Electromagnetic Properties of Hexaferrite Polymer Composite Materials

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Abstract. In this paper the results of studies of the electromagnetic characteristics of composite radio materials based on Z-, W-, M-type barium ferrites and epoxy resin are presented. The frequency dependences of the reflection, transmission, and absorption coefficients are shown. The obtained results of the electromagnetic response are in good agreement with the published data on the frequencies of natural ferromagnetic resonance for the types of ferrites considered. Based on the obtained data, the spectra of complex dielectric and magnetic permeabilities were calculated. It has been suggested that the above changes in the complex magnetic permeability are associated with the presence of domain wall resonance in the region of 1 GHz and natural ferromagnetic resonance at higher frequencies.

1. Introduction
Modern radio equipment and electronics are widely used in everyday life. So, various wireless transceivers are widely used. These are Bluetooth, Wi-Fi routers and adapters, cell phones, smart home appliances and equipment (IoT-devices), these are all kinds of devices and systems used for geopositioning and tracking of targets, radar stations, monitoring and control systems for robotic complexes. The frequency range of operation of such devices is very often in the range of microwave radiation [1-3]. But the use of this equipment creates a number of negative factors, such as a threat to biological objects, problems of electromagnetic compatibility and safety, as well as "pollution" of the general electromagnetic background. Composite materials that act as absorbers of microwave energy or shielding materials are widely used to solve this type of problem [4, 5]. Hexagonal ferrimagnets of various types are quite widely used as fillers for radio composites in the microwave range [6, 7]. Therefore, the study of the properties of such materials remains an urgent task.

2. Materials and Methods
2.1. Materials
The choice of matrix or binder composite is based on its purpose. Various types of sealants, varnishes, paints, epoxies, as well as rubbers of various types can act as a polymer matrix for samples of composite material. In our case, the base was an epoxy resin EDP-20 with a hardener made of polyethylene polyamine (PEPA). Barium ferrite powders were used as the active phase: BaAlFe$_{11}$O$_{19}$ (M type), BaCo$_{0.6}$Zn$_{1.4}$Fe$_{16}$O$_{27}$ (W type), BaCo$_{0.7}$Zn$_{1.3}$Fe$_{16}$O$_{27}$ (W type), BaCo$_{2.4}$Ti$_{0.4}$Fe$_{23.2}$O$_{41}$ (Z type).
Ferrites were obtained using SHS technology, followed by firing at 1200 °C and processing in a planetary mill. After grinding, the particle size was less than 100 microns.

2.2. Obtaining experimental samples
The manufacture of samples of composites was carried out according to the traditional technology of mixing the starting components. The sample production scheme is shown in Figure 1. After selecting the filler, we carefully weighed the components of the composite on a Shimadzu AUX-320 balance (error ~ 0.5 mg). This was followed by mixing the mixture to a homogeneous state with simultaneous sonication with a power of 20 W for 1 minute. Then, the resulting mixture was poured into special molds and polymerized at room temperature. After polymerization, the samples were machined with grinding machines to give the desired geometric shape. As a result, experimental samples were obtained in the form of washers with an inner diameter $d_{in} = 3$ mm, external diameter $d_{ext} = 7$ mm and thickness $h = 2.25$ mm.

![Production of samples of composite materials.](image)

Thus, 4 experimental samples were prepared. The samples contained 40 wt.% epoxy resin and 60 wt.% ferrite powders: $\text{BaAlFe}_{11}\text{O}_{19}$, $\text{BaCo}_{0.6}\text{Zn}_{1.4}\text{Fe}_{16}\text{O}_{27}$, $\text{BaCo}_{0.7}\text{Zn}_{1.3}\text{Fe}_{16}\text{O}_{27}$, $\text{BaCo}_{2.4}\text{Ti}_{0.4}\text{Fe}_{23.2}\text{O}_{41}$.

2.3. Measuring equipment
The electromagnetic properties of the obtained composites were measured by the waveguide method with the inclusion of a coaxial cell with an inner diameter $d_{int} = 3.05$ mm and external diameter $d_{ext} = 7$ mm. The vector network analyzer P4M-18 produced by the company “Micran” acted as a measuring installation. The measurements were carried out according to the “for pass” scheme (Figure 2). As a result, we obtained the family of frequency dependences of the reflection coefficients ($R$), transmission ($T$), and absorption ($A$) in the frequency range from 10 MHz to 18 GHz.

![Measuring installation: a) vector network analyzer P4M-18; b) Schematic diagrams of measurements "on pass".](image)
The “on pass” (Figure 2b) measurement scheme allows you to measure the values of the coefficients of the scattering matrix. In other words, to determine the value of parameters $S_{11}$, $S_{12}$, $S_{21}$, $S_{22}$. These $S$-parameters determine the coefficients of reflection and transmission of an electromagnetic wave through a sample placed in the measuring cell, with forward direction and reverse passage. This device (Figure 2a) allows you to track the phase change of the obtained coefficients of the scattering matrix. Based on the measured $S$-parameters, as well as their phase, it is possible to calculate the spectra of complex magnetic and dielectric constants.

3. Results and discussion

Figure 3 shows the frequency dependences of the reflection coefficients ($R$), transmission ($T$), and absorption ($A$) for the studied composite samples.

![Composite materials](image)

**Figure 3.** Frequency dependences of the reflection coefficients ($R$), transmission ($T$), and absorption ($A$) for composite materials based on epoxy resin and ferrites.

Analyzing the obtained dependences in Figure 3, it can be noted that with an increase in the frequency for all samples, a decrease in the transmission coefficient is noticeable. The sample containing $\text{BaAlFe}_{11}\text{O}_{19}$ powder has the lowest shielding level (up to 65%). The samples with $\text{BaCo}_{0.6}\text{Zn}_{1.4}\text{Fe}_{23.2}\text{O}_{41}$ and $\text{BaCo}_{2.4}\text{Ti}_{0.4}\text{Fe}_{23.2}\text{O}_{41}$ have the maximum of shielding level (up to 45%). This is due to an increase in the reflection and absorption coefficients. Thus, with increasing frequency, an almost linear increase in the reflection coefficient is observed. Moreover, for all ferrites, its value is
approximately the same, this is due to the same thickness of the samples and close electromagnetic parameters. The highest absorption at frequencies up to 10 GHz is possessed by a sample containing BaCo$_{0.6}$Zn$_{1.4}$Fe$_{16}$, and above 10 GHz a sample with BaCo$_{2.4}$Ti$_{0.4}$Fe$_{23.2}$O$_{41}$, whose absorption coefficient for this reaches 27%. The lowest absorption coefficient has a sample based on BaAlFe$_{19}$O$_{19}$ and reaches 2-3%. This behavior of the electromagnetic response is in good agreement with published data on the frequencies of natural ferromagnetic resonance (NFMR) for the considered types of ferrites.

Based on the measurements of the $S$-parameters, the complex permittivity and permeability were calculated. The calculation of electromagnetic parameters was carried out according to the Becker-Jarvis method using a program written in the Mathcad environment, based on the content of the article “Wideband Reference-Plane Invariant Method for Measuring Electromagnetic Parameters of Materials” [8]. So, Figure 4 shows the frequency dependences of the complex dielectric and magnetic permeabilities for samples of composites based on epoxy resin and ferrimagnets.

![Figure 4](image)

**Figure 4.** Frequency dependences of permeability (a) and permittivity (b) for a composite material based on epoxy resin and ferrites.

Analyzing the spectra of complex permittivity and magnetic permeability, it can be noted that at low frequencies, the highest $\varepsilon'$ and $\varepsilon''$ values belong to BaCo$_{0.6}$Zn$_{1.4}$Fe$_{16}$, reaching $\varepsilon' = 5.6$ rel. units and $\varepsilon'' = 0.6$ rel. units. At frequencies above 4 GHz all the materials under consideration have a similar dependence and the value of $\varepsilon'$ belongs in the range from 4 rel. units up to 5 rel. units, and $\varepsilon''$ up to 0.5 rel. units. On the frequency dependences of the complex magnetic permeability, there is a decrease in the real part and maxima on the imaginary part. Such changes in the complex magnetic permeability are probably associated with the presence of resonance of domain walls in the region of 1 GHz and natural ferromagnetic resonance at higher frequencies.

4. Conclusions

From the obtained spectra of the complex dielectric and magnetic permeabilities, W-type hexaferrites are more preferred to use in the microwave range at frequencies up to 5 GHz, because the natural ferromagnetic resonance of these materials is in this frequency range. From 6 to 12 GHz, Z-type hexaferrites demonstrate the better properties than W-type hexaferrites. M-type hexaferrite works at higher frequencies, which follows from the literature on its natural ferromagnetic resonance, which should be above 50 GHz for this structure.
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