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INFLUENCE OF NiZn FERRITE FILLER CONCENTRATION ON MAGNETIC AND PHYSICAL-MECHANICAL PROPERTIES OF MAGNETOPOLYMER COMPOSITE MATERIALS

Mariana Ušáková* — Jana Rekošová**
— Rastislav Dosoudil* — Elemír Ušák*

The influence of soft magnetic ferrite filler on various properties of the magneto-polymer composite with the polyamide (PAD) as a polymer nonmagnetic matrix was investigated. As the magnetic filler, soft magnetic spinel ferrite with the chemical composition of $Ni_{0.33}Zn_{0.67}Fe_2O_4$ synthesized by standard ceramic method was used. Magneto-polymer composites with various volume concentrations (0 to 30 vol.%) were prepared. The influence of powder filler on the magnetic properties of magneto-polymer composites such as complex permeability was investigated. The measurements of physical-mechanical properties of prepared composite samples confirmed that the presence of ferrite filler in the polymer matrix leads to their significant changes.

Keywords: ferrites, magneto-polymer composites, magnetic properties

1 INTRODUCTION

NiZn ferrites owing to their suitable properties such as high initial permeability, electrical resistivity, low magnetic and dielectric loss, mechanical hardness and chemical stability are perspective materials for various electronic devices. One of the possible applications is their usage as EM-wave absorbers for elimination of the electromagnetic interference (EMI). Such materials, however, cannot be applied in the microwave region because of the decrease of magnetic loss (μ'') due to their Snoek's limit, [1]. On the other hand, the ferrite polymer composite materials, in which the ferrite particles are dispersed in a polymeric matrix, are one of the suitable candidates for thin EM-wave absorbers in GHz range, as their Snoek's limit is enhanced to higher frequency region. Nowadays, the experimental and theoretical studies of these materials are a subject of many research works [2-4]. An advantage of these magneto polymer materials is their low weight, flexibility, elasticity as well as resistance to corrosion or other aggressive chemical atmosphere. Moreover, they exhibit good magnetic properties. The aim of this paper is to study the influence of soft magnetic ferrite filler on magnetic properties of magneto-polymer composites in the frequency range from 10 MHz to 6.5 GHz hereby investigated of the physical-mechanical property changes prepared samples.

2 EXPERIMENTS

2.1 Sample preparation

The ferrite powder with the chemical composition $Ni_{0.33}Zn_{0.67}Fe_2O_4$ synthesised by standard ceramic method, sintered at 1200°C for 6 h in air was used as the magnetic filler. The size of synthesized ferrite particles was reduced by crushing to the fraction of (0 – 40) μm. The samples of magneto-polymer composite were prepared by mixing the prepared ferrite powder with polyamide (PAD) matrix (Vestamid L – Polyamide 12 by Evonik) in the laboratory mixer BRABENDER at 190°C, the filler contents was 0 to 30 vol.%.

2.2 Experimental methods used

In order to investigate the density, specific surface area S_A and porosity of prepared ferrite powder filler, the mercury porosimetry was employed.

The size, shape, distribution and chemical analysis of the ions of ferrite particles were studied by scanning electron microscope (SEM) and laser particle size analyzer.

The Curie temperature T_C of prepared ferrite filler was obtained from the temperature dependencies of magnetic susceptibility measured by the bridge method within the temperature range of 30 – 400°C with constant temperature rise rate of 4°C/min.

The magnetisation characteristics along with basic magnetic parameters (coercivity, remanence, amplitude and/or initial permeability, *etc*) were measured by means of computer controlled experimental equipment built-up from commercially available instruments.

Electromagnetic properties, e.g. the frequency dependencies of real (μ') and imaginary (μ'') parts of the complex permeability of the magneto-polymer composites were obtained by means of impedance spectroscopy (coaxial S-parameter method) in the frequency range from 10 MHz to 6.5 GHz. The measured composite samples were prepared in the form of toroids with an outer diameter of 8 mm, an inner diameter of 3.5 mm and a height of 2 mm.

Physical-mechanical properties of the prepared composites were measured in accordance with valid technical standards by Instron equipment on the double side blade specimens (width 6.4 mm, length 100 mm, thickness 2 mm).

* Slovak University of Technology, Faculty of Electrical Engineering and Information Technology, Ilkovičova 3, 812 19 Bratislava 1, Slovakia, mariana@iris.elf.stuba.sk, ** Slovak University of Technology, Faculty of Chemical And Food Technology, Radlinského 9, 812 37 Bratislava, Slovakia, jana.rekosova@stuba.sk

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3 RESULTS AND DISCUSSION

3.1 Magnetic properties of bulk ferrite

To utilize the ferrite as the magneto-polymer composite filler it is necessary to choose the material with appropriate magnetic parameters, especially high initial permeability. The maximum value of the permeability of NiZn ferrites can be obtained for the material with chemical composition $Ni_{0.3}Zn_{0.7}Fe_2O_4$, [5], but the Curie temperature of this material (T_C = 70°C) is too low for practical use. However, small change of molar ratios of Ni^{2+} and Zn^{2+} ions consisting in the reduction of zinc amount causes an increase of T_C meanwhile the magnetic properties remain acceptable. Consequently, the ferrite with the chemical composition $Ni_{0.33}Zn_{0.67}Fe_2O_4$ with the Curie temperature T_C = 95°C was chosen as the magnetic filler.

An example of the hysteresis loop and amplitude curve of such ferrite measured at frequency f = 50Hz and maximum value of applied exciting field H_m = 200 A.m^{-1} is shown in Fig. 1. The magnetic parameters, such as the maximum flux density B_m, remanent flux density B_r, coercivity H_c, loop area, maximum value of amplitude permeability $\mu_{a,max}$ and initial permeability μ_i, are shown in Table 1. Magnetic characteristics confirm that the prepared ferrite is a suitable material for using as soft magnetic filler in the magneto-polymer composites.

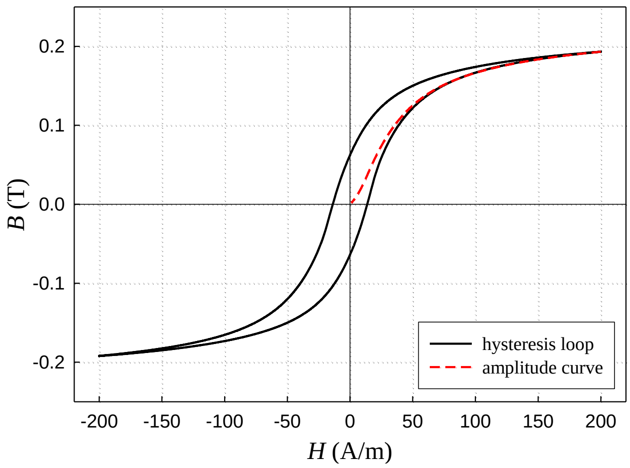

Fig. 1. The hysteresis loop and amplitude curve of $Ni_{0.33}Zn_{0.67}Fe_2O_4$.

Table 1: Magnetic properties of prepared bulk ferrite

T_C (°C)	H_m (A.m^{-1})	B_m (T)	B_r (T)
118	200	0.1932	0.06232
H_c (A.m^{-1})	Loop area (J.m^{-3})	$\mu_{a,max}$ (-)	μ_i (-)
13.6732	9.4538	2353	1316

3.2 Structural characteristics of ferrite powder fillers

The structural characteristics of ferrite fillers, such as the specific surface area S_A, total porosity, density ρ and particle size D are given in Table 2. SEM analysis (Fig. 2) showed irregular particles of polyhedral shape present in the sample, the particle size ranges from 1 μm to 40 μm. The size of NiZn ferrite particles was also confirmed by laser particle size distribution plotted in Fig. 3.

Table 2: Structural properties of prepared ferrite powder used as the filler

S_A (m^2.g^{-1})	*total porosity* (%)	ρ (g.cm^{-3})	D (μm)
0.61	45.65	5.11	26.49

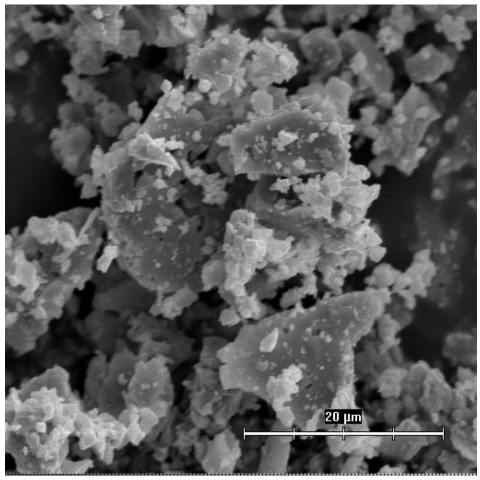

Fig. 2. SEM image of $Ni_{0.33}Zn_{0.67}Fe_2O_4$ ferrite powder particles.

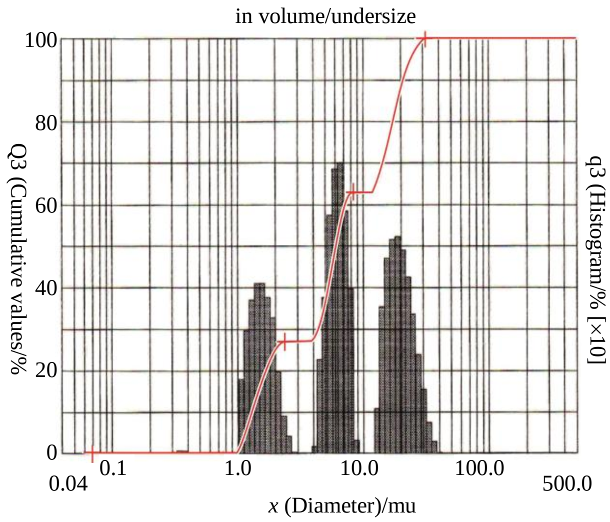

Fig. 3. Particle size distribution of $Ni_{0.33}Zn_{0.67}Fe_2O_4$ ferrite powder.

3.3 High-frequency electromagnetic properties of composite

The measured frequency dependencies of real (μ') and imaginary (μ'') parts of relative complex permeability $\mu = \mu' - j\mu''$, as shown in Fig. 4, exhibit the relaxation type of permeability dispersion on contrary to the resonance one observed in sintered bulk ferrite, [6]. Low-frequency permeability μ' (at the frequency of 10 MHz) increased from about 1.08 (for composite sample with 1 vol.% of magnetic filler) to about 3.45 (for composite sample with 30 vol.% of magnetic filler), and the resonance frequency f_r (at which the imaginary part μ'' had its maximum value) decreased from 6.25 GHz to about 663 MHz, Fig. 5. This can be explained as follows:the magne-

MHz, Fig. 5. This can be explained as follows: the magnetic nature of composite structure primarily depends on the spinel ferrite used as magnetic filler.

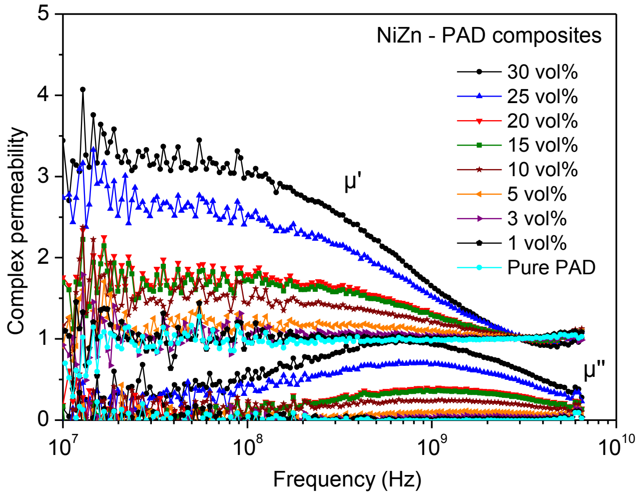

Fig. 4. Measured frequency dependencies of real (μ') and imaginary (μ'') parts of complex permeability

The variation in the frequency dependencies of μ is attributed to the mechanism of natural magnetic resonance (ferromagnetic resonance). In a weak ac magnetic field, basically the domain wall rotation mechanism dominates in the magnetising process of this ferrite, so that μ' in general has a small value. Moreover, the crystallites of ferrites are enveloped by non-magnetic coating of polymer matrix that does not allow the intra-crystalline interactions. This results in a weak dispersion phenomenon in the $\mu''(f)$ dependence. The change in the magnetisation is generally brought about by rotation of spins and/or domain wall displacement. These motions lag behind the applied magnetic field and consequently cause the increase of μ'' and simultaneously the decrease of μ'. The drop of μ'' and the shift of μ'' peaks at certain frequency are owed to the presence of demagnetising field in filler particles.

Another interesting fact can be pointed out in Fig. 5 - see the dependence of low-frequency permeability μ' upon filler volume concentration κ_v. This dependence can be approximated by piecewise linear dependence; for the filler concentrations below 20 vol.% the slope is relatively small, meanwhile above this value it suddenly increases. This seems to be associated with a kind of percolation effect – above some threshold a stronger interaction among ferrite filler grains appears.

3.4 Physical-mechanical properties of composite

The measurement of physical-mechanical properties of prepared composite samples confirmed, that the presence of ferrite filler in the polymer matrix leads to changes of these characteristics. The dependence of the tensile strength value as a function of ferrite contents is illustrated in Fig. 6. From the dependence is obvious at first rapid fall when the tensile strength value decreased about 46% for the sample with 10 vol.% ferrite filler in comparison with tensile strength value of ferrite free composite sample. Following increasing of ferrite concentration leads to slight decline of tensile strength value (about 21% for 20 vol.% of the filler) and for magneto-composites with ferrite filler contents higher as 20 vol.% remained unchanged (25 MPa). Similar decrease of tensile strength was found in case of magneto-polymer composites with increasing content of iron particles filled to natural rubber, [7]. The ferrite present in magneto-polymer composites influences Young's modulus values (Fig. 7). The dependencies Young's modulus from the ferrite filler contents exhibit considerable increase, Young's modulus for the maximum ferrite filler (30 vol.%) reached about 2.7 times higher value compared to pure polymer.

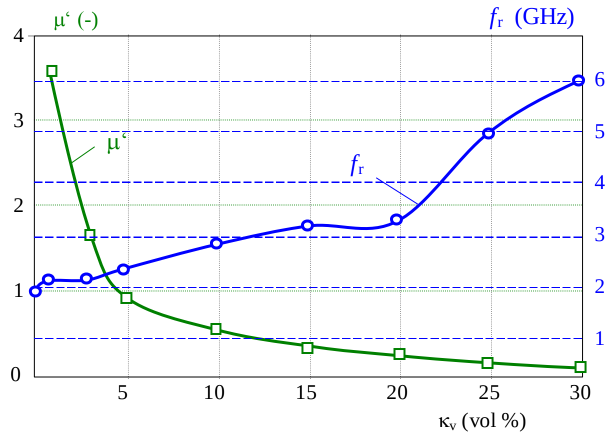

Fig. 5. Real part of complex permeability (μ') and resonant frequency (f_r) as a function of filler volume concentration (κ_v)

4 CONCLUSIONS

The filler volume concentration allows significant changes of the properties of magneto-polymer composite. As can be seen, higher concentration in general increases the low-frequency permeability, meanwhile the physical-mechanical properties get worse. The measurements of physical-mechanical properties of prepared composite samples confirmed that the presence of ferrite filler in the polymer matrix leads to their significant changes. The tensile strength value decreased for about 55 % in case of the sample with 30 vol.% of ferrite filler in comparison with tensile strength value of pure polymer matrix material.

Various filler volume concentration along with using diverse ferrite filler materials allows the modification of various magnetic properties aimed at tailoring the material for any particular application (required operating frequency range, permeability, *etc*).

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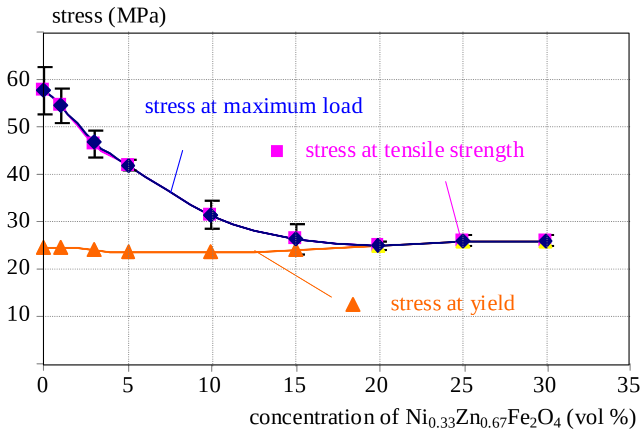

Fig. 6. Influence of ferrite filler concentration on tensile strength

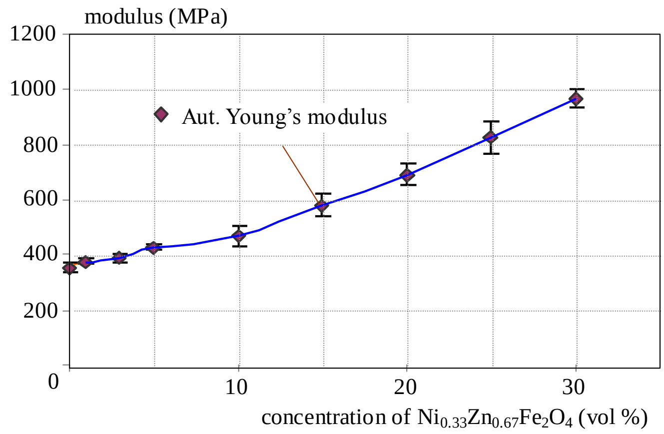

Fig. 7. Influence of ferrite filler concentration on Young's modulus

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Mariana Ušáková (Mgr, Ing, PhD), born in Spišská Nová Ves, Slovakia, in 1967. She graduated from the Faculty of Chemical and Food Technology at the Slovak Technical University in Bratislava, in Electrochemistry Branch in 1990 and received the PhD degree in Material Engineering branch in 2010. At present, she is a research worker at the Department of Electromagnetic Theory, Faculty of Electrical Engineering and Information Technology. Her research activities are mainly focused on soft and hard ferrites and magneto-polymer composite materials.

Jana Rekošová (Ing), was born in Skalica, in 1981. She graduated from Faculty of Industrial Technologies, in Púchov, in Material Engineering branch (Chemistry and Technology of Rubber Production) in 2009. She is a PhD student at the Slovak University of Technology in Bratislava, Faculty of chemical and Food Technology, Institute of Polymer Materials, Department of Plastics and Rubber. Her research activities are magnetically active materials based on elastomers and their physical-mechanical, structural and magnetic properties.

Rastislav Dosoudil (Doc, Ing, PhD) was born in Bratislava, Slovakia, in 1970. He graduated from Faculty of Electrical Engineering and Information Technology, Slovak University of Technology, in Bratislava, in Material Engineering branch, 1993 (technology of electronic equipments) and received the PhD degree in Theory of Electromagnetism in 2000. At present, he is an Associate Professor (2008) at the Institute of Electrical Engineering. He teaches electric circuits, electromagnetics and electronics. His research activities are mainly ferrite-polymer composite materials, complex permittivity/permeability phenomena, electromagnetic wave absorbing properties of composites, ferromagnetic resonance, and magnetic measurements.

Elemír Ušák (Assoc Prof, PhD), born in Piešťany, Slovakia, 1965. Graduated from the Faculty of Electrical Engineering, Slovak University of Technology, Bratislava in Solid State Physics branch, 1988 and received the CSc (PhD) degree in Theory of Electromagnetism in 1996. After habilitation in 2000 he became an associate professor at the Department of Electromagnetic Theory of FEIIT SUT in Bratislava.