

COMPOSITE MATERIALS BASED ON NANOSIZED FERRITE FOR MICROWAVE APPLICATIONS

Svetoslav Kolev^{1,2}, Todor Cholakov², Daniela Kovacheva³, Tatyana Koitzarova¹

¹Institute of Electronics, Bulgarian Academy of Sciences
72 Tsarigradsko Chaussee, 1784 Sofia, Bulgaria

²Neofit Rilski South-Western University, 66 Ivan Mihailov St.
Blagoevgrad 2700, Bulgaria

³Institute of General and Inorganic Chemistry
Bulgarian Academy of Sciences, Acad. Georgi Bonchev St.
Bl. 11, Sofia 1113, Bulgaria
E-mail: svet_kolev@swu.bg

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ABSTRACT

The fabrication of composites with magnetite Fe_3O_4 nanoparticles was investigated in this research as a method of shielding electromagnetic radiation. The effects were studied of various weight percentages of Fe_3O_4 nanoparticles on the microwave absorption. XRD, SEM, and a scalar network analyzer were used to characterize the sample. The results showed that the Fe_3O_4 nanoparticles had an average size of 175 nm. The nanocomposite sample with a thickness of 4 mm and a concentration of 1.5 g in 1 cm³ of silicon rubber had the highest level of reflection loss (-23 dB).

Keywords: nanoparticles, magnetite, microwave absorption.

INTRODUCTION

Nowadays, using electromagnetic (EM) absorber materials is among the most important solutions for countering the EM interference. The absorbers play an essential role in the development of wireless communication and high-frequency circuit devices in the gigahertz frequency range [1, 2]. A number of reports have dealt with improving the performance of EM absorbers, such as employing nanosized particles [1] with magnetic properties [3], utilizing multilayer materials [4], 3D-structure materials [5], developing core/shell structures [6, 7], and combining dielectric-magnetic materials to achieve dielectric-magnetic dual-loss [8].

The EM absorption mechanism on a microscopic scale is related to the change in the direction of the magnetic and electric dipole moments of the absorbing materials [9]. When the size of the material is reduced

to the nanoscale, then the EM waves have a higher probability of interacting with the magnetic or electric dipoles in the material, thus enhancing the absorption. Moreover, using a nanoscale material is an effective way of achieving high-energy absorption [10].

In this study, we report a method of preparing nanocomposites using nanosized Fe_3O_4 particles dispersed in silicon rubber. X-ray diffraction (XRD) and scanning electron microscopy (SEM) confirmed the purity and the nanoscale size of the magnetite particles. The EM wave absorption characteristics of the composites are presented, and absorption mechanisms are discussed.

EXPERIMENTAL

The magnetic materials, magnetite, (Fe_3O_4 nanosized powder) were characterized by different methods. Thus, XRD spectra were obtained using a

Bruker D8 Advance Twin diffractometer in view of phase identification and assessment of the phase purity, while SEM tests were performed by scanning electron microscopy (Philips ESEM XL30 FEG and JEOL 6390) to investigate the microstructure of the samples. Employing a Hewlett-Packard 8756A microwave scalar network analyzer, the microwave (MW) characteristics were measured in the frequency range 1-20 GHz. To determine the MW properties of the composites, we used the short-circuit measurement technique, whereby the electromagnetic wave passes through a single-layer absorber backed by a perfect conductor [11]. The toroidal samples under investigation were fit into a 50 Ω coaxial measurement cell.

The samples for the microwave measurements were prepared as a mixture of Fe_3O_4 nanosized powder and commercial silicon rubber (Mastersil, ASP); they have a ring shape of 7-mm outer diameter, 3 mm inner diameter and 4 mm thickness. We characterized three composite samples with stepwise increasing amounts

of magnetite powder homogeneously dispersed in the polymer matrix, namely, A (1.2 g cm^{-3}), B (1.5 g cm^{-3}) and C (1.8 g cm^{-3}).

RESULTS AND DISCUSSION

The XRD spectra of magnetite powder and of a composite sample are presented in Fig. 1. It is seen that the magnetic material was well crystallized - the spectra exhibit the characteristic peaks corresponding to magnetite only. Thus, the sample reveals a high degree of crystallization and very high purity.

Fig. 2a shows the typical morphology of a magnetite sample. The presence is seen of spherical particles with an average particle size of 175 nm. In the composite sample, quite bigger agglomerates are observed (Fig. 2c).

The critical diameter for single-domain magnetite particles is about 60 nm, so that the particles are in the so-called pseudo-single state. Our previous research on

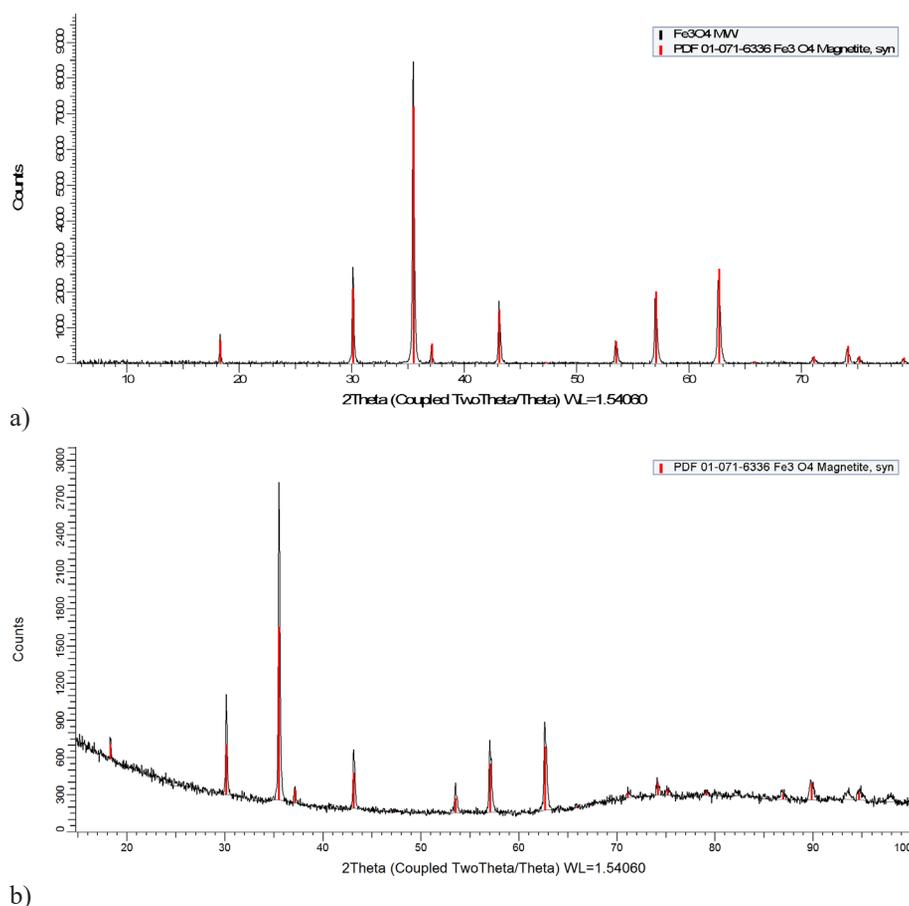


Fig. 1. X-ray diffraction (XRD) patterns of (a) magnetite powder and (b) of a composite sample.

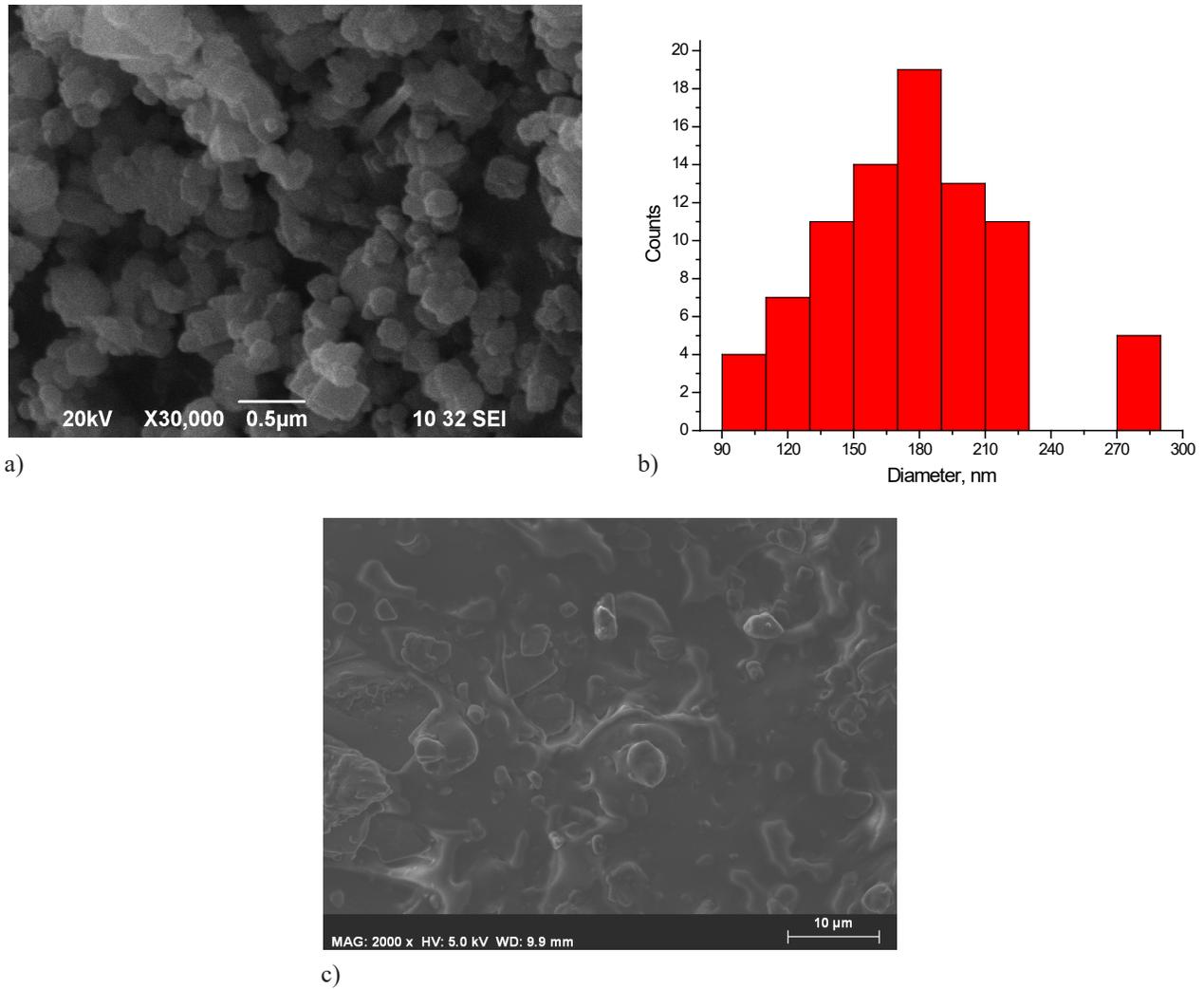


Fig. 2. SEM images of (a) magnetite powder, (b) particles size distribution and (c) composite samples.

the microwave properties has been focused on single and multi-domain magnetite. Therefore, the current study is important in completing our knowledge with data on magnetite in the pseudo-single state.

The main goal of our study was to investigate the microwave behavior of the composite samples (magnetite nano-powder dispersed in a polymer matrix). Fig. 3 shows the evolution of the microwave spectra of the reflection losses (R_L) for the samples of equal thickness (4 mm) with different fillings of the matrix (A, B, C). Since, as was shown in our earlier work [12, 13], the polymer matrix (silicon rubber) is transparent in the electromagnetic wave frequency range 1 - 20 GHz the absorption properties are due to the ferrite (Fe_3O_4) only. The results for the reflection losses R_L are obtained under the assumption that the

electromagnetic wave is incident perpendicularly to the surface of the composite samples backed by a perfect conductor. Based on the transmission line theory, the R_L as a function of the normalized input impedance can be given by [14]:

$$R_L(\text{dB}) = 20 \log \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (1)$$

where

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\varepsilon_r}} \tanh \left[j \frac{2\pi}{c} \sqrt{\mu_r \varepsilon_r} f d \right] \quad (2)$$

In principle, the maximum value of R_L of a material can be achieved by adjusting its permittivity and

permeability. Experimentally, a material process may be used to design a maximum R_L value at a certain frequency (range). In this work, the improvement of the microwave absorption properties resulted from changing these by dispersing different amounts of Fe_3O_4 nanoparticles in silicon rubber.

Since our purpose was to obtain a perfect microwave absorber, we considered two very important factors. The first important condition to be fulfilled is the impedance matching condition between the microwave absorber and free space. In what concerns obtaining zero reflection off the front surface of the samples, the characteristic impedance of the samples must be equal/close to that of the free

space. The best impedance matching could be achieved when the material has equal or very similar ϵ_r and μ_r . The second essential parameter is the microwave attenuation inside the microwave absorber. It should be sufficient to dissipate the incident microwaves by intrinsic magnetic and dielectric loss mechanisms. The reflection losses reaching a minimum corresponds to a minimal reflection and/or a maximal MW energy absorption. This can be observed for a specified density of filling and a certain thickness of the sample at the respective frequency - matching frequency (Fig. 3).

As the experimental results demonstrate, two matching areas exist - in the regions of 4 - 11 GHz and 13 - 19 GHz. Here, one can assume that the first

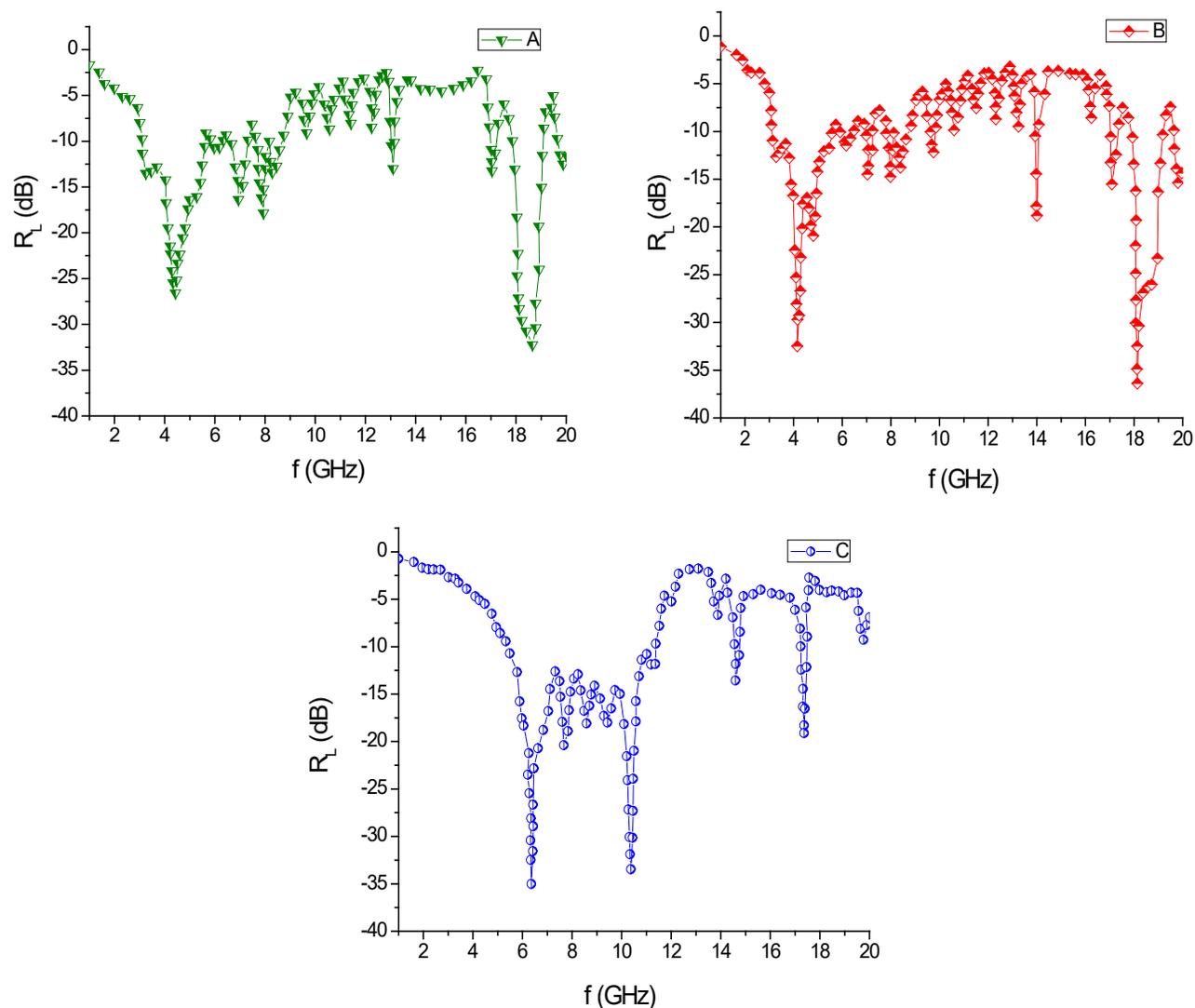


Fig. 3. Reflection losses of samples with different filler ratio: samples A, B and C.

matching frequency is related to the magnetic resonance region [15]. The second matching frequency could be associated with the behavior of a quarter-wavelength resonator [16].

The measurements displayed in Fig. 3 illustrate that the reflection losses depend on increasing the density of the magnetic filler in a specific manner. One can see that for sample A the value of the reflection losses is the lowest at two frequencies - at 4.42 GHz the reflection loss is -26.56 dB; the second minimum is at 18.65 GHz and the reflection loss is -32.21 dB. For sample B, the first minimum is observed at 4.14 GHz, the reflection loss is -32.48 dB; the second minimum is at 18.13 GHz, with a reflection loss of -36.37 dB.

As the filling density is increased, the deepest and broadest peaks for sample C are observed in the region 4 - 11 GHz - one broad peak with two minima at 6.35 GHz and at 10.37 GHz, with the reflection losses being -35.00 dB and -33.45 dB, respectively.

CONCLUSIONS

Our results prove that one can vary the matching frequencies and the reflection losses of a nanocomposite sample in a controlled way by varying the density of the ferrite filler while keeping the sample thickness. By increasing the amount of the magnetic filler, we increased the value of the reflection losses and observed zero reflection at certain frequencies. The nanocomposite structures prepared could find applications as detectors of MW electromagnetic waves or as antireflection coatings.

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REFERENCES

1. H. Husain, M. Sulthonul, B. Hariyanto, Y. Taryana, W. Klyusubun, S. Wannapaiboon, D. Darminto, S. Pratapa, The structural and magnetic characterization of ironstone-derived magnetite ceramic nanopowders, *J. Mater. Sci. Mater. Electron.*, 31, 15, 2020, 12398-12408.
2. T. Iwamaru, H. Katsumata, S. Uekusa, H. Ooyagi, T. Ishimura, T. Miyakoshi, Development of microwave absorbing materials prepared from a polymer binder including Japanese lacquer and epoxy resin, *Phys. Procedia.*, 23, 2012, 69-72.
3. J. Liu, R. Che, H. Chen, F. Zhang, F. Xia, Q. Wu, M. Wang, Microwave Absorption Enhancement of Multifunctional Composite Microspheres with Spinel Fe_3O_4 Cores and Anatase TiO_2 Shells, *Small.*, 8, 2012, 1214-1221.
4. G.S. Bae, C.Y. Kim, Design of Multi-Layer Radar Absorbing Coating and Transparent Shielding Film for RCS Reduction of Complex Targets, *Appl. Mech. Mater.*, 799-800, 2015, 1370-1378.
5. T. Hou, Z. Jia, Y. Dong, X. Liu, G. Wu, Layered 3D structure derived from MXene/ magnetic carbon nanotubes for ultra-broadband electromagnetic wave absorption, *Chem. Eng. J.*, 431, 2022, 133919.
6. M. Wu, A.K. Darboe, X. Qi, R. Xie, S. Qin, C. Deng, G. Wu, W. Zhong, Optimization, selective and efficient production of CNTs/ $\text{Co}_x\text{Fe}_{3-x}\text{O}_4$ core/shell nanocomposites as outstanding microwave absorbers, *J. Mater. Chem.*, 8, 34, 2020, 11936-11949.
7. L. Long, E. Yang, X. Qi, R. Xie, Z. Bai, S. Qin, C. Deng, W. Zhong, Positive and Reverse Core/Shell Structure $\text{Co}_x\text{Fe}_{3-x}\text{O}_4/\text{MoS}_2$ and $\text{MoS}_2/\text{Co}_x\text{Fe}_{3-x}\text{O}_4$ Nanocomposites: Selective Production and Outstanding Electromagnetic Absorption Comprehensive Performance, *ACS Sustain. Chem. Eng.*, 8, 2020, 613-623.
8. C. Sun, Z. Jia, S. Xu, D. Hu, C. Zhang, G. Wu, Synergistic regulation of dielectric- magnetic dual-loss and triple heterointerface polarization via magnetic MXene for high-performance electromagnetic wave absorption, *J. Mater. Sci. Technol.*, 113, 2022, 128-137.
9. B.D. Cullity, C.D. Graham, Introduction to magnetic materials, 2nd ed, IEEE/Wiley, Hoboken, N.J., 2009.
10. Z. Wang, L. Wu, J. Zhou, B. Shen, Z. Jiang, Enhanced microwave absorption of Fe_3O_4 nanocrystals after heterogeneously growing with ZnO nanoshell, *RSC Adv.*, 3, 2013, 3309.
11. J.Y. Shin, J.Y. Oh, The microwave absorbing phenomena of ferrite microwave absorbers, *IEEE Trans. Magn.*, 29, 1993, 3437-3439.

12. S. Kolev, A. Yanev, I. Nedkov, Microwave absorption of ferrite powders in a polymer matrix. *Phys. Stat. Sol. C*, 3, 2006, 1308-1315.
13. S. Kolev, T. Koutzarova, Nanosized ferrite materials for absorption and protection from microwave radiation, in: P. Petkov, D. Tsiulyanu, C. Popov, W. Kulisch (Eds.), *Nanoscience and Nanotechnology in Security and Protection against CBRN Threats*, Springer, Dordrecht, 2018, 273-283.
14. Y. Naito, K. Suetake, Application of ferrite to electromagnetic wave absorber and its characteristics, *IEEE Trans. Microw. Theory Tech.*, 19, 1971, 65-72.
15. D. Guo, W. Kong, J. Feng, X. Li, X. Fan, Microwave absorption properties of $\text{Sr}_x\text{Ba}_{3-x}\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ hexaferrites in the range of 0.1-18 GHz, *J. Alloy. Compd.*, 751, 2018, 80-85.
16. T. Inui, K. Konishi, K. Oda, Fabrications of broadband RF-absorber composed of planar hexagonal ferrites, *IEEE Trans. Magn.*, 35, 1999, 3148.