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Estimation of the Superhigh-Frequency Magnetic Permeability of Alsifer from the Measured Permeability of Composites

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Abstract—The magnetic permeability of alsifer was restored from the frequency dependences of the dielectric and magnetic permeabilities of powder alsifer (AlSiFe alloy)—wax matrix composites. The permeabilities were measured using the coaxial line technique within a frequency range of 0.05–20 GHz. The effect of the concentration, shape, and size of powder particles on the microwave magnetic properties of composites was considered. A good agreement between the measurement results and the Maxwell—Garnett formula generalized with consideration for the particle shape, the percolation threshold, and the skin-effect was obtained. The found sizes of particles agreed with electron microscopy and granulometry data. Both the frequency and the ferromagnetic resonance line figure of merit (FOM) for lamellar particles proved to be higher than for spherical ones. Alsifer powders were shown to be promising fillers for radioabsorbing materials.

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1. INTRODUCTION

When designing different high-frequency (HF) and superhigh-frequency (SHF) devices, it is necessary to know the magnetic properties of the materials used. Electromagnetic fields can penetrate into metals only to the skin-layer depth, which attains several fractions of a micron at superhigh frequencies, so the frequency dependences of the magnetic permeability μ of metals were estimated only from the properties of thin films until now.

There were several known attempts to estimate the SHF magnetic permeability of a metal from the losses in metallic grids [1], the dependence of the resonator FOM on the external magnetic field [2], and the losses in the coaxial line of transmission from a studied material [3]. In these works, different metals were studied, and the obtained estimates of u corresponded to isolated widely spaced frequency points and poorly agreed with each other. Some estimates of the HF permeability of a ferromagnetic from the measured magnetic permeability of a composite filled with dispersed metal particles (two-phase powder filler-polymeric binder mechanical mixture) appeared at a later date due to the development of the theory of effective properties of inhomogeneous materials. Thus, the magnetic permeability of iron was estimated from the measured permeabilities of composites with spherical iron particles in [4]. This estimate was obtained using the Maxwell–Garnett formula [5]. As a result, it has been found that the estimated magnetic permeability of a filler depends on the concentration of inclusions in the composite selected for the analysis of data. It is obvious that this result lies outside the limits of applicability for the used mixing formula.

In the work [6], the intrinsic magnetic permeability of alsifer particles was restored from the measured permeability of composites filled with particles of this alloy using the Wiener approximation [7] applicable only at a small difference between the properties of mixture components.

There also exist some known works [8, 9], in which similar estimates were obtained using the elective medium theory (EMT or the generalized Bruggeman formula for a symmetric mixture [10]). The correctness of application was substantiated for the EMT formula in these works only by the similarity of the measured concentration dependences of the static magnetic permeability of a composite to the curves predicted by the selected mixing formula. However, the obvious unfeasibility of the simultaneous description of the effective material parameters, dielectric permeability ε_{mix} , and magnetic permeability μ_{mix} of a composite with the selected mixing formula was ignored in this case.

An obvious shortcoming of the known attempts to determine the magnetic permeability of a metal from

the permeability of composites with its dispersed particles is the arbitrary selection of a mixing formula and the estimation of the formal parameters of this formula instead of the properties of particles. For this reason, the obtained results are determined not so much by the properties of metals as by the form of a used mixing formula. In this work, the mixing formula simultaneously describes the dependences of ε_{mix} and μ_{mix} on the composition of a composite, and its applicability is confirmed by comparing its experimentally determined parameters with the results of their estimation by independent methods.

The precise and most correct approach to finding the relation between the permeabilities of a heterogeneous system and its components is described by the Bergmann–Milton theory [11] through the characteristic spectral function related with the shape of inclusion clusters. The determination of this function form requires essential efforts for each new sample of mixtures, so the spectral function has not found practical application yet.

The paper [12], which is most closely allied with this work, is devoted to the derivation of a hybrid mixing formula, which takes into account the dependence of the shape distribution of ferromagnetic metal particles on their concentration. This formula describes the structural transformation of a mixture from a matrix mixture to a symmetric one upon attainment of the percolation threshold. The applicability of the expression derived in [12] is limited by near-spherical particles.

Below, the frequency dependence of the SHF magnetic permeability of a metal was estimated from the results of measurements for the frequency dependences of the effective material parameters ($\epsilon_{\mbox{\scriptsize mix}}$ and μ_{mix}) of composites filled with a studied highly dispersed metal powder. The selected object of study was alsifer $(Al_{0.054}Si_{0.096}Fe_{0.85})$, which is a promising filler for SHF composites. The dispersion dependences of the material parameters of wax-based matrix mixtures with a varied concentration of spherical or lamellar alsifer particles of known size were measured. The dependences of the material parameters of the studies composites on the volumetric concentration of inclusions p were described at each frequency using the modified Odelevsky formula [13, 14], which takes into account the shape of particles and the structural transformation of a mixture upon attainment of the percolation threshold. The Odelevsky formula more precisely takes into account the interaction between metallic inclusions in a mixture than the Maxwell-Garnett formula [5].

The applicability of the Odelevsky formula relating ε_{mix} and μ_{mix} of a two-component mixture with the corresponding parameters of filler particles ε_{incl} and μ_{incl} and a matrix ε_{host} and $\mu_{host} = 1$ is confirmed by the absence of dependence between the experimentally determined theory parameters (the inclusion shape determining the depolarization coefficient *N* and the

critical concentration of inclusions p_c) and the contrast between the matric and filler permeabilities μ_{incl}/μ_{host} . When the frequency *f* is changed, the inclusion permeability μ_{incl} variates due to frequency dispersion within broad ranges, and obtained *N* and p_c remain constant. If the used mixing formula did not correspond to the properties of the studied composites, there would be either a deviation between the calculated and measured dependences $\varepsilon_{mix}(p)$ and $\mu_{mix}(p)$ or an appreciable dependence of the mixing formula parameters on the contrast μ_{incl}/μ_{host} and, correspondingly, the frequency.

The measured set of the material parameters of a composite $\mu_{mix}(f, p)$ and $\varepsilon_{mix}(f, p)$ is used to determine the parameters N and p_c for each frequency f at different inclusion concentrations p. The found parameters are averaged over the frequency to decrease the contribution of error in the measured material parameters of mixtures. The calculated average parameters N and p_c and the measured values of $\mu_{mix}(f, p)$ are used to determine the frequency dependence of the effective magnetic permeability of a metal inclusion $\mu_{incl}(f)$. Using the known alloy conductivity and the inclusion sizes (estimated microscopically and refined with the depolarization coefficient N), the skin-effect contribution to $\mu_{incl}(f)$ is excluded, and the sought frequency dependence of the magnetic permeability of a metal inclusion $\mu_{metal}(f)$ is calculated. The derived frequency dependences $\mu_{\text{metal}}(f)$ for spherical and lamellar particles are approximated to estimate the ferromagnetic resonance parameters by the Lorentz line technique. Lamellar particles are approximated with an oblate spheroid in the process of calculation.

2. TECHNIQUE OF EXPERIMENT AND RESULTS

The alsifer powder used for the preparation of mixtures was obtained via melt aquaspraying. The granulometric composition of powders was measured on an Analyzette 22 laser analyzer. The initial powder is spherical with the most probable particle diameter of $4.2 \,\mu\text{m}$. The half-height granulometric density distribution width is $2-8 \,\mu\text{m}$.

Lamellar alsifer particles, whose electron microphoto is shown in Fig. 1, were obtained by grinding spherical particles in a ball mill in an ethanol medium. After grinding, the particles were classified on sieves, and the fraction between the sieves with a mesh of 63- and 40- μ m was selected for the preparation of mixtures; in this case, the most probable equivalent diameter of a lamella was nearly 50 μ m. The microscopically estimated thickness of particles was nearly 1 μ m.

The composites for the manufacturing of specimens were prepared by mixing together the calculated portions of powder alsifer and melted wax. Mixing was



Fig. 1. Electron microphoto of lamellar alsifer particles obtained by grinding spherical particles in a ball mill.

performed under cooling until the solidification of wax. Powder alsifer was with gasoline to improve distribution uniformity. The mass obtained after solidification was pressed as 2–4-mm thick disks (to fit the standard size of the coaxial line with a cross section of 7×3 mm). The volumetric metal concentration was monitored by measuring the density of disks with an error of 0.5–1%. The maximum volumetric content of metal in thus obtained specimens was nearly 65 and 18% for spherical and lamellar particles, respectively.

To measure the frequency dependence of the effective material parameters, the disks were placed into a feedthrough coaxial cell, where they were additionally pressed to decrease the air gaps, which exist between a specimen and the cell electrodes and invalidate the measured dielectric permeability of a specimen. The material parameters of mixture samples were measured by the transmission/reflection method [15] within a frequency band of 0.05–20 GHz on an Anritsu MS2028 vector network analyzer.

The dielectric permeability of mixture samples is nearly constant throughout the entire studied frequency range and may be considered as the static dielectric permeability. The measured dependences of the dielectric permeability of mixtures on the volumetric concentration $\varepsilon_{mix}(p)$ of metal spheres and lamellae are shown in Fig. 2.

The measured frequency dependence of the effective magnetic permeability of composites at different volumetric concentrations of spherical particles is shown in Fig. 3. Similar data for the composite filled with lamellar particles are given in Fig. 4.



Fig. 2. Static dielectric permeability of mixtures filled with lamellar (circles 1-5) and spherical (rhombs 6-11) alsifer particles versus their volumetric concentration.

3. MODIFIED ODELEVSKY FORMULA. CONSTRAINTS FOR THE ESTIMATED PERMEABILITY OF FILLER PARTICLES

The Odelevsky formula for matrix mixtures describes a broad range of transfer phenomena (generalized conductivity [14]) in two-component matrix



Fig. 3. Magnetic permeability (real (solid) and imaginary (dashed) parts) of composites filled with spherical alsifer particles versus frequency at a volumetric concentration of spheres p = 0.124, 0.26, 0.36, 0.48, 0.58, 0.63 (curve numbers correspond to point numbers in Fig. 2).



Fig. 4. Magnetic permeability (real (solid) and imaginary (dashed) parts) of mixtures filled with lamellar alsifer particles versus frequency at a volumetric concentration of lamellae p = 0.023, 0.041, 0.077, 0.122, 0.157. (curve numbers correspond to point numbers in Fig. 2).

heterogeneous systems (mechanical mixtures). According to the definition [14], a matrix mixture is composed of a matrix (continuous medium) and a filled (isolated inclusions spaced apart by matrix interlayers). In electrodynamics, the Odelevsky formula representing a generalization of the Maxwell–Garnett formula [5] (in the original paper [5], derivation was performed only for spheres, and the correction for the shape of inclusion was introduced for the first time in [14]) is used to describe the electric and magnetic polarization or, correspondingly, the dielectric and magnetic permeabilities of a mixture ε_{mix} (Eq. (1)) and μ_{mix} (Eq. (2)) as

$$\varepsilon_{\rm mix} = \varepsilon_{\rm host} \left[1 + \frac{p}{\left(1 - \frac{p}{p_c}\right)N + \frac{\varepsilon_{\rm host}}{\varepsilon_{\rm incl} - \varepsilon_{\rm host}}} \right], \quad (1)$$

$$\mu_{\rm mix} = 1 + \frac{p}{\left(1 - \frac{p}{p_c}\right)N + \frac{1}{\mu_{\rm incl} - 1}}.$$
 (2)

Equations (1) and (2) differ from each other in that the dielectric permeability of a composite ε_{mix} is determined not only by the filler permeability $\varepsilon_{\text{incl}}$, but also by the matrix permeability $\varepsilon_{\text{host}}$ (for wax, $\varepsilon_{\text{host}} = 2.18$). In these equations, the subscripts mix, host, and incl denote a mixture, a matrix, and inclusions, respectively, *N* is the depolarization (demagnetization) coefficient depending on the inclusion shape [16], p is the volumetric content of inclusions, and p_c is the percolation threshold (critical concentration of inclusions, at which they form an infinite percolation channel (cluster), and the structure of a mixture is transformed from a matrix into a symmetric structure, in which both components are equivalent and form mutually penetrating channels).

The depolarization coefficient for oblate spheroids (disks) at $d/D \le 1$, where D is the greater diameter, and d is the thickness, is given by the formula [17]

$$N = \frac{1}{22 - 2(d/D)^{2}} \times \left[1 - \frac{d/D}{\sqrt{1 - (d/D)^{2}}} \arccos(d/D)\right].$$
(3)

From Eq. (3) it follows that $0 \le N \le 1/3$.

In our calculations, lamellar alsifer particles were approximated with uniformly shaped oblate spheroids. For this reason, the polarizability of lamellae perpendicular to the field is close to zero (the depolarization coefficient is close to unity, i.e., $N \rightarrow 1$), and the effective concentration of lamellae in the isotropic composite (the method used to prepare the specimens gives an isotropic mixture) is 2/3 of the concentration determined from the density of specimens.

In Eqs. (1) and (2), N and p_c are experimentally determined parameters. Theoretically, the depolarization coefficient governed only by the inclusion shape can be estimated by Eq. (3) from the microphotos of filler particles, but the particle size and shape obtained from them is only estimative. In addition, a polydisperse mixture always contains clusters composed at least of two neighbors in real practice even in the case of ideally spherical inclusions, thus leading to a certain decrease in the effective depolarization coefficient of a particle even in a dilute mixture in comparison with its theoretical estimate.

The critical concentration p_c is a parameter characterizing the interaction between inclusions. At $p_c \rightarrow \infty$, there is no interaction between filler particles. At $p_c = 1$, only dipole interaction between particles is taken into account, and Eqs. (1) and (2) are reduced to the Maxwell–Garnett formula [5]. In real practice, $0 < p_c < 1$, and the more appreciable is the distinction of a filler particle from a sphere, the lower is p_c . Let us note that no direct relation $p_c \approx N$ as in the symmetric Bruggeman formula [10] is observed for the studied specimens.

Equations (1) and (2) impose some constraints on the possibility to determine the permeability of inclusions from the measured permeability of a composite. Thus, if the permeability of inclusions is high, the second summand in the denominator of Eqs. (1) and (2) is close to zero, and the permeability of a mixture is determined only by the shape of filler particles and the interaction between them. Just a greater contrast $|\varepsilon_{incl}/\varepsilon_{host}| \sim 10^9$ between the filler and binder permeabilities enables the more precise estimation of *N* and p_c from the experimental data on the concentration dependence of the dielectric permeability of a mixture with metal inclusions $\varepsilon_{mix}(p)$ in comparison with their estimation from the concentration dependence of the magnetic permeability. Due to a great contrast, Eq. (1) is simplified as

$$\varepsilon_{\text{mix}} \approx \varepsilon_{\text{host}} \left| 1 + \frac{p}{\left(1 - \frac{p}{p_c}\right)N} \right|.$$
 (4)

In this case, the effective dielectric permeability of a composite depends only on three factors, such as the volumetric concentration p, the inclusion shape described by the depolarization coefficient N, and the inclusion interactions described by the multiplier $(1 - p/p_c)$. The properties of inclusions do not almost produce any effect on the permeability of a mixture.

At a small contrast between the inclusion and matrix permeabilities, e.g., at frequencies above the ferromagnetic resonance frequency, where $\mu_{incl} \sim 1$, the first summand in the denominator of Eqs. (1) and (2) becomes much lower than the second one and, otherwise, the mixture permeability ceases to depend on *N* and p_c and, according to the Wiener formula [7], is governed only by the permeability and concentration of inclusions, i.e., $\mu_{mix} \approx 1 + p(\mu_{incl} - 1)$.

Hence, the analysis of the concentration dependences of the magnetic permeability of matrix mixtures filled with magnetic powders provides the estimation of the inclusion permeability μ_{incl} from the mixture permeability μ_{mix} only within a range depending on the shape and concentration of filler particles. Differentiating Eq. (2), let us determine the effect of the depolarization coefficient N and the filler concentration p on the inclusion permeability estimation error $\Delta\mu_{incl}$ at a certain specimen permeability measurement error $\Delta\mu_{mix}$ specified by experimental conditions:

$$\Delta \mu_{\text{incl}} = \frac{1}{p} \left[(\mu_{\text{incl}} - 1) \left(1 - \frac{p}{p_c} \right) N + 1 \right]^2 \Delta \mu_{\text{mix}}.$$
 (5)

From Eq. (5) it follows that the inclusion permeability estimation error $\Delta \mu_{incl}$ is proportional to the dilution of a composite ($\Delta \mu_{incl} \sim 1/p$), the squared depolarization coefficient N^2 , and the squared effective magnetic permeability of inclusions μ_{incl}^2 .

Hence, the estimation of the magnetic permeability of inclusions μ_{incl} by the Maxwell–Garnett model (Eq. (2) at $p_c = 1$) has a satisfactory precision only in the region of frequencies, where μ_{incl} is rather low. To determine higher μ_{incl} , it is necessary to measure the permeability of composites with concentrations, which lie near the percolation threshold and can be described by the Odelevsky formula (Eq. (2)), which more correctly takes into account the interaction between inclusions.

When measuring only the magnetic permeability μ_{mix} , it is necessary to determine three parameters at once from an array of experimental data, such as the demagnetization coefficient *N*, the critical concentration p_c , and the sought inclusion permeability μ_{incl} . Such a measurement represents a complicated experimental problem, which can be simplified only by engaging some additional composite structure data.

In the paper [13], it is proposed to determine the depolarization coefficient N from the dielectric permeability of a composite ε_{mix} (where the contrast of permeabilities is high, i.e., $\varepsilon_{incl}/\varepsilon_{host} \rightarrow 10^9$) with a minimum filler content. The magnetic permeability μ_{mix} is also measured at contents $p \ll p_c$ to eliminate the effect of interaction between inclusions. The method [13] imposes high requirements to the precision of measurements for ε_{mix} , as the depolarization coefficient N is determined by the difference between two close parameters, such as the matrix permeability ε_{host} and the mixture permeability ε_{mix} with a low filler content.

In contrast to the work [13], the parameters N, p_c , and the inclusion permeability μ_{incl} as such are found here simultaneously by minimizing the sum of the mean-square deviation between the permeabilities $\varepsilon_{mix}(p)$ and $\mu_{mix}(p, f)$, which are calculated by Eqs. (2) and (3) and obtained as a result of measurement.

In this case, experimental data are processed for particles with the same shape over all the concentrations at each frequency of measuring the material parameters $\varepsilon_{mix}(p)$ and $\mu_{mix}(p)$. In contrast to the method [13], the reliable estimation of *N* and p_c by this technique requires composite permeability data from as broad a range of concentrations as possible.

At specified measurement errors $\Delta \varepsilon_{mix}$ and $\Delta \mu_{mix}$, the maximum inclusion permeability μ_{incl} estimated by this technique is limited by the possibility to reliably distinguish the dielectric and magnetic permeabilities of a composite $\varepsilon_{mix}/\varepsilon_{host} - 1$ and $\mu_{mix} - 1$. Correspondingly, as follows from Eq. (5), the closer is the concentration of inclusions *p* to the percolation threshold p_c , the higher values of μ_{incl} can be determined.

4. PROCESSING OF EXPERIMENTAL DATA. CALCULATION OF THE MAGNETIC PERMEABILITY OF A PARTICLES AND ITS METAL

The above-described approach and measured $\varepsilon_{mix}(p)$ and $\mu_{mix}(p, f)$ were used to determine the model parameters N and p_c . These parameters found from the data for different frequencies have close values. The



Fig. 5. Magnetic permeability (real (solid) and imaginary (dashed) parts) of spherical particles μ_{incl} (black lines) and their metal μ_{metal} (dark-grey lines, the skin-effect contribution was excluded from the particle permeability μ_{incl} in the case of μ_{metal}) versus frequency and approximation of the alsifer magnetic permeability μ_{metal} with the Lorentz line (light-grey lines).

obtained deviations of 1-5% are produced by the errors in measuring the material parameters of composites. The independence of the calculated parameters from the contrast between component properties confirms the applicability of the selected mixing model.

Black lines in Fig. 5 represent the frequency dependence of the magnetic permeability $\mu_{incl}(f)$ of spherical alsifer particles. Each black point in this dependence was obtained by analyzing the corresponding array of values for $\varepsilon_{mix}(p)$ and $\mu_{mix}(p, f)$ given in Figs. 2 (curve of spheres) and 3, respectively.

Similarly, a black line in Fig. 6 corresponds to the frequency dependence obtained for the magnetic permeability $\mu_{incl}(f)$ of lamellar alsifer particles by processing the corresponding array of values for $\varepsilon_{mix}(p)$ and $\mu_{mix}(p, f)$ shown in Fig. 2 (curve of disks) and 4, respectively.

From the comparison of the dependences $\mu_{incl}(f)$ for spherical and lamellar particles it can be seen that the magnetic permeability of lamellar particles is higher than for spherical ones at frequencies below 2 GHz probably because of the exclusion of the inner volume of a spherical particle due to the skin-effect (according to microscopy data, the thickness of lamellae is nearly 1 μ m, and the average diameter of spheres is 4.2 μ m). At a frequency of 100 MHz (the lower boundary of the experimental frequency band), the magnetic permeability of both inclusions does not attain saturation and is far from its static value.



Fig. 6. Magnetic permeability (real (solid) and imaginary (dashed) parts) of lamellar particles μ_{incl} (black lines) and their metal μ_{metal} (dark-grey lines, the skin-effect contribution was excluded from the particle permeability μ_{incl} in the case of μ_{metal}) versus frequency and approximation of the alsifer magnetic permeability μ_{metal} with the Lorentz line (light-grey lines).

At an alsifer conductivity $\sigma_{incl} \approx 1.6 \times 10^7 \Omega^{-1} m^{-1}$ and a thickness (minimum size) *d* of inclusions, it is possible to exclude the skin-effect contribution from the above estimated magnetic permeability of inclusions $\mu_{incl}(f)$ at each frequency point and calculate the frequency dependence of the magnetic permeability of the inclusion composing alloy $\mu_{metal}(f)$. The correction for the skin-effect contribution is applied for spheres and an infinite plane [17], and the magnetic permeability of an inclusion is related with the magnetic permeability of the inclusion material $\mu_{incl} =$ $\mu_{metal} f(\theta)$ via the multiplier $f(\theta)$, which is a function of the optical thickness θ of an inclusion, i.e.,

$$\theta = 2\pi f d \sqrt{\varepsilon \mu}.$$
 (6)

This function is $f(\theta) = \frac{\tan(\theta)}{\theta}$ for the plane and

 $f(\theta) = 2 \frac{\tan(\theta) - \theta}{(\theta^2 - 1)\tan(\theta) + \theta}$ for a spherical inclusion.

The frequency dependence of the alsifer magnetic permeability $\mu_{metal}(f)$ for spheres with the skin-effect contribution, which was numerically excluded from $\mu_{incl}(f)$, is shown in Fig. 5 as dark-grey lines. Similarly, the frequency dependence of $\mu_{metal}(f)$ for lamellar particles is plotted in Fig. 6. Let us note that the error in the lamella thickness has an appreciable effect on the calculated magnetic losses at low frequencies: the overestimation of this thickness leads to the appearance of negative values for the imaginary part of μ_{metal} .

Thus, the thickness obtained for lamellar particles from the analysis of electron microphotos is close to 1 µm, and the average diameter of a lamella is nearly 50 µm. At a depolarization coefficient N = 0.00854found as described above, the ratio of the thickness of an equivalent ellipsoid (disk) to its diameter is $d/D \approx$ 0.0011. For the imaginary part of the magnetic permeability µ^{''}_{metal} (dark-grey dashed line in Fig. 6) to be close to zero at frequencies below 500 MHz, the thickness of a lamella must be nearly $d \approx 0.65$ µm. Whence, the equivalent diameter of a lamella at $d/D \approx 0.0011$ is

size of a sieve separator (mesh side, 63 μ m). Let us note that the equivalent diameter of a lamella exceed 100 μ m for the processing of experimental data (Figs. 2–4) by the Maxwell–Garnett formula, being much greater than a sieve mesh, and the deviations of the calculated mixture permeability $\mu_{mix}(f)$ from its value measured at high and low filler

contents exceed 15%.

 $D \approx 58 \,\mu\text{m}$, which is in good agreement with the mesh

A decrease in the real part of the magnetic permeability μ'_{metal} of the inclusion material with decreasing frequency at frequencies below ~300 MHz in Figs. 5 and 6 seems to be due to the limitations of the method used to estimate the magnetic permeability of metal. On the one hand, the measurement error for $\mu_{mix}(f)$ grows with decreasing frequency due to a small optical thickness of the studied specimens. On the other hand, the magnetic permeability of inclusions $\mu_{incl}(f)$ grows due to frequency dispersion and a decrease in the skin-effect contribution, and the error determined by inequality (5) appears. The accuracy of measuring the magnetic permeability of metal at frequencies below 500 MHz may be improved with the use of more precise methods for the measurement of $\mu_{mix}(f)$ (at megahertz frequencies) and specimens with a higher content of inclusions. However, a further increase in the filler content proves to be problematic for the used specimen manufacturing technology, as specimens become brittle and break down during their removal from a press mold.

To estimate the magnetic permeability of alsifer at frequencies below 500 MHz, the obtained frequency dependences of the magnetic permeability of alsifer were approximated with the resonance (Lorentz) curve

$$\mu_{\text{metal}} = 1 + \frac{\mu_{\text{static}} - 1}{1 - \left(\frac{f}{F}\right)^2 + i\frac{\Gamma f}{F}}.$$
(7)

In this equation, μ_{static} is the static magnetic permeability of metal, *F* is the ferromagnetic resonance frequency, Γ is the attenuation coefficient, and *f* is the frequency, at which μ_{metal} is calculated. In Figs. 5 and 6, the calculated frequency dependences of μ_{metal} are dark-grey, and their approximations are presented as light-grey lines.

From these figures it can be seen that the attenuation coefficient for the approximating Lorentz functions is rather high. The quantitative comparison of the resonance frequencies F is, however, problematic here, so it is necessary to consider the absorption maximum frequency. The static magnetic permeabilities obtained for spherical and lamellar alsifer particles are close to 300. The alsifer absorption maximum in lamellar particles corresponds to a frequency of nearly 3 GHz, being appreciably lower for spherical particles, i.e., nearly 1 GHz.

The shift of the alsifer absorption maximum for lamellar particles towards higher frequencies in comparison with the absorption maximum of the same alloy in spherical particles may be due to the dependence of ferromagnetic resonance conditions on the shape of a particle. Thus, when one dimension of a uniformly magnetized ferromagnetic is much smaller than the others (a film or a plane), the ferromagnetic resonance frequency will be governed not only by the field of anisotropy, but also by the difference between the demagnetization coefficients. In this case, the static permeability proves to be close to its value observed in a body with close dimensions in all directions (in a sphere). In other words, the parameters of the frequency dependence of the magnetic permeability of a ferromagnetic are determined not only by its composition, but also by the shape of a body, in which ferromagnetic resonance is observed.

5. CONCLUSIONS

It has been shown that the used Odelevsky model is in good agreement with the measured frequency and concentration dependences of the material parameters of composites. The applicability of Eq. (2) is confirmed by that the parameters N and p_c remain constant with a change in the contrast between the permeabilities of mixture components within broad ranges, and the equivalent lamella diameter D calculated from the demagnetization coefficient N and the skin-layer thickness (Eq. (6)) corresponds to the mesh size of a sieve separator.

The processing of experimental data by the Maxwell–Garnett formula leads to that the deviation between the calculated and measured permeabilities of a composite lies outside experimental error limits. Moreover, the lamella diameter D (calculated at $p_c =$ 1) is almost two times greater than the sieve mesh size (100 and 63 µm, respectively).

An obvious shortcoming of matrix models is the presentation of a composite as a regular structure similar to a crystal lattice. It is assumed that there are no local concentration fluctuations, and the interaction characterized by the parameters N and p_c between all

inclusions is equivalent. In real practice, the distribution of inclusions by their effective shape takes place and is described by the Bergman-Milton characteristic geometric function [11, 18], and the summand $(1 - p/p_c)N$ in the denominator of Eqs. (3) and (4) is only a refined approximation in comparison with (1 - p)N in the Maxwell-Garnett formula.

The accepted approximations lead to small deviations (3–5%) of the mixture permeability $\mu_{mix}(f)$ calculated from the found parameters N, p_c , and $\mu_{incl}(f)$ from its measured value at minimum and maximum filler contents ($p \rightarrow 0$ and $p \rightarrow p_c$, respectively). A similarly small deviation of the permeability calculated by the Odelevsky model from its measured value was obtained when studying the composites with iron powders [13].

Hence, the Odelevsky formula modified with allowance for the percolation threshold more precisely describes the properties of matrix mixtures in comparison with the Maxwell–Garnett formula. The found parameters of the mixing formula are confirmed by the results of independent measurements.

The broader is the describable range of filler contents and contrasts between the permeabilities of mixture components for a selected mixing formula, the lower is the error of restoring the permeability of a particle from the permeability of mixtures.

In comparison with the processing of measurement results for the magnetic permeability $\mu_{mix}(f, p)$ alone, the simultaneous processing of the frequency and concentration dependences of both material parameters $\varepsilon_{mix}(f, p)$ and $\mu_{mix}(f, p)$ of a mixture provides the more precise estimation of the mixing formula parameters and, correspondingly, the more accurate restoration of the frequency dependence of the alsifer permeability $\mu_{metal}(f)$ in the region of high μ_{metal} .

An appreciable increase in the error of estimating μ_{metal} at frequencies below ~300 MHz is due to both an increase in the error of measuring the material parameters of a composite by the transmission/reflection method and a nonlinear relation between the particle and mixture permeabilities (Eq. (2)). The precision of estimating μ_{metal} can be improved with the use of a more precise method of measuring the material parameters and specimens with a higher filler content.

In principle, the precision of estimating high values of μ_{metal} can be improved by decreasing the effective magnetic permeability of inclusions μ_{incl} and filling mixtures with particles of greater thickness *d*. In this case, the parameter μ_{incl} will be lower due to the skineffect and, correspondingly, the error of its measurement for the same shape and concentration of inclusions will be smaller. However, the accuracy of measuring the effective permeability of inclusions is increased due to the screening of a major part of the inner particle volume, and the skin-effect contribution can precisely be taken into account only for a sphere or an infinite plane. It seems that the growth of particles in size will lead only to that the error of estimating μ_{incl} will be replaced by the skin-effect correction error.

The measured frequency dependences of the dielectric and magnetic permeabilities of mixtures indicate that the studied alsifer particles may serve as a filler for electromagnetic radiation absorbents and materials providing electromagnetic compatibility at frequencies below 1 GHz, as it superior to carbonyl iron in magnetic losses. At the same time, the restored magnetic spectra of the Al_{0.054}Si_{0.096}Fe_{0.85} alloy indicate that the obtained values of the magnetic permeability of a composite $\mu_{mix}(f)$ are limited first of all by the skin-effect and can be appreciably elevated by increasing the dispersion of particles.

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REFERENCES

- 1. W. Arkadiew, Ann. Phys. 58, 12 (1919).
- 2. J. H. E. Griffits, Nature 158, 670 (1946).
- G. F. Hodsman, G. Eichholz, and R. Millership, Proc. Phys. Soc. Sec. B 62, 377 (1949).
- P. Lederer and C. Brewitt-Taylor, IEEE Proc. Sci. Meas. Technol. 147, 209 (2000).
- J. C. Maxwell Garnett, Phil. Trans. R. Soc. London A 203, 385 (1904).
- M. Han, D. Liang, K. N. Rozanov, and L. Deng, IEEE Trans. Magn. 49, 982 (2013).
- 7. O. Wiener, Abh. Math. Phys. Klasse Konigl. **32**, 509 (1912).
- L. V. Panina, A. S. Antonov, A. K. Sarychev, V. P. Paramonov, E. V. Timasheva, and A. N. Lagarkov, J. Appl. Phys. 76, 6365 (1994).
- 9. A. N. Lagarkov, V. N. Semenenko, V. A. Chistyaev, and I. T. Iakubov, J. Magn. Magn. Mater. **324**, 3402 (2012).
- 10. D. A. Bruggeman, Ann. Phys. (Leipzig) 24, 636 (1935).
- K. N. Rozanov, M. Y. Koledintseva, and J. Drewniak, J. Magn. Magn. Mater. 324, 1063 (2012).
- A. V. Osipov, K. N. Rozanov, N. A. Simonov, and S. N. Starostenko, J. Phys.: Condens. Matter 14, 9507 (2002).
- K. N. Rozanov, A. V. Osipov, D. A. Petrov, S. N. Starostenko, and E. P. Yelsukov, J. Magn. Magn. Mater. 321, 738 (2009).
- V. I. Odelevskii, Cand. Sci. Dissertation (Sci. Res. Inst. No. 34, Leningrad, 1946).
- 15. A. M. Nicolson and G. F. Ross, IEEE Trans. Instrum. Meas. **19**, 377 (1970).
- 16. J. A. Osborn, Phys. Rev. 67, 351 (1945).
- 17. F. Frank and R. Mizes, *Differential and Integral Equations of Mathematical Physics* (ONTI, Leningrad, 1937) [in Russian].
- K. N. Rozanov, Z. W. Li, L. F. Chen, and M. Y. Koledintseva, J. Appl. Phys. 97, 013905 (2005).

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