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Method of Measuring Permittivity of Composite Materials with Hexagonal Ferrite Inclusions

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Abstract— A new simple method for measuring complex permittivity of substantially lossy composite materials is presented. In this method, a sample of the material under study should completely fill in the cross-section of the single-mode transmission line (waveguide), and the length of the sample must be an integer of a half-wavelength in the waveguide filled with this material. The oscillator frequency is swept linearly, the minima of the reflection coefficient are measured, and then analytical formulas are used to calculate real and imaginary parts of permittivity. The method was tested on magneto-dielectric samples containing hexagonal ferrite powders, as well on such dielectric materials, as PMMA, schungite composites, and alabaster. This method can be a useful technique for measuring dielectric properties of absorbing materials designed, for example, for electromagnetic shielding purposes.

Keywords- composite, complex permittivity, waveguide, reflection coefficient, hexagonal ferrite, schungite, PMMA, alabaster

I. INTRODUCTION

In many practical applications, it is required to reduce reflections of electromagnetic waves of the RF and microwave frequency ranges from metal surfaces. This problem may be solved by placing a layer (or layers) of wideband composite radio absorbing materials (CRAM), in particular, magneto-dielectric absorbing materials. When studying frequency dependences of a reflection coefficient from an object coated with CRAM, it is important to know material properties at the frequencies of the intense absorption of electromagnetic waves, in particular, complex permittivity \( \varepsilon \) and permeability \( \mu \) over the frequency range of operation.

This work is focused on measuring complex permittivity of ferrite-based CRAMs. Application of ferrites in CRAMs is very attractive. Ferrites possess high permittivity, high permeability, and extremely low d.c. conductivity [1-4]. Constitutive parameters of ferrites are frequency-dependent. Different types of ferrites have different frequency characteristics, and their frequency characteristics may be tailored by choosing appropriate chemical structure, adding dopant ions, and setting a regime for synthesis. Wideband frequency characteristics can be formed by using mixtures of different types of ferrites or statistical distribution of their physical parameters (geometry of ferrite particles, their orientation, deviation of their internal field of magnetic anisotropy, etc.).

Study of a CRAM containing a mixture of hexagonal ferrite powders in a dielectric bond material is of a special interest. Resonance absorption of electromagnetic energy in such a CRAM takes place close to the frequencies of natural ferromagnetic resonance (NFMR) of hexagonal ferrite inclusions, specifically, in the microwave band. The phenomenon of the NFMR can be employed effectively without any bias magnetic field [5, 6]. Typically, hexagonal ferrites effectively absorb electromagnetic energy due to the NFMR at frequencies higher than approximately 2.5 GHz (up to 200 GHz), because their internal field of magnetic crystallographic anisotropy \( H_A \) ranges from a few units to a few dozens of kiloersted [7-9]. In the frequency range of the NFMR, due to the dramatically increasing permeability values \( \mu' \) and \( \mu'' \) (where complex permeability is \( \mu = \mu' - j\mu'' \)), there is a substantial increase of electromagnetic energy absorption (up to 20-30 dB). The corresponding complex permittivity \( \varepsilon = \varepsilon' - j\varepsilon'' \) also affects significantly the absorption of electromagnetic energy. However, measuring complex permittivity over a wide frequency range is a practically difficult problem, especially, when losses in the material under study are high. Herein, a new method for determining of the real and imaginary parts of \( \varepsilon' \) and \( \varepsilon'' \) for substantially lossy materials is proposed.

II. IDEA OF MEASUREMENT METHOD

The total dielectric susceptibility spectrum \( \chi_e(\omega) = \varepsilon_e(\omega) - \varepsilon_\infty \) for any dielectric is schematically shown in Fig.1 [10, 11]. It is known that in the microwave frequency range the frequency characteristic of \( \varepsilon' \) of homogeneous inorganic dielectrics, including ferrites, is practically constant [11]. To be more precise, \( \varepsilon_e(\omega) \) for ferrites follows the Debye frequency dependence [12], however, in the limited frequency range of interest (microwaves) for hexagonal ferrite composites \( \varepsilon'(\omega) \) may be considered as constant.
This property laid the basis for the proposed method of measuring complex permittivity of lossy dielectrics, including ferrite-based CRAMs.

The idea of the method is the following. First, a rectangular sample of a hexagonal ferrite composite material is made. The dimensions of the sample are chosen to completely fill the cross-section of a rectangular waveguide. The length of the sample is such that the possible gaps between the sample and the waveguide do not affect the results of measurements (typically, about 10-15 mm). Then the sample is placed in the waveguide, and frequency is linearly swept in some range \( f_{\text{min}} \ldots f_{\text{max}} \). Fig.2 shows the disposition of a sample under study in a waveguide, and Fig. 3 demonstrates the typical behavior of the reflection coefficient from the sample.

It is important to mention that measurement of \( \varepsilon \) is concurrent with the measurement of the electromagnetic energy absorption \( \alpha \). The components of the complex permittivity are determined in the characteristic points of the curve shown in Fig. 3. Depending on the chosen point of the reflection coefficient minimum, the obtained permittivity values are different, and this leads to ambiguity. Since \( \varepsilon' \) of the hexagonal ferrite CRAM does not change much over the frequency range of interest, the permittivity is measured outside the NFMR, where the magneto-dielectric medium behaves just as a lossy dielectric. It is known [13], that when engineering a material with the desired parameters and determining whether it is good enough for absorbing electromagnetic energy, the transmission coefficient through a sample in the waveguide is measured as well. The proposed method of measuring \( \varepsilon' \) and \( \varepsilon'' \) also allows for simultaneous measuring the transmission coefficient, which makes this method practically convenient.

\[
L = n \cdot \frac{\lambda_w}{2}. \tag{1}
\]

where \( n \) is the number of half-wavelengths along the length of the sample.

After some algebraic manipulations, the resultant formula for calculating \( \varepsilon' \) is


\[ \varepsilon' = \left( \frac{\lambda_0}{2} \right)^2 \left\{ \left( \frac{n}{L} \right)^2 + \left( \frac{2}{\lambda_{cr}} \right)^2 \right\}, \quad (2) \]

where \( \lambda_0 = 30 / f_i \) [cm] is the wavelength in free space, \( i = 1, 2, \ldots \), and \( \lambda_{cr} \) is the cut-off wavelength [cm]. Typically, in the rectangular waveguide, the fundamental mode TE\(_{10}\) is used. The cut-off wavelength for the TE\(_{10}\) mode is \( \lambda_{cr} = 2a \), where \( a \) is the dimension of the waveguide wide side. In the formula (2), \( n \) is the unknown parameter, which corresponds to the number (N\(_0\)) of the minimum of the reflection coefficient, and it should be found. Using the fact that the real part \( \varepsilon' \) in the neighboring minima of the reflection coefficient from the sample is practically constant with respect to the frequency, the formula (2) can be written for two neighboring minima. After some transformations, the quadratic equation for calculating \( n \) is obtained:

\[ \left( \frac{1}{f_1} - \frac{1}{f_2} \right)n^2 - 2 \frac{2L}{\lambda_{cr}} \left( \frac{1}{f_1} - \frac{1}{f_2} \right) - 1 = 0. \quad (3) \]

where \( f_1 \) and \( f_2 \) are the frequencies at the minima of the reflection coefficient [GHz], and \( L \) is the sample length [cm].

It is important to mention that the value \( n \), found as a solution of the equation (3), is not necessarily integer. This is because there is always some experimental error in determining frequencies \( f_1 \) and \( f_2 \) at the minima of the reflection coefficient, and also the length of the sample \( L \) is measured with some accuracy. To determine \( \varepsilon' \), one can use the simplified formula (2). However, to determine \( \varepsilon'' \), one must use the exact expression for complex reflection coefficient \( \Gamma \) in the rectangular waveguide with the single fundamental TE\(_{10}\) mode [14]:

\[ \Gamma = \frac{(\tilde{Z}_u^2 - (W_0^H)^2) \tanh(j \tilde{\gamma} \cdot L)}{2 \tilde{Z}_u W_0^H + (\tilde{Z}_u^2 + (W_0^H)^2) \tanh(j \tilde{\gamma} \cdot L)}, \quad (4) \]

where \( W_0^H \) is the wave impedance for the TE\(_{10}\) mode in the empty waveguide, \( \tilde{Z}_u \) is the wave impedance of the waveguide filled with a magneto-dielectric medium, \( \tilde{\gamma} \) is the complex propagation constant inside the rectangular waveguide in the magneto-dielectric medium, \( L \) is the length of the sample.

Formula (4) is used for any fixed \( \varepsilon' \) to calculate the dependence of the minimum reflection coefficient \( \alpha \) [dB] as a function of \( \varepsilon'' \) (loss in the composite medium). The frequencies \( f_1 \) and \( f_2 \) are found from the experimental frequency dependence of the reflection coefficient from the sample.

III. EXPERIMENTAL RESULTS

A. Composites containing hexagonal ferrites

Demonstration of the proposed method is the experimental determination of \( \varepsilon' \) and \( \varepsilon'' \) values for a composite medium containing a powder of the doped M-type hexagonal barium ferrite \( BaO \cdot 5.12Fe_2O_3 \cdot 0.7In_2O_3 \cdot 0.18Sc_2O_3 \) in the 3-cm wavelength region. The mass concentrations and corresponding volume fractions of the hexagonal ferrite powder and the bond material (paraffin) are as in Table 1. Two edge frequencies of the 3-cm waveband were used: 8.5 GHz and 12.04 GHz. The resultant real part of permittivity is \( \varepsilon' = 4.1 \pm 0.05 \).

<table>
<thead>
<tr>
<th>TABLE I</th>
</tr>
</thead>
<tbody>
<tr>
<td>VOLUME AND MASS FRACTIONS OF FERRITE AND PARAFFIN</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>( \rho ), g/cm(^3)</th>
<th>( m ), g</th>
<th>( V ), cm(^3)</th>
<th>( f_v ), GHz</th>
<th>( \text{f} \text{m} ) %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ferrite</td>
<td>5.0</td>
<td>1.5</td>
<td>0.3</td>
<td>0.3</td>
<td>70.4</td>
</tr>
<tr>
<td>Paraffin</td>
<td>0.9</td>
<td>0.63</td>
<td>0.7</td>
<td>0.7</td>
<td>29.6</td>
</tr>
<tr>
<td>Total</td>
<td>2.13</td>
<td>2.13</td>
<td>1.0</td>
<td>1.0</td>
<td>100</td>
</tr>
</tbody>
</table>

The loss increases with the frequency increase, and the following results are obtained. At the frequency \( f=8.5 \) GHz, the imaginary permittivity is \( \varepsilon'' = 0.14 \), and at the frequency \( f=12.04 \) GHz, the imaginary permittivity is \( \varepsilon'' = 0.186 \). The real part of permittivity \( \varepsilon' \) was calculated using (2), and the imaginary part was determined from the dependence in Fig. 4.

![Figure 4. Dependence of the reflection coefficient upon loss (imaginary part of permittivity).](image)

The effective permittivity of the composite was also calculated using the Maxwell Garnett (MG) mixing rule [15],

\[ \varepsilon_{\text{eff}} = \varepsilon_{\text{par}} + \frac{3f_{\text{fer}}(\varepsilon_{\text{fer}}/\varepsilon_{\text{par}}-1)}{\varepsilon_{\text{fer}}/\varepsilon_{\text{par}}-2-f_{\text{fer}}(\varepsilon_{\text{fer}}/\varepsilon_{\text{par}}-1)}, \quad (5) \]
In the MG model it was assumed that the ferrite particles are spherical and non-interacting with each other. The paraffin permittivity in the calculation was taken as \( \varepsilon_{\text{par}} = 2.3 - j0.025 \), and the permittivity of the bulk polycrystalline hexagonal ferrite was taken as \( \varepsilon_{\text{fer}} = 16 - j1.0 \). The calculated effective permittivity for these input data is \( \varepsilon_{\text{effMG}} = 4.02 - j0.087 \). The real part agrees well with the measured results, while the measured imaginary part is higher than the calculated.

Another set of measurements were conducted in the 3-cm waveband using the standard metal rectangular waveguide technique with the fundamental mode TE\(_{10}\). The central frequency was 9.8 GHz. A sample made of the same hexagonal ferrite \( \text{BaO} \cdot 5.12\text{Fe}_2\text{O}_3 \cdot 0.7\text{In}_2\text{O}_3 \cdot 0.18\text{Sc}_2\text{O}_3 \) in a paraffin base was studied. The results of measurements are summarized in Table II. It is seen the higher mass concentration of hexagonal ferrite leads to the higher \( \varepsilon' \) and \( \varepsilon'' \).

### Table II

**DIELECTRIC PROPERTIES OF HEXAGONAL FERRITE–PARAFFIN MIXTURE AT DIFFERENT MASS CONCENTRATIONS**

<table>
<thead>
<tr>
<th>Mass concentration, ( f_m ), %</th>
<th>Frequency, ( f ), GHz</th>
<th>( \varepsilon' )</th>
<th>( \varepsilon'' )</th>
</tr>
</thead>
<tbody>
<tr>
<td>60%</td>
<td>9.8</td>
<td>3.7</td>
<td>0.05</td>
</tr>
<tr>
<td>70%</td>
<td>9.8</td>
<td>4.1</td>
<td>0.16</td>
</tr>
<tr>
<td>70.4%</td>
<td>8.5</td>
<td>4.15</td>
<td>0.14</td>
</tr>
<tr>
<td>70.4 %</td>
<td>12.0</td>
<td>4.05</td>
<td>0.186</td>
</tr>
</tbody>
</table>

**B. PMMA**

The proposed method was also applied to determining complex permittivity of non-magnetic dielectric materials.

First, a dielectric constant of PMMA samples (Polymethylmethacrylate, a type of organic glass) was measured in the frequency range 8.5-12.0 GHz. The measured \( \varepsilon' \) for five different PMMA samples were 2.56, 2.54, 2.56, 2.52, and 2.54 (average \( \varepsilon' = 2.54 \)). Loss in PMMA samples was very low, and \( \varepsilon'' \) was not measured. This matches the reference and published data for PMMA in the microwave frequency range: \( \varepsilon' = 2.2-2.8 \), and \( \varepsilon'' = 0.01-0.3 \). [16-19].

**C. Composites containing schungite**

Shungite is known to be an amorphous carbon-rich material occurring in Precambrian schists [20]. The only so far known mine of natural shungites is in Karelia (Russia). Recently, interest of engineers to shungites has increased, since they possess an amazing morphology. The schungite carbon \( C_{60} \) has a fullerene metastable molecular structure that is not likely to graphitize. The main element of a schungite carbon is a globule – a multilayer structure of about 10 nm diameter with a pore inside. Schungite forms a matrix that contains periodically arranged dispersed silicates of about 1 \( \mu \text{m} \) size. This structure makes schungites attractive for engineering applications that require effective absorption of electromagnetic radiation (see, for example, [21]).

Two groups of samples were prepared and studied: (1) pure shungite in a paraffin base; and (2) cobalt added to shungite. The graphs of real and imaginary permittivity for schungite-paraffin composite versus mass concentration of schungite are presented in Fig. 5 (a, b). As is seen from these figures, both \( \varepsilon' \) and \( \varepsilon'' \) increase with mass concentration of schungite. When cobalt is added to schungite (\( f_m = 50% \)), both \( \varepsilon' \) and \( \varepsilon'' \) have a trend of increasing with the increase of concentration of cobalt, as is shown in Fig. 6 (a, b).
Another series of tests were measuring complex permittivity of a sample made of alabaster, which is a type of plaster, or clay. The sample is made of alabaster powder mixed with water for hardening. It was dried naturally at room temperature of 22°C and air humidity of 23% for a few days, and its permittivity was measured by the proposed method in the frequency range 8.5-12.0 GHz. Both $\varepsilon'$ and $\varepsilon''$ decrease as the sample dries. The results of measurements are presented in Table III.

The published data differs from the published data. The possible reasons are, most likely, because (1) our sample was made of alabaster, but not plaster; and (2) there is possible uncontrolled porosity in our sample under study, which leads to the lower values of $\varepsilon'$ and $\varepsilon''$, especially, when the sample dries up.

**TABLE III**

<table>
<thead>
<tr>
<th>Days</th>
<th>$\varepsilon'$,</th>
<th>$\alpha$,</th>
<th>$\varepsilon''$,</th>
<th>$\tan \delta$</th>
<th>Humidity in the sample</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>5.2</td>
<td>-10</td>
<td>0.2</td>
<td>0.038</td>
<td>high</td>
</tr>
<tr>
<td>5</td>
<td>5.15</td>
<td>-14</td>
<td>0.18</td>
<td>0.034</td>
<td>high</td>
</tr>
<tr>
<td>7</td>
<td>4.8</td>
<td>-18</td>
<td>0.142</td>
<td>0.029</td>
<td>intermediate</td>
</tr>
<tr>
<td>9</td>
<td>3.7</td>
<td>-23</td>
<td>0.07</td>
<td>0.019</td>
<td>intermediate</td>
</tr>
<tr>
<td>10</td>
<td>3.4</td>
<td>-26</td>
<td>0.034</td>
<td>0.01</td>
<td>low</td>
</tr>
<tr>
<td>12</td>
<td>3.3</td>
<td>-31</td>
<td>0.01</td>
<td>0.003</td>
<td>low</td>
</tr>
</tbody>
</table>

IV. CONCLUSION

The method of measurement proposed in this paper is simple and can be used to determine microwave complex permittivity of a lossy material, including composite radio absorbing materials based on ferrites that exhibit natural ferromagnetic resonance. There is a good agreement between the measured complex permittivity of a composite absorbing material containing hexagonal ferrite powder and computations based on the Maxwell Garnett formulation. The proposed technique was also applied to measuring complex permittivity of non-magnetic dielectric (PMMA, schungite-paraffin composites, and alabaster).

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