STUDY OF MAGNETIC PROPERTIES OF SAMARIUM SUBSTITUTED NICKEL-ZINC FERRITES

Mariana Ušáková* — Elemír Ušák* — Martin Šoka*

NiZn ferrites with iron ions partly substituted by samarium having the chemical composition $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ (x = 0.00, 0.02, 0.04, 0.06, 0.08 and 0.10) were synthesized by means of standard ceramic technology at the temperature of 1200°C for 6 hours. The influence of a small amount of Sm^{3+} ions substituting Fe^{3+} on the resulting magnetic properties of prepared ferrite samples such as Curie temperature, coercive field, remanent magnetic flux density, hysteresis loop area, amplitude and initial permeability has been studied. The chemical composition of prepared samples was studied using scanning electron microscopy with energy-dispersive X-ray spectroscopy. As confirmed by the measurements of magnetic properties at room temperature, the substitution of Sm strongly influences the resulting magnetic properties of studied material in an easily controlled way.

Keywords: rare earths, Sm-doped NiZn ferrites, magnetic properties, hysteresis loops

1 INTRODUCTION

Rare-earth (RE) ions belong to a wide group of perspective additives for "tuning up" the magnetic properties of spinel ferrites with regard to any particular application. The components based on nickel-zinc ferrites are widely used in various electronic devices, such as transformers, microwave absorbers, *etc*, thanks to very good magnetic properties, mechanical strength, chemical stability and low cost of used materials, [1, 2]. Even a relatively small substitution of Fe ions by RE ions remarkably influences structural and magnetic properties of ferrites. Substituting RE ions are characterized by the ionic radii larger than that of Fe, resulting in the deformation of crystalline lattice and the changes in the magnetocrystalline ordering as well as magnetic behavior of materials with spinel structure, [3, 4]. Earlier studies were focused on the investigation of the effect of some RE additives (La, Gd and Eu) and Y with various amounts on the magnetic and structural properties of Ni-Zn ferrites [5, 6]. In this study, the influence of increasing concentration of samarium on the magnetic behaviour of the spinel ferrites has been examined.

2 SYNTHESIS OF THE SAMPLES

Samarium-substituted NiZn ferrite samples with the chemical composition $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ (x = 0, 0.02, 0.04, 0.06, 0.08 and 0.10 ions per formula unit - i/fu) were prepared by means of standard ceramic technology based on solid state reaction. Commercially available relevant oxides, *ie* NiO, ZnO, Fe_2O_3 and Sm_2O_3 of high purity (AR grade) in stoichiometric ratios were used as the raw materials. The raw oxides were homogenized by wet-milling in the ball mill, dried and thermally treated at the temperature of 950°C for 1 hour. Prepared semi-product was homogenized and dehydrated once more. A part of dried calcined powder was blended in polyvinyl-alcohol (acting as binding matrix) and pressed into the pellet shape with the diameter of 15 mm. Subsequently, the pellets were sintered in the furnace at the temperature of 1200°C for 6 hours. Further, the holes were drilled into the pellets by means of diamond routers and drill bits. Thus, ring-shaped samples with outer diameter of about 12 mm and inner diameter of 6 mm used for the measurement of magnetic measurements have been obtained. The semi-product remained after the first thermal treatment, homogenization and dehydration was sieved using the sifter with 500 μm mesh hole size and finally annealed at the temperature of 1200°C for 6 hours. Prepared powder samples were used for the measurement of the temperature dependencies of magnetic susceptibility as well as for scanning electron microscope (SEM) analysis.

3 CHARACTERIZATION TECHNIQUES

The surface morphology and chemical composition (the presence of individual chemical elements) of prepared ferrite materials were studied using scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDX) for semi-quantitative analysis.

The Curie temperature was determined from the temperature dependencies of magnetic susceptibility measured by automated balancing bridge, based on the detection of inductivity changes in a measuring coil due to the presence of the measured sample inside the coil.

Basic magnetic properties, such as the coercive field, remanent magnetic flux density, hysteresis loop area (proportional to the total magnetization losses), amplitude and initial permeability were evaluated from low-frequency magnetization curves measured by automated experimental setup built up from commercially available instruments. For each sample, the series of quasi-static minor loops at the frequency f = 50 Hz and sinusoidal exciting field with the amplitude H_m changing from 10 to 1500 A/m with the step of 10 A/m was measured.

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4 RESULTS AND DISCUSSION

Quantitative analysis of prepared ferrites determined the weight content of Ni, Zn, Sm, Fe and O ions, the results are summarized in Tab. 1. Only small amount of Sm^{3+} in the sample with composition the $Ni_{0.42}Zn_{0.58}Sm_{0.02}Fe_{1.98}O_4$ did not allow to confirm the presence of Sm by this method.

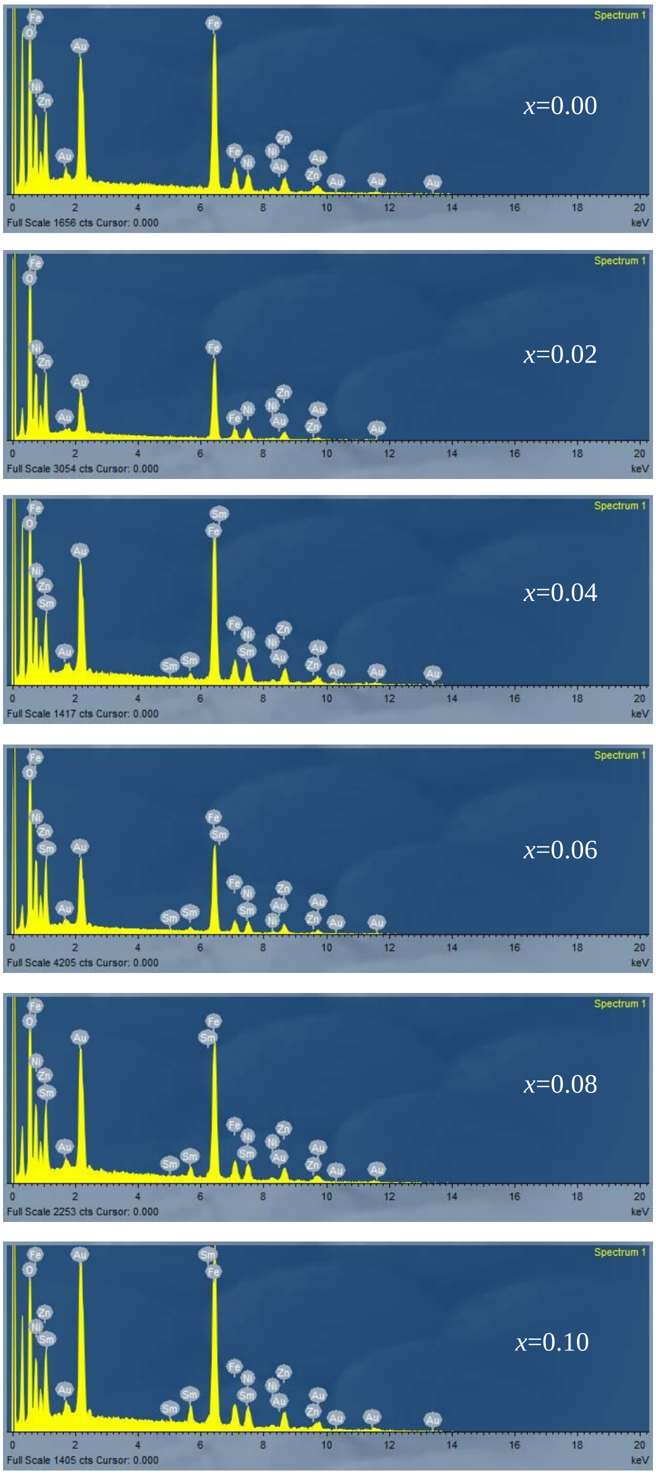

Fig. 1. EDX analysis of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite

Table 1. The mass content of individual ions in $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite

x (i/fu)	w_{Ni} (%)	w_{Zn} (%)	w_{Sm} (%)	w_{Fe} (%)	w_O (%)
0.00	9.24	16.38	-	48.10	26.28
0.02	7.88	19.14	-	35.79	37.19
0.04	9.85	13.70	3.35	46.90	26.17
0.06	7.58	19.04	2.83	36.06	34.49
0.08	8.36	15.46	6.08	43.08	27.02
0.10	9.39	13.41	8.11	47.23	21.85

Evident differences in weight contents against theoretical values, especially for sample with $x = 0.02$ and 0.06 are probably associated with structural inhomogeneity and irregular location of particular ions in the ferrite structure due to ceramic preparation technique.
Figure 1 confirmed EDX results in graphical form where the presence of small content of Au^{3+} (as the residue of golden coating of the specimen) is visible.

The temperature dependencies of magnetic susceptibility $\chi(T)$ for powdered ferrite samples of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$, $x = 0 - 0.10$ i./f.u. are plotted in Fig. 2. A relatively sharp fall of the magnetic susceptibility close to Curie temperature T_C due to the loss of the magnetic behavior is apparent from the figure. The values of T_C were determined from the inflexion point of $\chi(T)$ dependencies. The dependence of T_C on Sm contents shows oscillations of T_C, within 183-187°C except for $x = 0.04$, Fig. 5. Slight increase of T_C for the sample with the composition $Ni_{0.42}Zn_{0.58}Sm_{0.04}Fe_{1.96}O_4$ ($T_C = 199.8$°C) may originate from direct relationship between the lattice parameter and the magnetic interaction within ferrite sublattice which is in agreement with [7, 8]. Since the parameters from X-ray diffraction analysis are not yet available, this behavior (even if well supported by published information) needs to be subjected to further analysis.

For the illustration of a significant influence of Sm contents, the hysteresis loops of all the samples at the same amplitude of $H_m = 1500$ A/m are in Fig. 3. From these loops, classical hysteretic parameters, such as *eg* coercive field H_c, remanent magnetic flux density B_r and hysteresis loop area A_l were found. Further, the amplitude permeability μ_a for each minor loop was determined as a ratio of the flux density amplitude B_m and exciting field amplitude H_m divided by the vacuum permeability μ_0.

As can be seen in Fig. 3, an increasing substitution of Fe by Sm ions decreases saturated magnetization (here in correlation with B_m, since the samples are close to saturation), which is in accordance with the fact that Sm ions occupy only the octahedral sublattice.

The coercive field and the ratio of remanent and maximum flux density B_r/B_m corresponding to the loops in Fig. 3 plotted as a function of x are in Fig. 4. One can see almost linear *increase* of H_c with Sm contents, whereas B_r/B_m (except for a maximum for $x = 0.04$) with increasing x tends to saturate to a nearly constant value slightly above 0.5.

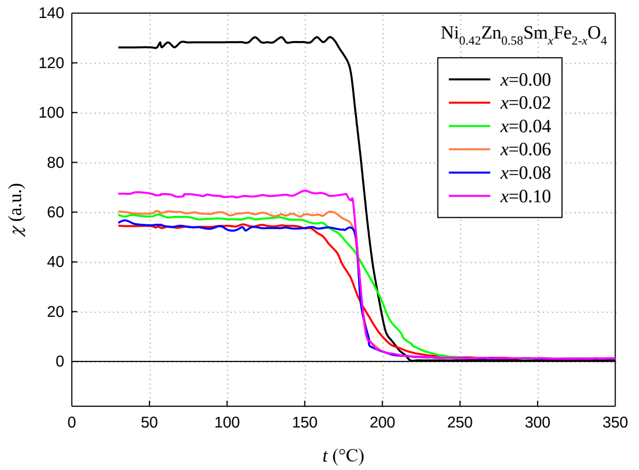

Fig. 2. The temperature dependencies of magnetic susceptibility of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite.

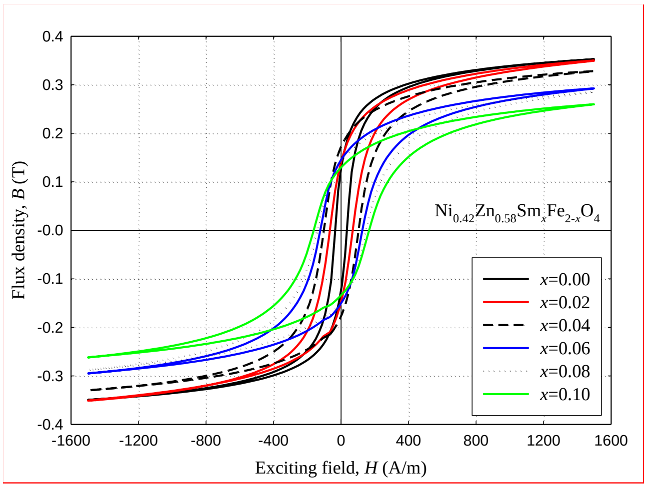

Fig. 3. The hysteresis loops of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite for various Sm content x measured at H_m = 1500 A/m.

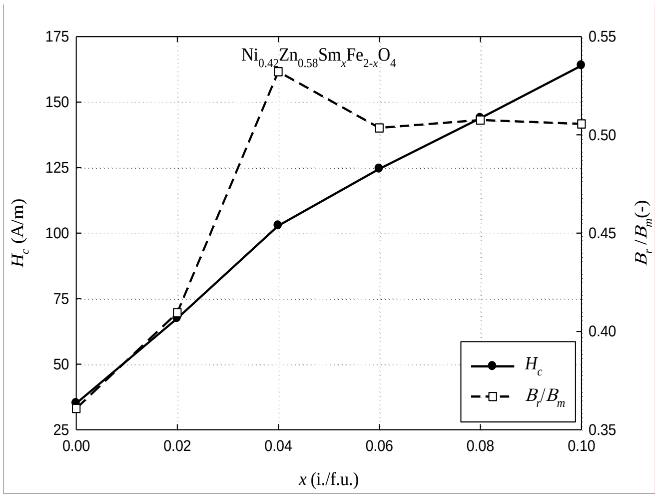

Fig. 4. The dependence of coercive field H_c (•, solid) and the ratio of remanent and maximum flux density B_r/B_m (□, dashed) on Sm content x of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite.

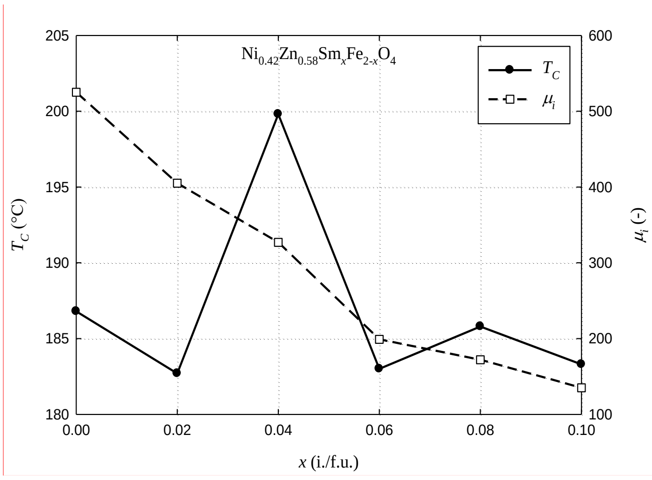

Fig. 5. The dependence of Curie temperature T_C (•, solid) and initial permeability μ_i (□, dashed) on Sm content x of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite.

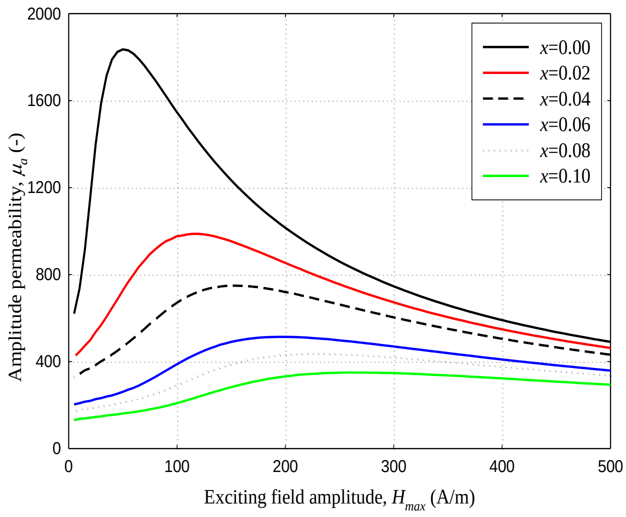

Fig. 6. The amplitude premeability μ_a as a function of aapplied field amplitude H_m for various Sm content x of $Ni_{0.42}Zn_{0.58}Sm_xFe_{2-x}O_4$ ferrite.

Note that for another type of ferrite with the composition $Cu_{0.5}Zn_{0.5}Sm_xFe_{2-x}O_4$ the coercivity *decreases* almost linearly with addition of Sm, which implies almost unlimited manipulation of the magnetic behavior of these materials via the chemical composition, [7].

In Fig. 5, along with Curie temperatures, the dependence of initial permeability μ_i upon x is displayed as well. Initial permeability, found as an extrapolation of the dependencies of the amplitude permeability μ_a upon the applied field H_m (provided in Fig. 6) to zero field, exhibits its monotonous decrease that can possibly be described by the second order polynomial function. The amplitude permeability related to the applied field amplitude exhibits its no extraordinary behaviour - as the amount of Sm x increases, in accordance with observed shape of hysteresis loops the curves become wider with decreasing peak values of amplitude permeability.

5 CONCLUSIONS

The analysis of experimental data approves samarium as a very good iron substituting element for nickel-zinc ferrite with initial composition $Ni_{0.42}Zn_{0.58}Fe_2O_4$ since important parameters representing quasi-static magnetic properties exhibit simple dependencies upon the amount of Sm incorporated into the basic chemical formula of the polynomial-like character of the first or second order. Consequently, by adding defined amount of Sm, the magnetic parameters can be easily controlled by a straightforward way and thus tailored for any particular application required in industrial practice.

Acknowledgement

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