SOFT MAGNETIC COMPOSITES DESIGNED FROM FeSi POWDER AND Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ SINTERED BY SPARK PLASMA AND MICROWAVE SINTERING PROCESSES

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Abstract
Soft magnetic composites (SMCs) base on FeSi particles covered as a core by the ferrite dielectric coating Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$. The secondary ferrite phase was synthesized by a sol-gel auto-combustion method in situ with FeSi particles. The uniaxial compaction at 600 MPa was used for production of cylindrical- and ring-shaped samples. The usual final step during the sample preparation with the help of PM technology is sintering or direct hot pressing. The present work sheds light on differences in the microstructure and basic mechanical properties caused by the use of three different sintering processes. The microwave sintering and spark plasma sintering processes were used as convenient alternatives with respect to conventional sintering in furnace at the air atmosphere. The homogeneous distribution of the insulating coating is responsible for a high specific electric resistivity of the final composite material, which indicates perspective application of this SMC material at medium and high frequencies.

Keywords: soft magnetic composites, Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ ferrite, FeSi particles, microwave sintering, spark plasma sintering

INTRODUCTION
Soft magnetic composites (SMCs), which are of technological relevance in electromagnetic applications, can be described as ferromagnetic powder particles surrounded by an electrical insulating film [1]. PM technologies are indeed able to produce SMCs with a high enough density and sufficiently stable mechanical properties, whereas an insulating layer between magnetic powder particles ensures a high electric resistivity that minimizes the overall magnetic losses [2]. The main advantage of SMCs prepared by PM methods with respect to laminated soft magnetic materials lies in their unique magnetic properties such as an isotropic three-dimensional ferromagnetic behavior, low eddy current losses, as well as, relatively lower total core losses at medium and high frequencies [3,4]. In this regard, another promising way to produce SMCs with excellent properties consists in using spinel ferrites as a coating with an extremely high electric resistivity and almost zero eddy current losses in ac magnetic fields, which will not basically affect the magnetic characteristics of metal cores (high magnetic flux density, low coercitive force) [5].

Other perspective method for production of SMCs of activating the sintering process involves the use of electrical current. These advantages include: lower sintering temperature, shorter holding time, and marked comparative improvements in properties of
In recent times the microwave synthesis has emerged as an efficient technique for synthesis of new materials [8-10]. It is cost effective, time saving and environment friendly. When exposed to microwave radiations, materials may reflect, absorb or remain transparent. The absorption of microwave radiation causes rise in temperature of material due to dielectric heating. Energy consumption is substantially lower during the microwave heating in comparison to conventional furnace [11].

The goal of the present work lies in the use of two different alternative sintering processes for a preparation of soft magnetic composites (SMCs): microwave sintering and spark plasma sintering. Sintering conditions are predominantly determined by the individual components of the designed composite. The investigation of microstructure, basic material characteristics and their optimization according to the used sintering process represents a challenging task of our work.

**EXPERIMENTAL**

The sol-gel synthesis accompanied with the auto-combustion process was used for a preparation of $\text{Mn}_0.8\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ [12]. The $\text{FeSi/MnZnFe}_2\text{O}_4$ particles were created by mixing of 5g of the prepared gel with 20 g of FeSi particles and heated at 200°C for 12 hours in laboratory dryer. The final composition of samples included 6.1wt% of $\text{Mn}_0.8\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$. The prepared composite powders were cold compacted into cylindrical specimens ($\Phi 10 \times 5 \text{ mm}$) at 600 MPa. The first sintering process was realized at 780°C for 30min in laboratory chamber (LCH, HI CERAMIC) in air. The alternative sintering processes were realized in air atmosphere using single mode microwave sintering device (MW, microwave generator 750 W) and spark plasma sintering (SPS, FCT Systeme GmbH, model HP D10-SD), 10min at 35MPa, pulses 25ms/2ms. The sintering temperature profiles of both MW and SPS sintering processes are shown in Figs. 1 and 2. The microstructure and morphology of all the samples were examined by the scanning electron microscope SEM (JEOL JSM-7000F or TESCAN Lyra3) equipped with the energy dispersive X-ray analyzer (EDX). The measurement of coercivity was performed using the Forster Koerzimat HCJ 1.097.

![Fig.1. Time-temperature dependence of microwave sintering process.](image1)

![Fig.2. Time-temperature dependence of spark plasma sintering process.](image2)
RESULTS AND DISCUSSION

The well-described sol-gel synthesis of Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ accompanied with the auto-combustion process, coating process and detailed characterization of the spinel ferrite by TEM and XRD analysis are described in our previous work [12]. The original FeSi particles distributed by Höganäs corporation were used as ferromagnetic cores (Fig.3a). Note that the MnZn ferrite can not instantaneously produce as a continuous coating on FeSi particles because of very fine and porous nature of prepared ferrite from a sol-gel synthesis. One may instead observe a porous layer of the MnZn ferrite surrounding FeSi particles before compaction (Fig.3b). The pressing is responsible for a breakdown of the as-prepared ferrite structure and consequently, the non-continuous coating of the MnZn ferrite surrounding spherical FeSi particles is build up when using conventional sintering process (see Figs. 4a,b). The inhomogeneous distribution with large agglomerates of ferrite phase between individual FeSi particles is seen from Fig.4a. Moreover, the significant porosity was raised during this sintering process in the bulk. Two alternative sintering processes, the microwave and spark plasma sintering were therefore used for a comparison with the usual sintering in air. The ferrite network of the secondary phase was confirmed from the polished samples of FeSi/Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ created after microwave sintering process (Fig.5a). The more adhesive and compact coating in comparison with coating created by conventional sintering process is evident from insignificant porosity and adhesive coating of the fractured surface of FeSi/Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ (Fig.5b). The typical ferrite layer structure created after MW sintering process is visualized from the zoom of Fig 5b. (Fig 5c). EDX analysis properly confirmed the presence of ferrite phase Fig.5d. The resulting microstructure created from SPS process and distribution of ferrite phase in composite material is shown in Figs. 6 and 7. Two different temperatures of sintering processes: 800 °C (Fig.6) and 1000 °C (Fig.7) were used. The significant difference between fractured surfaces are evident from both SPS processes. It is evident that the boundaries between Fe particles in the sample treated at 1000 °C disappeared and no signal of ferrite phase was detected from EDX analysis (Fig.7b). In addition, the compaction procedure has caused a significant deformation of FeSi particles (Fig.7a).

Fig.3. SEM images from: a) the original FeSi spherical powder (97% Fe and 3% Si).
Fig. 4. SEM images from sintering process in air: a) the cross section of FeSi particles surrounded by $\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrite, b) the individual FeSi particle covered by $\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrite with EDX analysis.

Fig. 5. SEM images from MW sintering process: a) the cross section of FeSi particles surrounded by $\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrite, b) the fractured surface of FeSi/$\text{Mn}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$, c) zoom from Fig. 4b of the ferrite phase, d) EDX analysis of ferrite originated from place marked on Fig. 4c.
Fig. 6. SEM images from SPS process at 800 °C: a) the cross section of FeSi particles surrounded by Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ ferrite, b) the fractured surface of FeSi/Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$, c) zoom on FeSi particle covered by Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$, d) EDX analysis from place marked in the Fig 6c.

The differences between fraction surfaces after SPS processes at 800 °C and 1000°C are more evident from the Figs. 6b and 7c. It is quite apparent that the compaction procedure at 800 °C did not cause any noticeable deformation of the spherical FeSi particles however the sintering temperature set up to 1000 °C was so high that it caused the formation of interconnection between FeSi particles. The rise of interconnection during increasing temperature caused the totally different mechanism of breakage. The examination of fracture surface of FeSi/Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ composite at 800 °C confirmed that the damage of overall structure was propagated through ferrite phase (Fig.6b,c). In contrary to this, the both type of fractured surfaces was observed on fig 7c. The ductile fracture surface (Fig.7d) originated from the zoom of the top in Fig.7c and the typical river pattern of brittle fracture surface (Fig.7d) visualised from the zoom of botton part of Fig 7c.
Table 1 shows only negligible increase in a density of the samples sintered under MW and SPS processes. The more significant increase in a density is evident only in the case of sample SPS at 1000 °C because of a mutual sintering of individual FeSi particles. The emergence of mutual contacts is also apparent from the values of the electrical resistivity, which has confirmed conductive character of the sample. The preliminary measurements of coercitive fields were done on the samples sintered by the convention method in the air at 800 °C. The measured values of coercitive field were around 926 A/m what is too high for soft magnetic material. The further measurements for the samples sintered under MW and SPS processes are currently in progress.
Tab. 1. The used temperature, density, mechanical hardness and electrical resistivity.

<table>
<thead>
<tr>
<th>Sintering process</th>
<th>$T_{\text{max}}$ [°C]</th>
<th>$\rho$ [g.cm$^{-3}$]</th>
<th>HV [GPa]</th>
<th>$R$ [$\Omega$.cm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LCH</td>
<td>800</td>
<td>6.46 ± 0.01</td>
<td>1.31 ± 0.08</td>
<td>2.4 ± 0.4</td>
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<tr>
<td>MW</td>
<td>800</td>
<td>6.59 ± 0.01</td>
<td>1.44 ± 0.02</td>
<td>4.7 ± 1.7</td>
</tr>
<tr>
<td>SPS</td>
<td>800</td>
<td>6.56 ± 0.01</td>
<td>1.39 ± 0.01</td>
<td>0.02 ± 0.002</td>
</tr>
<tr>
<td>SPS</td>
<td>1000</td>
<td>6.91 ± 0.01</td>
<td>1.48 ± 0.01</td>
<td>conductive</td>
</tr>
</tbody>
</table>

CONCLUSION

The composite material was designed from the spherical FeSi particles and the soft magnetic ferrite Mn$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ with the inverse spinel structure. The auto-combustion process of ferrite preparation was responsible for preparation of composite powder coated by the MnZn ferrite in the form of large soft agglomerates, which were used for the subsequent compaction and final sintering of green compacts. Three different sintering processes were used for final heat treatment of the composite sample. It has been found that in the case of MW sintering the fine porous MnZn ferrite structure provides a very convenient secondary phase during the compaction process and subsequent creation of tight arrangement of spherical FeSi particles onto dense body without any significant porosity. SPS sintering process has improved the density of the samples at given temperatures, but it could not be applied at 1000 °C for a preparation of the sample with the insulating ferritic phase.

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REFERENCES