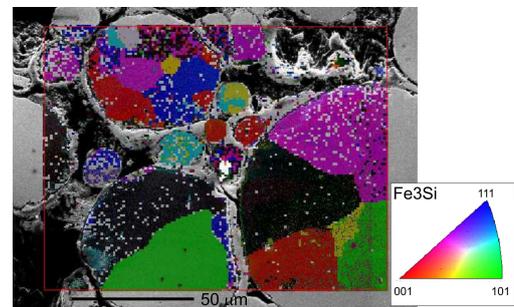


Fig. 3. EBSD image of $\text{FeSi}/\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ composite sample.

The individual grains evidently have different crystallographic orientation, because different colored parts can be recognized from the electron backscattering diffraction (EBSD) image shown in Fig. 3. The relevant color in the EBSD map represents an average misorientation of fcc crystallographic axes, which is expressed by the Euler rotational angles within Bunge notation [3] in each analysed point. The crystal orientation map visualised by EBSD analysis determines the grain structure of FeSi particle.

The MFM image of $50 \times 50\ \mu\text{m}^2$ cross-section is shown in Fig. 4 for the sample sintered by the conventional heat treatment (Fig. 4a) and by means of microwave device

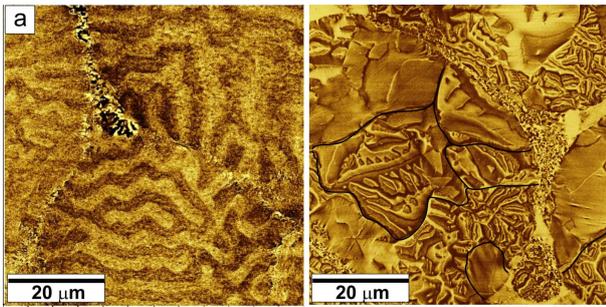


Fig. 4. MFM image of FeSi/Mn_{0.8}Zn_{0.2}Fe₂O₄ composite sample prepared by: a) conventional sintering process in the laboratory chamber at 780 °C in air; b) in the microwave sintering device.

(Fig. 4b). One may recognize several distinct features though the same method of the sample preparation and FIB polishing were used for both types of the samples.

The progressing of large domains of the same type was observed in FeSi particle in the case of conventionally sintered sample (Fig. 4). By contrast, a possible fall out of MnZn ferrite microparticles could cause the minor scratches in the microwave sintered sample, which results in the deformed domain structure of FeSi particles (Fig. 4b). A clear demarcation of grain boundaries with different domain structure determined from EBSD analysis can be observed. In the magnetic image, the outline of the grain is reproduced as a thin black line for better visualization.

Figure 5a visualizes the morphology of the MnZn ferrite coating, which has a function of dielectric and magnetic active binder connecting FeSi particles. It is known that the ferrite coating basically influences the domain wall motion; several recent studies have verified a subtle relation of the domain structure of polycrystalline MnZn and NiZn ferrites on the average grain size [4]. The prepared MnZn ferrite has average particle size from nanometric to micrometric dimensions (Fig. 5a). In general, the grain size can be controlled by the sintering temperature and type of sintering, which allow one to tune the grain size by maintaining a constant composition. It has been shown that a critical grain D_{cr} for the MnZn ferrites, below which the grains become monodomain, is about 4 μm [3, 5]. Recently, it was found from magnetic susceptibility measurements that the energy dissipation at MHz frequencies is substantially lower in the samples with lower grain size $D < D_{cr}$. This fact can be attributed to absence of the domain walls in these small grains.

It is quite obvious from MFM image (Fig. 5b) that the most MnZn ferrite grains have smaller grain size than the critical value D_{cr} as evidenced by a large content of small monodomain particles. It is tempting to conjecture that the prepared MnZn ferrite may act as a magnetically active insulating spacer in between FeSi particles, which may possibly promote a passing of the magnetic domains between different FeSi particles separated through the thin MnZn ferrite (see the upper part of Fig. 5b).

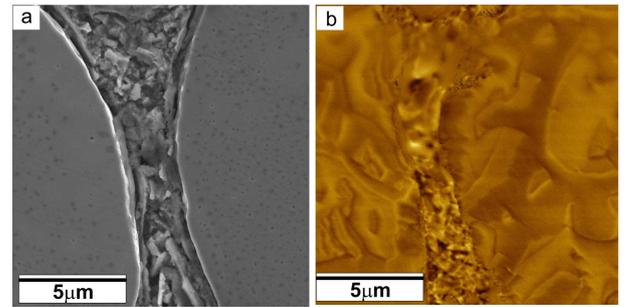


Fig. 5. a) SEM and b) MFM images of FeSi/Mn_{0.8}Zn_{0.2}Fe₂O₄ composite sample prepared by microwave sintering device with the main focus on the MnZn ferrite insulating part.

4. Conclusions

In the present work we have described a design of the novel soft magnetic composite FeSi/Mn_{0.8}Zn_{0.2}Fe₂O₄ prepared from the FeSi powder and MnZn ferrite using uniaxial compaction and two different sintering processes. The unconventional microwave sintering has been used to reduce the growth of grain size in both FeSi particles as well as MnZn ferrite. The final composite samples were subsequently polished by FIB technology before performing MFM imaging in order to reduce the stress introduced by other polishing methods. The MnZn ferrite with inverse spinel structure turns out according to MFM as a good magnetically active coating, which promotes a passing of the magnetic domains between individual FeSi particles.

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