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On Properties of Magneto-dielectric Composites in the Effective Medium Approximation

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The development of ultra-broadband composite absorbers of electromagnetic waves depends largely on optimal combination of the medium characteristics that determine their absorption capacity and conditions for impedance matching and destructive interference. It is possible to achieve a higher absorption level in a wider frequency band by combining a variety of mechanisms enhancing the loss of electromagnetic field energy, for example, by combining specific constituents in a composite matrix. The analysis of various mixing models for constituent parameters is carried out in the effective medium approximation for ferrite-dielectric composites. It appears that the standard mixing rules do not explain the increase in the effective permittivity of ferrite composites in comparison with that of bulk ferrites. The proposed mechanism of such increase is based on the conductive properties of the ferrite granules and the equivalent capacitance effect. The developed model of permittivity calculation is based on the equivalent capacitor circuits and gives a satisfactory agreement with the experimental data.

Keywords: Radar absorbing material, Ultra-wideband absorber, Electromagnetic radiation, Effective medium approximation, Reflection coefficient, Effective permeability and permittivity.

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1. INTRODUCTION

Rapid developments in the electronics and telecommunication require control over the electromagnetic radiation (EMR) to ensure electromagnetic compatibility and ecological safety in a wide frequency range. This problem remains a challenge and it is vital to investigate novel designs of electromagnetic materials for efficient shielding and absorption of EMR [1-3]. Many applications spreading far beyond military usage are related with a need to make certain objects invisible (or less visible) to radar, which is known as stealth technology [4]. Composite materials with magnetic phase such as polymer with ferrites can be developed to meet these needs through tailoring the spectra of the effective permittivity and permeability.

Electromagnetic energy can be lost in two main ways: by being absorbed into a coating material (radio absorbing coating, RAC) that converts it into another form of energy such as heat; and by destructive interference of the waves reflected from different layers of coating. The absorption efficiency also depends on the impedance matching [5]. Wave scattering due to geometric heterogeneity of the complete structure is also possible but typically used for large-scale applications. Single-layer interference RACs are usually operated in a narrow band. Today's requirement is to expand the frequency band of EMR protection, which is possible with multilayer structures having different electromagnetic properties. In this context, radio-absorbing materials (RAM) should provide high absorption over a wide frequency range. The ability of natural materials for broadband operation is very limited and the quest for novel designs of broadband RAMs becomes imperative [6-9].

Ferrite materials can be designed to have a relatively wide absorption spectrum considering all the losses: magnetic, dielectric and resistive [10]. Moreover, unlike many other RAM, they can operate at relatively low frequencies (lower than 1 GHz and down to 100 kHz). In particular, Ni-Zn ferrites could be efficient at such frequencies [10, 11]. Unfortunately, they are quite expensive, and nickel compounds used in their manufacture is highly toxic [11 - 13]. A good substitute discussed here is Mn-Zn ferrite of a spinel type. This is a soft magnetic material having a high permeability at frequencies below 100 MHz. Beyond this frequency the permeability substantially decreases due to the suppression of domain wall dynamics. Low values of the magnetic anisotropy of some kinds of this ferrite limit the natural ferromagnetic resonance frequency by 100 MHz [13, 14]. But it is well known that the radio-absorbance depends on the composition and structure of materials [15]. Therefore, the combined systems like the composite materials (CM) with various fillers should be very attractive objects as RAM. Thus, one of the types of such media is a combination of ferrite and insulator materials with different values of permittivity, permeability and conductivity [16]. For example, the essential absorption in various CM with ferrite fillers can be achieved by increasing the dielectric loss in the frequency range above 100 MHz [13, 15]. Conducting particles can also be added to increase resistive losses but it may be difficult to control the increase in the real part of permittivity to realize efficient impedance matching.

R. Feynman in his famous lectures on physics considered that ferrites were one of the most difficult field for theoretical description, but the most interesting for investigations and practical applications [17]. This is even more relevant to CM with various ferrite fillers.

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The analysis of electromagnetic response from such heterogeneous systems (HS) as CMs is a challenging task [18, 19]. Effective parameters depend on many factors including intrinsic properties of the fillers and the host matrix, their shape and topology. The latter determines the filler layout: how inclusions are distributed in the matrix. The problem is simplified in the effective medium approximation when the interaction between the inclusions is replaced by that corresponding to the case when the inclusion is embedded in the medium with the effective parameters: effective permittivity and permeability. There are a number of so called mixing formulas (see, for example, review in [20]). Typically, such parameters as inclusion concentration and intrinsic properties are included explicitly in the mixing rules. Various approaches are different by the method how the topological characteristics are accounted for. This work develops the model allowing the evaluation of the effective parameters of ferrite composites accounting for conductive properties of ferrite particles. This model explains an increase in the effective permittivity of ferrite composites in comparison with that for bulk ferrites, observed experimentally.

Unfortunately, a rigorous analysis of the effective electromagnetic response is usually difficult due to a lack of reliable information about both the structure and properties of constituent components and about the topology of CM.

2. TOPOLOGICAL STRUCTURE OF FERRITE COMPOSITES USED AS RAM

The electromagnetic properties of composites are analyzed in terms of the effective theory approximation. This allows avoiding the direct numerical calculations and providing the conceptual simplicity. However, the method ignores fluctuations assuming that the local electric and magnetic fields are the same in each component. On the other hand, it is well known that the efficiency of RAM depends not only on the properties of fillers but also on the topological structure: on the way they are distributed in the matrix. The effective medium theory can be modified to account for the internal structure. In this section the important structural parameters are introduced.

We consider a model of a composite material consisting of fillers embedded in a dielectric matrix. The fillers of a certain shape and a characteristic size D can be distributed randomly or periodically forming a crystal-type structure. The effective parameters are determined in terms of a volume concentration C_v. On the other hand, the technological parameter is the mass concentration C_m. The mentioned concentrations are related as

$$C_v = \rho_1 C_m / [\rho_2 C_m + \rho_1 (1 - C_m)], \qquad (1)$$

where ρ_1 and ρ_2 are the densities of each component of the CM. The ratio $k = \rho_1 / \rho_2$ shows to which extent the mass and volume concentration differ.

The densities of practically important ferrites are in the ranges of 3.6-5.8 g/cm^3 for hexagonal and spinel ferrites, 5.2-7.2 g/cm^3 for garnets, and about 7-9 g/cm^3 for ferromagnetic metals. The density of polymers for dielectric matrix is about 1 g/cm^3. Dashed area in Fig. 1 showing the relationship between C_v and C_m corresponds to practically important ferrite materials. Among them, Mn-Zn ferrite of types 700NM and 2000NM having the density of 4.3-4.7 g/cm^3 which was used here for the investigations of radio-absorbing properties [21].

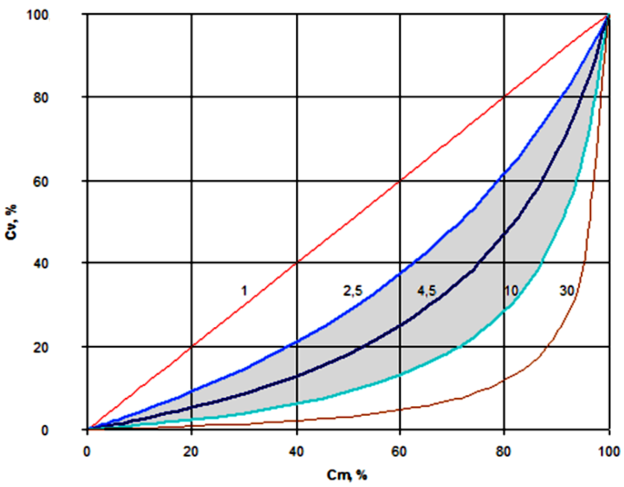

Fig. 1 – Diagram $C_v = f(C_m)$ for different values of the parameter k

It is proposed to approximate an ordered composite material having a conducting phase as a capacitor of width h filled with a dielectric medium with the effective permittivity ε. In another approach, a system of capacitors consisting of adjacent conducting particles and dielectric layers with matrix permittivity ε_2 and inter-particle thickness $d = 2\delta$ is considered. The proposed structures are depicted in Fig. 2 for particles of cubic and spherical shapes.

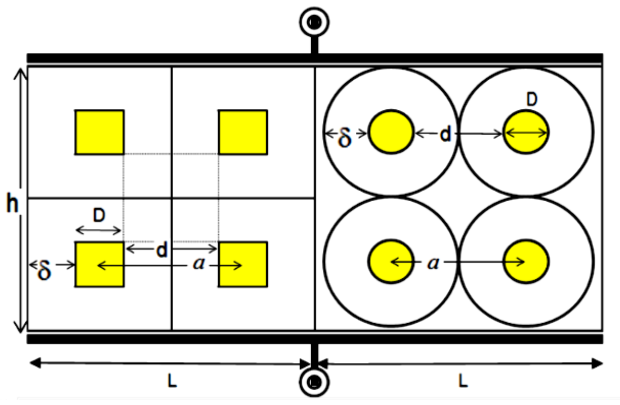

Fig. 2 – Scheme of packing particles and characteristic sizes in the structure of CM

The averaged distance a between the spherical particles and minimal spacing d between them are defined as

$$a = D(K_p / C_v)^{1/3},\ d = D[(K_p / C_v)^{1/3} - 1] \qquad (2)$$

where K_p is the maximal packing factor. For example, for spherical particles in a simple cubic lattice this parameter is 0.5236. These parameters are known to affect the electromagnetic properties of composites and the proposed effective medium approach more accurately accounts for the composite microstructure. For cubical particles in a simple cubic lattice the minimum inter-particle spacing is defined as

$$d = D\,(1 - C_v^{1/3}) / C_v^{1/3}, \qquad (3)$$

Assuming a flat particle surface, the averaged distance between them is typically defined (see, for example, [22]) as

$$d_{av} = D\,(1 - C_v) / 3C_v. \qquad (4)$$

This differs from (2) and gives an upper limit due to including a portion of the dielectric matrix outside the inter-particle space.

A more rigorous approach to calculate inter-particle spacing accounting for a spherical surface yields:

$$d_{av} = D\,\{[6K_p / (\pi C_v)]^{1/3} - 1\}, \qquad (5)$$

Equation (5) is different from (3) and takes into account more accurately the particle layout. However, using (5) for spherical particles gives the same result as (3) for cubic particles in a cubic lattice.

Figure 3 shows the inter-particles distance vs. mass concentration for different types of ordered composites. The average particle size is chosen to be $D = 400$ µm, and $k = 4.5$.

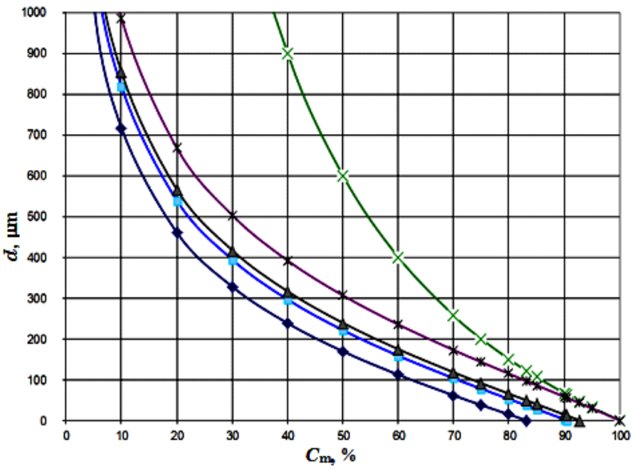

Fig. 3 – Inter-particles distance d vs. mass concentration C_m for different types of ordered composites. ($D = 400$ µm, $k = 4{,}5$). Equation (2) was used for different lattices of spherical particles: simple cubic, volume-centered cubic, close-packing (face-centered cubic or hexagonal close-packed); results of (4): ; results of (5) for spherical particles and (3) for cubic particles:

3. EFFECTIVE MEDIUM APPROXIMATION FOR CALCULATING THE PERMITTIVITY

There are a number of approaches to calculate the effective dielectric and magnetic responses of CM. Expression for values of the permittivity and permeability for CM are known as the mixing formulas by Lorentz, Rayleigh, Kondorsky-Odelevsky, Lichtenekker, Ollendorff, Bruggeman, Maxwell, Maxwell-Garnett, Wagner and others [see in 15, 18, 20, 22, 23].

For example, Maxwell-Garnett formula for the effective permittivity is obtained by considering the ratio of the averaged displacement and electric field. A rigorous approach in this approximation is related to randomly spaced spherical particles but can be generalized to particles of other shapes introducing the effective depolarization factor. The Bruggeman theory is better suited for percolating composites but essentially utilizes Maxwell-Garnett approach where the spherical particles are embedded into a matrix with the effective permittivity. In principle, this approach can be used for any concentrations. Other generalizations include coated particles, separate percolation portions (Bergman approach) and brick-wall models based on capacitor equivalent circuits. The latter is approximate not allowing rigorous calculations but it is frequently used for ceramics to consider different properties of grain bulk and boundaries. This approach is advanced here for composite microstructure in attempt to account for various structural properties and to explain the experimental results on the permittivity for ferrite composites the value of which can be considerably higher than that for bulk samples.

3.1 Experimental Results

Various magnetodielectric materials (bulk ferrites, CM with ferrite granules ~ 10 … 650 µm size disperged in dielectric matrix and nanocomposites with a much smaller particles) were previously studied experimentally [21, 24-27]. Therefore it is desirable that used models should describe above listed systems in all frequency range of interest (100 kHz … 10 GHz or even up to 30 GHz). It appears that the explanation of obtained results using various mixing rules is difficult in the frequency range of interest (100 kHz … 10 GHz).

The effective permittivity of CM can be mainly contributed by the conducting properties of ferrite inclusions [28]. It is possible to achieve enhancement of absorption capacity in ferrites with the conducting grains due to the formation of insulating inter-grain boundaries with increased permittivity [11, 29]. Therefore, the structure composed of semiconductive Mn-Zn ferrite particles isolated from each other by a dielectric medium, is characterized by high values of electrical capacity and leads to increased values of dielectric permittivity.

This was confirmed by the experimental studies of composites consisting of granules of Mn-Zn ferrites in paraffin wax matrix as shown in Fig. 4 where the spectra of the real part of permittivity are shown for different concentrations. For mass concentration of 70-80 % (corresponds to 34-47 % volume concentration) a significant increase in the permittivity is seen (more than three times in comparison for that typical of bulk ferrite or matrix ($\varepsilon_1 \sim 5.5$ for ferrite and $\varepsilon_2 \sim 2.2$ for paraffin).

Such increase is not possible to explain within standard mixing formulas. Even in the case of relatively low concentrations of about 13 % the composite permittivity is higher than calculated value by the standard formulas, but lower than ε_1. Further we develop a model based on conducting properties of ferrite granules [30], attempting to explain the observed behavior.

3.2 Composites with Conducting Particles

Let us consider a composite with conducting inclusions (for example, weakly conducting Mn-Zn ferrite) of

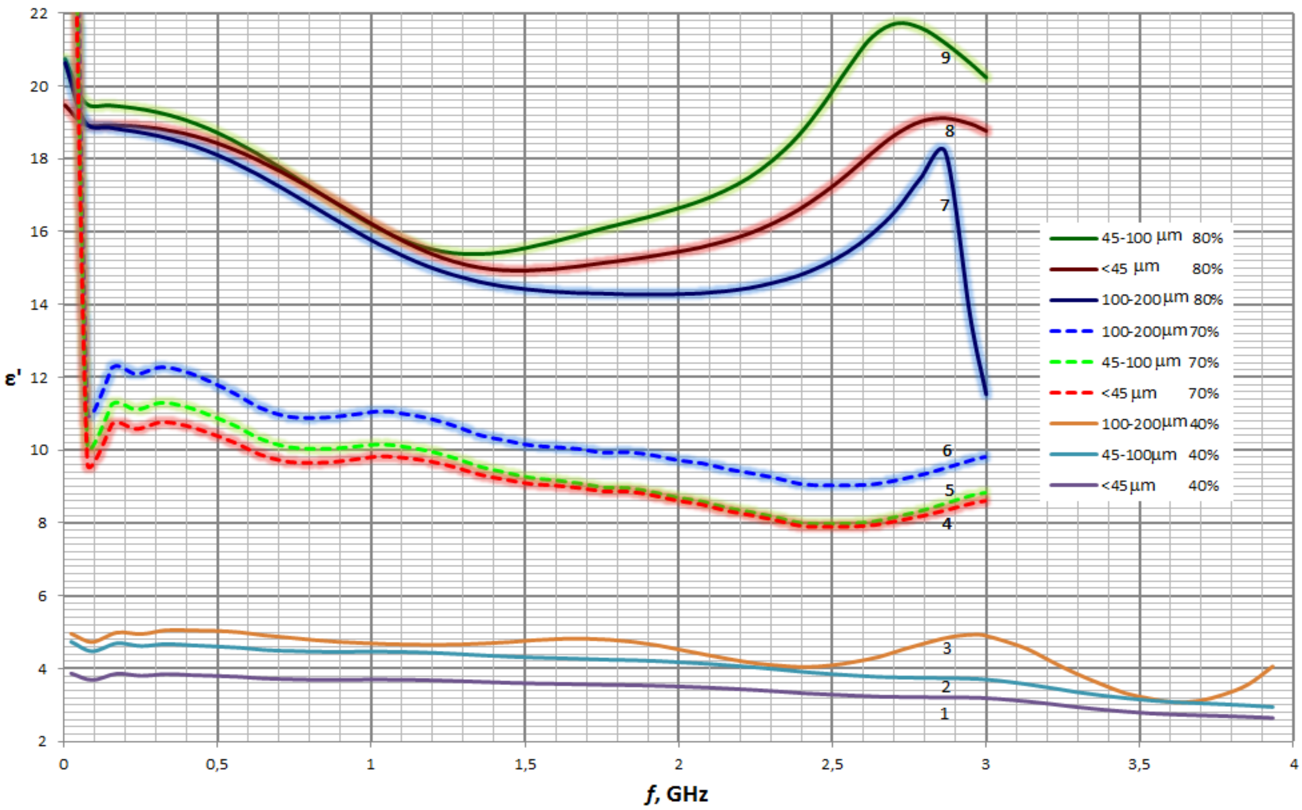

Fig. 4 – Permittivity spectra of ferrite composites for various concentrations (of the mass) and ferrite granular sizes

cubic shape in a cubic lattice as shown by the left diagram in Fig. 2. The composite sample is approximated by a capacitor of width h, the plate area L^2 and filled with a medium with the effective permittivity ε. The sample can also be subdivided into a number of connected capacitors having a width d and plate area D^2.

In the limit of $L >> h >> D >> d$ (dense composite) the formula for a system of plane capacitors with parallel-serial connections can be used. The total amount of capacitors is assumed to equal the number of particles $N = C_v L^2 h / D^3$. The capacitance of such system is

$$C_{CM} = C_v L^2 (d + D)\varepsilon_2 / \{4\pi dD[h / (d + D) - 1]\}. \quad (6)$$

Substituting (3) to (6) gives:

$$C_{CM} = L^2 C_v \varepsilon_2 / [4\pi(1 - C_v^{1/3})(hC_v^{1/3} - D)]. \quad (7)$$

The considered system is equivalent to the capacitor with the plate area of L^2 filled with a dielectric medium with additional permittivity $\Delta\varepsilon$ and having a width $h - d$. Then,

$$C_{CM} = \Delta\varepsilon L^2 / [4\pi(h - d)]. \quad (8)$$

Comparing (7) and (8) yields:

$$\Delta\varepsilon = \varepsilon_2 (D / d) C_v^{1/3} = \varepsilon_2 C_v^{2/3} / (1 - C_v^{1/3}), \quad (9)$$

where the total value of the permittivity is given by expression $\varepsilon = \varepsilon_2 + \Delta\varepsilon$.

The considered model is certainly a strong approximation and the concentration in (9) should be reasonably smaller than 1. However, this demonstrates the possibility to increase few times the effective permittivity of composites in comparison with the permittivity of matrix.

For the case of a diluted composite with a small volume concentration of ferrite fillers, the system of isolated conducting spheres is considered ($a >> R$, a is the distance between the centers and R is the radius) as shown in Fig. 5. The interaction between the two oppositely charged spheres almost does not change the charge distribution on the surface, so it is considered that the field is generated by the uniformly charged spheres, so the effective charges $+q$ and $-q$ could be placed in their centres at the distance a from each other. The deviation from a uniform charge distribution would result in the shift of the effective charge location from the center by $R' < R$. When $C_v \to 0$, $R' \to 0$ and $d \to a$.

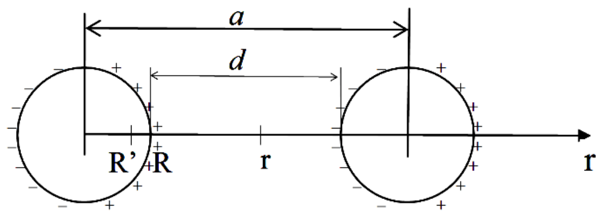

Fig. 5 – Geometry and charge distribution of two conducting spheres considered for the calculation of the effective permittivity

Taking the centre of the left sphere as origin, the electric field as a function of distance r is written as:

$$E(r) = (q / \varepsilon_2)\,[1 / (r - R')^2 + 1 / (r + R' - a)^2]. \quad (10)$$

In this approximation, the potential difference between the spheres is

$U = \int E(r)dr = (q / \varepsilon_\partial)\int[1/(r - R')^2 + 1/(r + R' - a)^2]dr$, (11)

If the effective charge shift is neglected, the integration limits are $r = R$ and $a - R$. The result of the integration is:

$$U = (2q / \varepsilon_2)\,(2R - a) / (R[R - a)], \qquad (12)$$

Equation (12) is then used to evaluate the capacitance of the system of two spheres ($C = q / U$), which gives

$$C = \varepsilon_2 D\,(d + D / 2) / (4d). \qquad (13)$$

Following the method of equivalent capacitance circuit described above the upper estimate of additional permittivity in the limit for low concentrations of filler is evaluated

$$\Delta\varepsilon = \varepsilon_2 \pi C_v^{1/3}(1 - C_v^{1/3} / 2) / (1 - C_v^{1/3}). \qquad (14)$$

Equation (14) also demonstrates the possibility to increase the effective permittivity of composites with conducting fillers. Thus for composite with ferrite fillers of mass concentration $C_m = 40$ % ($C_v = 13$ %) the increase in the permittivity value is 3.5 times. Further increase in $\Delta\varepsilon$ is possible when considering the difference between spherical and cubic particle packing. For the same particles size and concentration this increase is up to $(6 / \pi)^{1/3} \approx 1.24$ times.

3.3 Composites with Non-conducting Particles

The resulting value of permittivity for the existing models of HS in the effective medium approximation should be between the upper and lower estimates:

$$\varepsilon_{\min} = \left(\frac{\varepsilon_1}{p} + \frac{\varepsilon_2}{1 - p}\right)^{-1} \text{ and } \varepsilon_{\max} = \varepsilon_1 p + \varepsilon_2(1 - p), \quad (15)$$

where $p \equiv p_1$ is the concentration of the first phase (corresponds to the volume fraction of filler particles C_v in sections 2 and 3.2), $p_2 \equiv 1 - p$ is the concentration of the second phase.

The model of equivalent capacitive circuits can be generalised to take account of the permittivity of the filler which can be complex due to conducting properties. Let us consider a two-phase CM consisting of filler particles of the size D and permittivity ε_1 embedded in the matrix of permittivity ε_2. For cubic particles in a cubic lattice as shown in Fig. 2 an elementary cell has a size of $D + d$, and the volume concentration of the fillers is

$$p = [D / (D + d)]^3. \qquad (16)$$

The elementary cell is represented by a system of capacitors: three capacitors connected in series and one capacitor with parallel connection as shown in Fig. 6. The capacitors connected in series have a plate area of D^2, a width of D and $d / 2$ and permittivity of ε_1 and ε_2, respectively. The plate area of that connected in parallel is $(D + d)^2 - D^2$, a width is $D + d$ and the permittivity is ε_2.

The total capacitance of this system equals to that of the equivalent capacitor filled with the medium having the effective permittivity ε. Then, the balance equation is

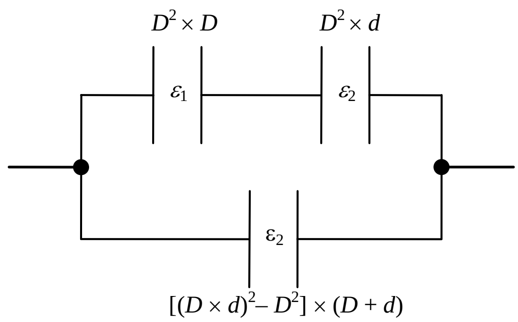

Fig. 6 – Representation of two-phase medium in Fig. 2 as an equivalent circuit of three capacitors

$$C_{CM} = \varepsilon(D + d)^2 / (D + d) = D^2 / [d / \varepsilon_2 + D / \varepsilon_1] + \\ + \varepsilon_2(D + d)\,[1 - D^2/ (D + d)^2], \qquad (17)$$

Equation (17) written in terms of the filler concentration p has a form:

$$\varepsilon = \varepsilon_2\left[1 + \frac{p(\varepsilon_1 - \varepsilon_2)}{\varepsilon_1\left(1 - p^{1/2}\right) + \varepsilon_2 p^{1/2}}\right] = \\ = \varepsilon_2\left[1 + \frac{p(\varepsilon_1 - \varepsilon_2)}{\varepsilon_1(\varepsilon_1 - \varepsilon_2)p^{1/2}}\right]. \qquad (18)$$

In the case of a highly conducting filler ($\varepsilon_1 \to \infty$) equation (18) reduces to the form similar to (9):

$$\varepsilon = \varepsilon_2[1 + p / (1 - p^{1/3})]. \qquad (19)$$

In [29] a problem of finding the effective permittivity of polycrystalline ferrite was considered in similar approximations: cubic conducting ferrite grains in a simple cubic lattice of a dielectric matrix. The matrix is formed by inter grain boundaries. The additional permittivity obtained in this work was

$$\Delta\varepsilon = \varepsilon_2(D / d), \qquad (20)$$

from which we can conclude:

$$\Delta\varepsilon = \varepsilon_2 p^{1/3} / (1 - p^{1/3}). \qquad (21)$$

It differs from equation (9) obtained here by a factor of $p^{1/3}$ and gives an enhanced value of the effective permittivity.

4. DISCUSSION

The values of the effective permittivity of CM at a low concentration of conductive inclusions can be found by using expression (14). In the case of high concentrations, formulas (9), (19), (21) should be used. Equations (9) and (19) lead to similar results for $p > 0.75$.

For CM described in section 3.1, the values of the effective permittivity ε obtained from (18) for limiting concentrations are: 3.54 at $p = 0.52$ (cubic lattice), 4.10 at $p = 0.68$ (volume-centered cubic lattice), and 4.34 at $p = 0.74$ (close-packing lattice). In the case of conducting particles, equation (19) yields much higher values: 8.0, 14.6 and 19.25, respectively, which are close to the experimental data of Fig. 4. Therefore, we can assume that the capacitor effect results in the additional contribution $\Delta\varepsilon$ to the permittivity of CM. The value of $\Delta\varepsilon$

should be added to the matrix permittivity or to the averaged permittivity calculated by mixing rules, for example, by equation (18)

$$\varepsilon = \varepsilon_2 + \Delta\varepsilon. \quad (22)$$

Equation (14) obtained for low concentrations gives $\Delta\varepsilon = 1.7\text{-}5.3$ for the $p = 0.01\text{-}0.13$. Then $\varepsilon = 3.9\text{-}7.5$. These values which also correspond to the experimental results are considerably higher than those obtained from any mixing formula for dielectric composites, but do not exceed the value of the upper estimate in (15) and the constituent permittivity with the highest value (ferrite).

Some discrepancies between predicted and experimentally found data in our case may also be due to certain differences of material parameters of porous granules and high-density ferrite ceramics. Comparison of dielectric layer thickness obtained by calculating the parameters of the CM structure from geometrical considerations with the one calculated from the measured permittivities of CM and ferrite assesses ~ 20 % granule porosity value.

The developed models should be considered to be qualitative. However, the obtained results describe surprisingly well the experimental data.

CONCLUSIONS

The paper discusses various models of the electromagnetic response from composite materials with ferrite fillers. The mechanism of increasing the effective permittivity in comparison with that of bulk ferrites is proposed which is based on the capacitance effect. The developed model of the equivalent capacitor circuits although been qualitative gives a satisfactory agreement with the experimental data. Therefore, the proposed approach of calculating the effective permittivity can be used for designing broadband radio absorbing structures.

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