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Engineering of Ferrite-Graphite Composite Media for Microwave Shields

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Abstract— An electromagnetic shielding of objects using ferrite-graphite composites is considered. The analytical model, using the Maxwell Garnett formulation for multiphase mixtures, results of computations based on this model and plane-wave formulation, and some experimental results are represented.

Keywords- shielding; dielectric base material; ferrite- graphite composite, Maxwell Garnett formulation

I. INTRODUCTION

Materials exhibiting simultaneously significant dielectric and magnetic properties have been of great interest to modern science and technology, not only from the viewpoint of solidstate physics, but also because of their potential for practical applications in electronic devices and systems [1-3]. The rapid evolution of high-speed digital electronics, wireless communications, industrial, medical, and concomitant increasing exposure to microwaves necessitates the development of effective, compact, and economical absorbers of electromagnetic energy to ensure electromagnetic compatibility and ecological safety in a wide frequency range. Ferrites possess a unique combination of high permittivity, spontaneous magnetization, and extremely low d.c. conductivity. These properties make them attractive for developing wideband absorbers [4-9].

The main problem with the development of polymeric composites filled with ferrite powders is how to control frequency dispersion of complex permeability and permittivity of composites. This can be achieved by variation of the composite filler types, size of inclusions, their concentrations, and morphology of mixtures through the alignment of inclusion particles. Attempts are also made to control the magnetic properties of inclusions through the microstructure of composites, etc.

It is known that the absorbing materials containing dispersed hexagonal ferrites (HF) in a dielectric base can effectively absorb electromagnetic energy at microwave frequencies. HF exhibit the phenomenon of natural ferromagnetic resonance (NFMR) [12], and this can be employed effectively without any bias magnetic field. The frequency of the NFRM is proportional to the magnetic field associated with a ferrite's crystallographic anisotropy, which, in turn, depends on the ferrite's chemical structure [13]. Alexander A. Kitaitsev, Andrey A. Shinkov Moscow Power Engineering Institute (Technical University), School for Radio Engineering and Electronics Moscow, Russia e-mail: KitaitsevAA@mpei.ru

Composites can consist of mixtures of dispersed hexagonal ferrites having different chemical contents. Due to the statistical distribution of the values of their fields of crystallographic anisotropy of hexagonal ferrites within the composite, it may be possible to obtain wideband absorption characteristics [14-16]. However, hexagonal ferrites are known to exhibit the phenomenon of NFMR only at frequencies higher than approximately 2.5 GHz (up to 200 GHz), because their internal field of magnetic crystallographic anisotropy ranges from a few units to a few dozens of kilooersted [16]. The reflection coefficient from a metal surface coated with composites containing hexagonal ferrite powders is typically -30...-20 dB at frequencies above 2.5 GHz. At lower frequencies, hexagonal ferrites do not substantially absorb electromagnetic energy, and, as a result, the coated surfaces might have high reflection.

There is another class of ferrites that can be used for absorbing materials - spinel ferrites. They are known to be soft ferrites with high permeability at d.c. and frequencies below of about 100 MHz, depending on the particular composition. Their permeability substantially decreases beyond about 100 MHz due to the dynamics of the domain magnetic walls [17]. These ferrites absorb due to a large imaginary part of the permeability (real and imaginary parts of the permeability as functions of frequency are related by the integral Kramers-Kronig causality relations [18]). The static permeability value and the maximum loss frequency are related according to Snoek's law, limiting the bandwidth for spinels' applications as microwave absorbers [19]. The internal field of crystallographic anisotropy for spinel ferrites ranges from a few units to dozens of oesrteds, i.e., three orders lower than that of hexagonal ferrites. Consequently, the NFMR phenomena are nearly nonexistent for spinels at frequencies exceeding approximately 100 MHz. Neither hexagonal ferrites, nor spinels effectively absorb energy in the frequency range from 100 MHz to 2.5 GHz. However, the substantial absorption in this frequency range can be achieved by increasing dielectric loss in the mixture of ferrites and bond material.

Ponomarenko, *et.al.* [20] have demonstrated the theoretical possibility of modifying both magnetic and dielectric properties of ferrite composites in microwave frequency range

due to metallization of ferrite particles. However, in the same paper [20], the experiments with NiZn ferrite particles electrolytically coated with a Fe skin demonstrate that along with impact of a conducting layer on frequency dispersion of NiZn ferrite particles, there is a substantial decrease of absolute values of both real and imaginary parts of permeability. This is due to the skin-effect in conducting coats, which is for such metals as Fe and Cu in microwave frequency range is about a few micrometers. It is almost impossible to control such thicknesses on the surface of ferrite particles using electrolytical sputtering. However, there is a unique class of conducting polymers, and their conductivity can be varied in a wide range from 10^{-11} to 10 S/m [21]. The technology of coating different inorganic materials with them, including soft magnetic materials, has been developed [22]. Kazantseva, et.al. [23,24] describe the way of synthesizing composite materials using polyconnected polyaniline and polycrystalline particles of MnZn ferrite, and present some experimental data on dielectric and magnetic properties of such composites in the frequency range from 1 MHz to 10 GHz.

Herein, the dielectric loss in composites containing NiZn and MnZn ferrites is increased by adding conducting particles in the mixture. These are carbon inclusions. The mechanism of absorption in such a composite is the following. Supposedly, the energy absorption process happens in two stages: first, electromagnetic energy is concentrated in the absorbing layer, and second, it dissipates. Electromagnetic energy concentrates due to the high magnetic μ' properties of ferrite inclusions and due to the polarization of conductive particles in the dielectric base, leading to high effective permittivity of the mixture ε' . The energy dissipates mainly because of high \mathcal{E}'' of the mixture in the frequency range of interest, due to the conductivity of carbon inclusions. Thus, the energy is absorbed synergistically by the mixture of ferrite and carbon particles, but not by ferrite and by carbon as separate phases. The experiments proving these speculations have been conducted, and the results are described below. Also, the analytical model, based on the Maxwell Garnett mixing formulation [25-27] and the results of computations are presented.

II. FORMULATION FOR FREQUENCY DEPENDENCES OF MIXTURES CONTAINING FERRITE AND GRAPHITE INCLUSIONS

Composite media at comparatively low concentrations of inclusions are successfully described by the Maxwell Garnett (MG) model [25]. Herein, the generalized MG mixing formula for multiphase mixtures with randomly oriented ellipsoidal inclusions is used [26, 27],

$$\varepsilon_{eff} = \varepsilon_b + \frac{\frac{1}{3} \sum_{i=1}^n f_i (\varepsilon_i - \varepsilon_b) \sum_{j=1}^3 \frac{\varepsilon_b}{\varepsilon_b + N_{ij} (\varepsilon_i - \varepsilon_b)}}{1 - \frac{1}{3} \sum_{i=1}^n f_i (\varepsilon_i - \varepsilon_b) \sum_{j=1}^3 \frac{N_{ij}}{\varepsilon_b + N_{ij} (\varepsilon_i - \varepsilon_b)}}, \qquad (1)$$

where ε_b and ε_i are the relative permittivities of a base dielectric and the *i*-th type of inclusions, respectively; f_i is the volume fraction occupied by the inclusions of the *i*-th type; N_{ij} are the depolarization factors of the *i* -th type inclusions [28], and the indices j=1,2,3 correspond to x,y, and zcoordinates. If the inclusions are thin cylinders, their two depolarization factors are close to 1/2, and the third can be calculated as in [29], $N \approx (1/a)^2 \ln(a)$, where a = l/d is the aspect ratio of cylinders.

The conducting (herein, carbon) particles with conductivity σ_c , have the complex relative permittivity

$$\varepsilon_{c}(j\omega) = \varepsilon' - j\varepsilon'' = \varepsilon' - j\frac{\sigma_{c}}{\omega\varepsilon_{0}}.$$
 (2)

The MG mixing rule is applicable, when the concentration of the conducting particles in the mixture is below the percolation threshold, $p_c \cong 4.5/a <<1$ [30].

The base material might be quite transparent over the frequency range where high shielding effectiveness is desired. The behavior of many polymeric materials in the microwave range can be described by the Debye frequency dependence,

$$\varepsilon_{b}(j\omega) = \varepsilon_{\infty b} + \frac{\varepsilon_{sb} - \varepsilon_{\infty b}}{1 + j\omega\tau_{b}}.$$
(3)

Ferrite inclusions have frequency dispersion of both permeability and permittivity. Our experimental studies of frequency dependences of real and imaginary parts of complex permittivity and permeability of bulk NiZn ferrite have shown that they behave according to the Debye law [31].

Suppose that the magnetic susceptibility of a bulk polycrystalline ferrite $\chi_{fer \ bulk} = \mu_{fer \ bulk} - 1$ is known. Let it be powdered in such a way that any particle of ferrite is not smaller than a single magnetic domain, and all the magnetic moments are 3D randomly oriented. If the volume fraction of ferrite in a mixture with non-magnetic phase(s) is f_{fer} , then the effective magnetic susceptibility of the mixture is calculated as [32]

$$\chi_{eff} = \frac{2}{3} f_{fer} \chi_{fer \ bulk} = \mu_{eff} - 1.$$
(4)

III. MODELING RESULTS

Teflon was chosen as the base material for modeling. In the frequency range of interest (up to 10 GHz) it may be considered as nondispersive material with $\mathcal{E}'_b = 2.2$ (loss tangent is smaller than $3 \cdot 10^{-4}$) [33]. The commercially available carbon inclusions for the modeling are taken with the

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following parameters: conductivity $\sigma_{carb} = 1000$ S/m, aspect ratio of cylindrical particles a = 50, and volume fraction $f_{carb} = 0.08$ (8%). High-frequency NiZn ferrite, for example, the type 45BHP manufactured by *Ferrite Domen* (Russia) [34], used in switching devices, has $\mu_s = 45$ with cutoff frequency $f_c = 80$ MHz. Its dielectric frequency characteristic is close to that obtained in [32], with $\varepsilon_{s fer} = 14$, $\varepsilon_{\infty fer} = 6$, and $\tau_{fer} = 7.95 \cdot 10^{-11}$ s. The resulting effective permittivity and permeability versus frequency are shown in Figures 1 and 2. Shielding effectiveness of 1-cm thick slab of composites: ferrite-graphite-Teflon and just graphite-Teflon obtained in a plane-wave formulation is shown in Figure 3.



Figure 1. Relative permittivity of ferrite-graphite mixture in Teflon base.



Figure 2. Relative permeability of ferrite-graphite mixture in Teflon base.



Figure 3. Shielding effectiveness for a slab of a composite material: 1- mixture ferrite-graphite-Teflon, and 2 - mixture of graphite and Teflon.

As seen from this figure, the presence of ferrite inclusions substantially increases S.E. The shortcoming of adding ferrite inclusions is the increase of the composite density and total mass of an enclosure made of it, since the ferrite density is about 4.8 g/cm³ versus carbon density of 2.27 g/cm³.

IV. EXPERIMENTAL RESULTS

The experimental research of graphite-ferrite and graphite mixtures in the paraffin base was carried out in the Moscow Power Engineering Institute (Technical University). Paraffin in the frequency range below 10 GHz has dielectric properties very close to those of Teflon: its complex permittivity is $\mathcal{E}_b = 2.25$ -j5.63·10⁻⁴ at *f*=10 GHz [35,36]. Density of paraffin is 0.9 g/cm³.



Figure 4. Absorption in different composite samples placed in coaxial fixture.

Three samples having the shape of washers were made: ferrite-paraffin, graphite-paraffin, and ferrite-graphite paraffin.

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Size of ferrite and graphite particles was approximately the same, in the range $60-90 \,\mu m$, and their volume fractions in the sample were approximately the same. Mass concentration of MnZn ferrite (type 3000HM, with $\mu_s = 3000$ and $f_c = 2$ MHz [34]) was 70%, and of carbon 5%. The samples were placed in a coaxial fixture with the diameters 16 mm x 7 mm. Volume fractions of phases in the 14-mm long washer made of graphite-ferrite-paraffin mixture were $f_{fer} = 32.8\%$, $f_{gr} =$ 4.9%, and $f_{par} = 62.3\%$, respectively. The ferrite-paraffin sample was made with exactly the same mass of ferrite (it is known that absorption is proportional to the total mass of ferrite), and the graphite-paraffin sample contained the same mass of carbon as the first sample. The lengths of the sample, hence, were slightly different. Transmission coefficient (in terms of power) and VSWR in the line were measured, and then the absorbed power $P_{abs} = P_{inc} - P_{tr} - P_{refl}$ in the calculated. The samples was absorption $A = -10\log_{10}(P_{abs} / P_{inc})$ as a function of frequency for the samples is represented in Figure 4. It is seen that the graphiteparaffin sample (line marked by circles) is slightly absorbing. Most of the absorption is assured by the presence of a dispersed ferrite (lines marked by stars and crosses). The line marked by triangles corresponds to the case when two washers - ferrite-paraffin and graphite-paraffin - were placed one after another, with a ferrite-paraffin washer first to the incoming wave. It is interesting that the absorption by the washer made of the mixture of ferrite and graphite absorbs about 4 times more than the cascade of two separate ferrite-paraffin and graphite-paraffin washers. This proves that the mechanism of absorption is due to the concentration of energy by carbon inclusions and absorption by ferrite particles.



Figure 5. Transmission coefficient in cables coated with composites hexagonal ferrite-graphite.

Figure 5 shows the transmission coefficient in the coaxial fixture made of Sc-doped BaM-type hexagonal ferrite and graphite, effectively absorbing in the frequency range 8-18 GHz. The dimensions of the fixture and concentrations of

hexagonal ferrite and graphite were the same as described above.



Figure 6. Transmission coefficient in cables coated with ferrite-graphite-paraffin



Figure 7. Intensity of radiation: 1- cable with two copper shields; 2- cable with a ferrite-graphite-paraffin layer between copper shields.

Another experiment was conducted by coating the central conductor of coaxial cables by ferrite-graphite composite layers and measuring transmission coefficient through the cable. Different powders of ferrites: 400HM, 3000HM, and 6000HM were used to make a composite. Length of the cable was 45 cm, the diameter of the central conductor was 1.35 mm, and the diameter of the outer conductor was 4 mm (400HMd4, 3000HMd4, 6000HMd4) or 6 mm (3000HMd6). Thickness of the composite layer was 2 mm (400HMd4, 3000HMd4, 6000HMd4) and 3 mm (3000HMd6). The concentration of ferrite and graphite was as mentioned above. The measured transmission coefficient in the frequency range below 1 GHz for different types of ferrites are shown in Figure 6. The main oscillation is transmitted without attenuation, while the unwanted higher-order harmonics are absorbed. Thus, a low-pass filter has been realized.

Figure 7 demonstrates the effectiveness of placing a composite layer between the copper shields around the central conductor. Radiation intensity versus distance from the cable

was measured for two cables: one with two copper shields around the central conductor, and the second having a 2-mm thick layer of ferrite (3000HMd4) – graphite composite. Presence of the ferrite-graphite composite layer leads to decrease of radiation from the cable at least 10 dB in all the points along the cable length.

V. CONCLUSION

Ferrite-graphite composites considered herein can be useful for shielding purposes, including development of low-pass filters based on coaxial cable structures. Long structures, such as coaxial cables, containing ferrite-graphite coating between metal shields, have shown substantial reduction of radiated emission. The mechanism of absorption in ferrite-graphite composites is due to two effects – concentration of electromagnetic energy in a dielectric base due to polarization of carbon particles, and energy absorption by ferrite inclusions.

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