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Engineering, Modeling and Testing of Composite Absorbing Materials for EMC Applications

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1. Introduction

Non-conducting wideband absorbing materials are important for improving immunity of electronic equipment and solving various problems of electromagnetic compatibility (Celozzi et al., 2008). Application of absorbing materials for the design of shielding enclosures, coatings, gaskets, or filtering patches is preferrable compared to metal structures for several reasons. Typically enclosures of electronic equipment are made of highly-conducting metal to achieve the required shielding levels (Neelakanta, 1995). However, requirements to make electronic devices of smaller size and weight neccessitate substitution of metal by electrically conducting composites (Sichel, 1982) and polymers (Fox et al., 2008; Wang & Jing, 2005). Moreover, if a conducting surface has sharp edges, slots, and apertures, currents on this surface may drive unintentional antennas and enhance noise coupling paths as culprits of undesirable emissions (Paul, 2006). One of the ways to eliminate this problem is to use absorbing materials, including applications of magneto-dielectric composite materials for electromagnetic shielding purposes.

Engineering of absorbing materials with desirable frequency responses and advanced physical properties is of great importance. These materials can be either bulk or sheet, and, depending on a particular application, they may be shaped as needed. Frequency characteristics of composite absorbing materials may be either wideband, or frequency selective. The materials of interest should provide the required attenuation of surface currents on the extended conducting surfaces, and/or satisfy requirements on damping electromagnetic near-fields of the sources, or far-field electromagnetic waves of the given configuration, direction of incidence, and polarization. Advanced physical, chemical, and mechanical properties of absorbing materials are of great importance as well. The materials must comply with requirements on thermal, chemical, and mechanical stability and durability, incombustibility, non-toxicity, environmental friendliness, and adhesion with surfaces on which they will be placed.

It is important that when applied to electronic products, the engineered absorbing materials would allow for compliance with requirements and regulations from the point of view of

electromagnetic compatibility (EMC), electromagnetic immunity (EMI), signal integrity (SI), and power integrity (PI) over frequency ranges of interest. For applications in high-speed digital electronic designs, the materials that would absorb electromagnetic energy in RF, microwave, and potentially mm-wave bands, are of special interest. The necessity of using absorbing materials becomes more and more important, since operating frequencies of electronic devices increase, package density grows dramatically, and the number of electronic equipment and devices of mass production continues to increase. Thin absorbing noise-suppressing composite sheets and coats are of special interest for such applications, especially because of microminiaturization trends and of convenience to apply directly on the surfaces to be protected, as well as their broadband performance. Some examples of applying thin sheet absorbing materials are shown in Fig. 1. Thin sheet absorbers can wrap cables, or be applied directly on the sources of noise, or at some optimal distance from the source. They can be put directly on the electronic module enclosure, around or over vents, holes, sharp edges, and wedges, or may be placed as patches inside cavities to damp unwanted resonances.
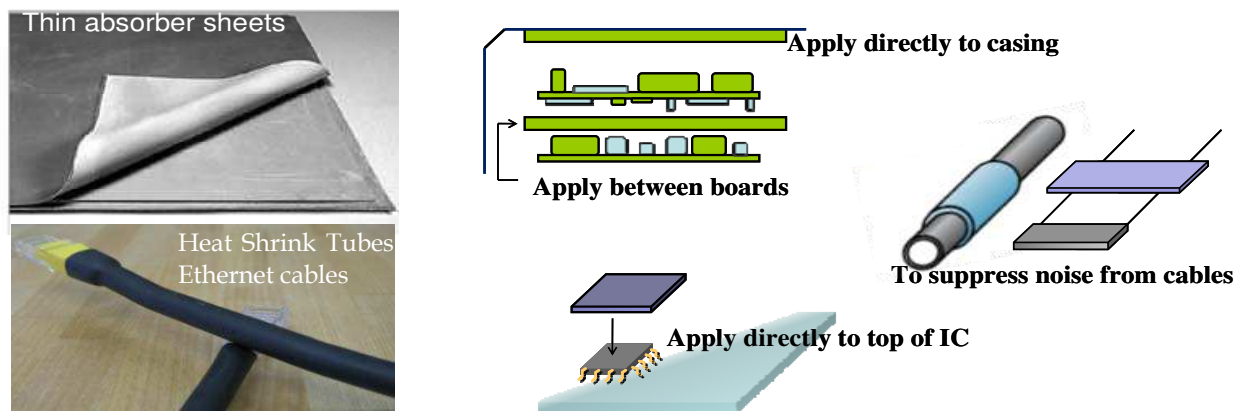

Fig. 1. Some examples of applying thin sheet absorbing materials

Also, electronic composites, whose properties can be controlled by thermal or electromagnetic means, play an important part in modern micro- and nano-electromechanical systems such as sensors, filters, switches, and actuators (Taya, 2005). However, they will not be considered herein, since the specific topic of the present work is absorbing composites for electromagnetic waves and fields.

An absorbing, non-conducting composite material may contain conducting non-magnetic inclusions (*e.g.*, carbon or non-magnetic metal particles) at concentrations below the percolation threshold as fillers in a dielectric host (matrix, base) material (Lagarkov & Sarychev, 1996; Sihvola, 1999; Tretyakov, 2003). Electromagnetic wideband radar absorbing materials may include conducting magnetic (*e.g.*, iron or permalloy) powders (Birks, 1948; Merrill et al., 1999; Absinova et al., 2007) or combinations of granular ferrite and conducting particles, e.g., carbonyl-iron powders (Park, et al., 2000). Electromagnetic wave absorbers also widely use non-conducting soft ferrites with spinel structure, e.g., Ni-Zn, Ni-Zn-Co, or Mn-Zn ferrites (Naito & Suetake, 1971; Shin & Oh, 1993; Kazantseva et al., 2004; Lisjak et al., 2006), or hexagonal ferrites (Mikhailovsky et al., 1965; Ota et al., 1999; Iijima et al., 2000). Combining inclusions of different types (dielectric, conducting, and magnetic) in a multi-phase composite material may yield substantial increase in the absorption level in the

desired frequency range. The absorbers can form either thick or thin films (paint) placed on the surfaces to be protected from unwanted radiation or coupling paths, or foam, as in anechoic chambers. The absorbing materials may form multilayered resonant structures, such as a Jaumann absorber or a Salisbury screen (Knott et al., 2004).

A current objective is to develop a methodology to efficiently design and evaluate novel products based on absorbing magneto-dielectric composite materials primarily for EMC/EMI purposes. Fig. 2 shows a flowchart for engineering design of noise-suppressing materials and electromagnetic filtering structures.
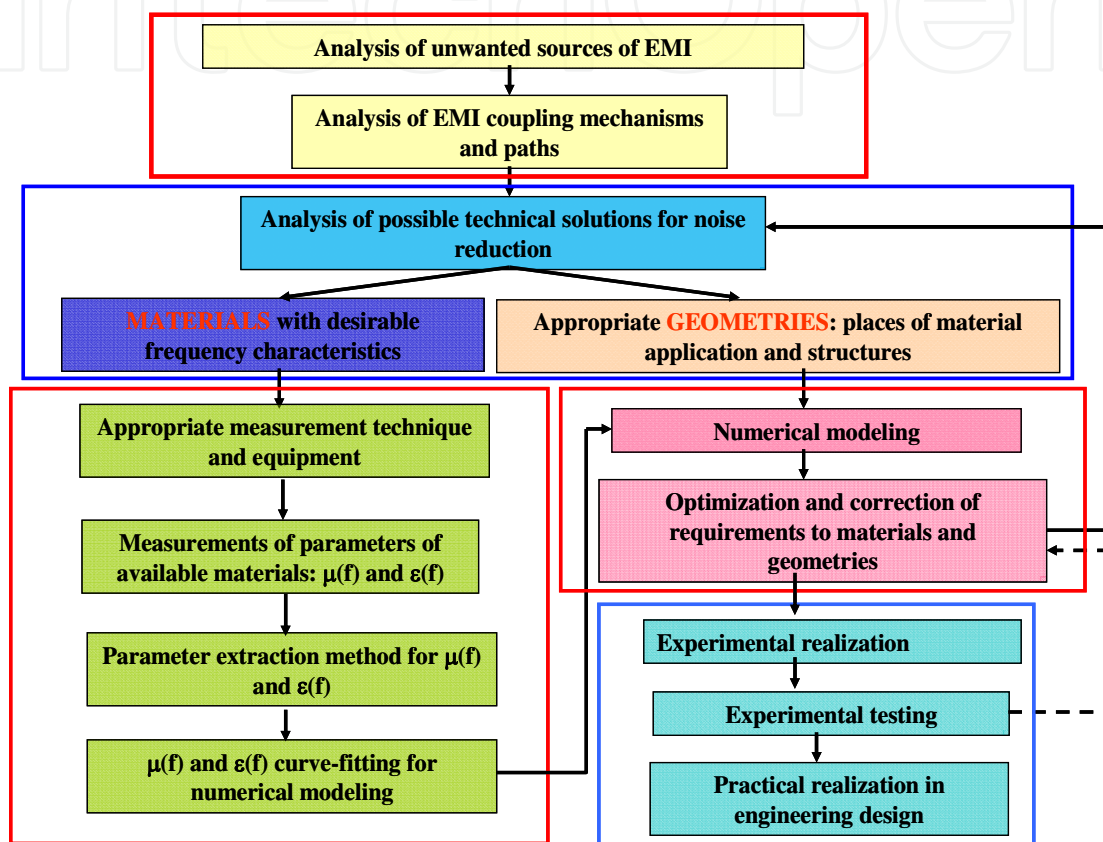

Fig. 2. Flowchart for engineering design of EMI noise-suppressing materials and structures

The analysis starts from diagnozing and caracterizing sources of unwanted radiation and/or undesirable electromagnetic interference coupling paths and mechanisms. Then possible technical solutions to eliminate or minimize those noise effects are proposed and analyzed. Any technical solution is based on a combination of proper material (or materials) and geometry. This means deciding, where the chosen materials should be placed, and what their configuration should be to achieve the required noise reduction in the intended frequency and dynamic (power or field amplitude) ranges. If choosing an appropriate material among already existing – either available commercially, or specially synthesized in laboratory conditions, it is important to measure their electromagnetic properties. These are the complex frequency-dependent dielectric susceptibility $\chi_e(\omega)$ or permittivity $\varepsilon(\omega) = \varepsilon_\infty + \chi_e(\omega)$, and complex magnetic susceptibility $\chi_m(\omega)$ or permeability $\mu(\omega) = 1 + \chi_m(\omega)$ in the frequency range of interest. Alternatively, a material is characterized by its dielectric and magnetic susceptibility kernels $\xi_e(t)$ and $\xi_m(t)$, which are the impulse responses corresponding to $\chi_e(\omega)$ and $\chi_m(\omega)$. There are different measurement techniques for

evaluating electromagnetic properties of materials (Chen, 2004;). The choice of a specific measurement technique depends on the frequency range of interest, material type (dielectric, magneto-dielectric, or conductive; isotropic or anisotropic; linear or nonlinear; exhibiting narrowband resonances, or having comparatively flat frequency dependence in the frequency range of interest); instrument availability; required accuracy and repeatability. There are some other factors, for example, how time-consuming and resource-consuming those measurements are. Different methods of characterization of dielectric materials are summarized in (Von Hippel, 1995; Chen, 2004), and various approaches to extract parameters of ferrite materials over different frequency ranges are described in (Polder, 1950; Rado, 1953; Mullen & Carlson, 1956; Lax & Button, 1963; Korolev et al., 2008).

Currently there are numerous automated methods for wideband characterization of dielectric and magnetic bulk materials (Nicholson & Ross, 1970; Weir, 1974; Barry, 1986), as well as of thin films (Bekker et al, 2004; Booth et al., 1994). The existing techniques are typically based on various transmission/reflection measurements, either in the frequency domain, or in time domain, in transmission lines, cavities, open space, or using special probes terminated with material samples under test. The literature is replete with various measurement techniques (Nicholson & Ross, 1970; Fellner-Feldegg, 1972; Weir, 1974; Baker-Jarvis, et al., 1993, 2001; Musil & Zacek, 1986; Ghodgaonkar et al., 1990; Zheng & Smith, 1991; Ganchev et al., 1995; Jargon & Janezic, 1996; Wang et al., 1998; Roussy et al. 2004; Ledieu & Acher, 2003; Bekker et al., 2004).

For practical EMC purposes, it is convenient to have systematic methodology to help EMC engineers to design and evaluate effectiveness of EMI noise-suppressing materials and structures based on their measured dielectric and magnetic responses. An overview of this methodology is presented in Fig. 3, corresponding to the flowchart in Fig. 2.

The dielectric and magnetic properties of the samples, depending on the measurement technique, are extracted either discrete frequency points, or over selected continuous frequency bands. Then the continuous frequency characteristics over the entire frequency range of interest can be restored. This is done using an appropriate curve-fitting technique. For further analytical and/or numerical modeling of structures containing dipersive materials, it is important that the resultant continuous material frequency dependencies would satisfy passivity and causality requirements over the entire frequency range of interest. Passivity means that the initially measured transmission and reflection coefficients in a passive system is in a physically meaningful range between 0 and 1 in unitless system, or fall below 0 in logarithmic units. Causality means that real and imaginary parts of complex permittivity $\varepsilon(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega)$ and complex permeability $\mu(\omega) = \mu'(\omega) - j\mu''(\omega)$ related through the Kramers-Krönig relations (Landau & Lifshitz, 1960; Lucarini et al., 2005), and, hence, no response could be obtained before a passive linear system has been excited by an external source. After the continuous causal complex functions $\varepsilon(\omega)$ and $\mu(\omega)$ are obtained, they can be used in building either an analytical, or numerical electromagnetic model to predict effects of the absorbing material upon characteristics of the particular noise radiation and coupling scenario under study.

Validation of theoretically predicted or numerically simulated results is an important stage of the methodology. Creating an experimental test vehicle, which is suitable and comparatively simple for modeling, and at the same time captures the main features of noise generation, coupling physics, and attenuation by an absorbing material, is probably the most difficult and time-consuming task. Mutual correction and fine tuning of the experimental setup and a modeling prototype are always required at this stage.

Test sample preparation   Material parameter measurement   Extraction of μ and ε parameters

Experimental validation

Parametrical variation

Obtaining smooth frequency dependencies

Numerical modeling

Curve-fitting to series of Debye and Lorentzian terms
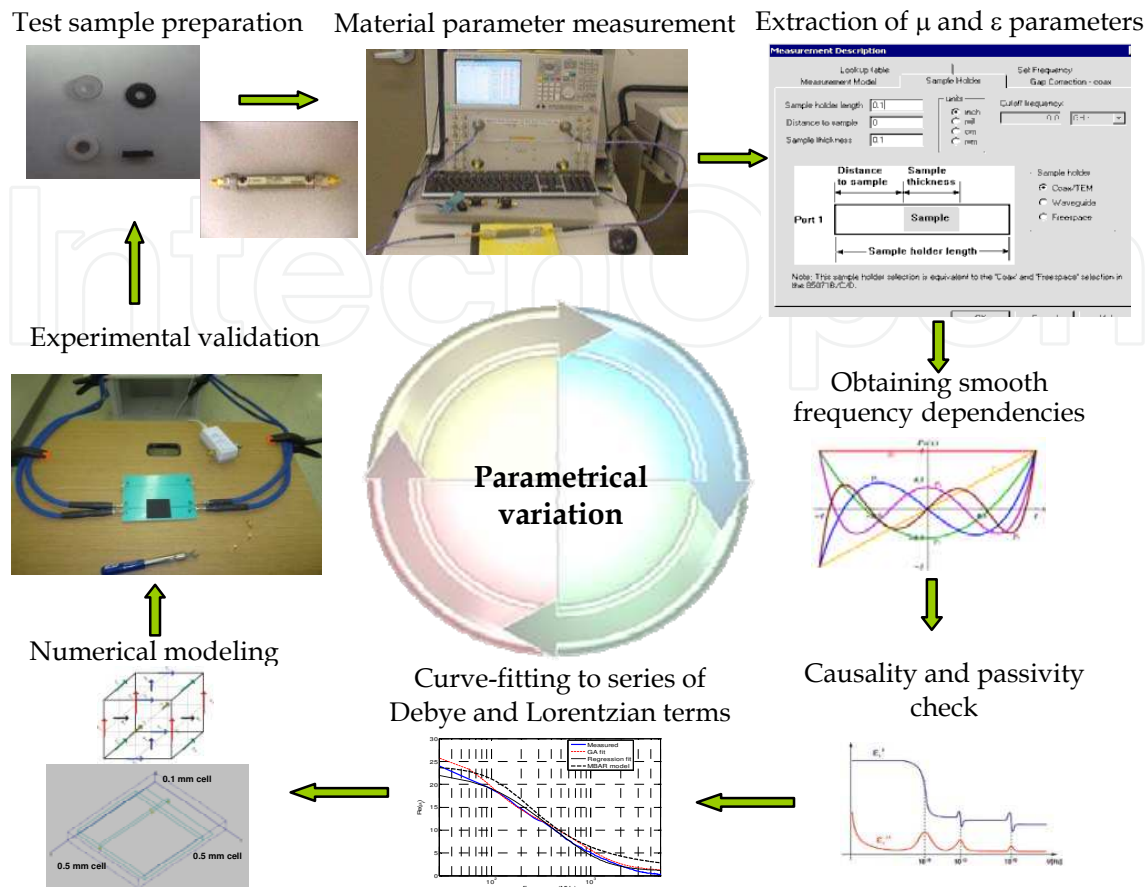
Causality and passivity check

Fig. 3. An EMC absorbing material design methodology

As soon as the agreement between measured and modeled results over the entire frequency range under consideration is achieved, the model and experimental setup may be subjected to some parametrical variations for developing practically useful design curves. Based on those design curves, applicability of the specific absorbing material in the particular practical scenario can be predicted. Different kinds of commercially available or new laboratory-synthesized materials could be compared with each other from the point of view of noise reduction effectiveness according to the imposed criteria, and the optimal solutions could be chosen.

2. Analytical model to predict frequency characteristics of a composite

Prediction of wideband frequency responses of effective electromagnetic parameters (permittivity and permeability), as well as concentration dependencies of composites, are important for engineering new electromagnetic absorbing materials. Herein, the general case of a magneto-dielectric composite material with either non-conducting ferrite, or conducting magnetic alloy inclusions in a dielectric polymer host, is considered. In addition, the composite may contain some fractions of non-magnetic conducting or dielectric inclusions. In the general case, each material phase may have its own frequency-dependent

permittivity and permeability, and the resultant homogenized material would also have α frequency dispersion of its effective dielectric and magnetic properties.

2.1 Background for developing a new mixing rule

A composite material is a medium comprised of at least two different material phases: a matrix (host, bond) material and inclusions. A matrix is typically a polymer dielectric, while inclusions may be either conducting or non-conducting, and may have their own dielectric and magnetic intrinsic properties. Depending on how many phases there are in the mixture, composites can be biphasic or multiphasic. The material phases may be intrinsically isotropic or anisotropic, depending on their crystallographic structure or morphology. The latter is determined by synthesis or manufacturing process specifics, such as temperature and pressure regimes, presence of catalysts, doping ions, and the kind of mechanical processing that was applied (milling, extruding, moulding, or pressing). Inclusions may be of identical or different sizes and shapes, and may possess the shape (form) anisotropy. In the general case, the axes of shape anisotropy do not necessarily coincide with crystal or texture axes of their constituent material. Inclusions inside the host matrix may be arranged in a regular periodic 3D or 2D structure, or may be randomly dispersed. They may either be homogeneously distributed over the space, in a the plane, or forming clusters. As for orientations of non-spherical or intrinsically anisotropic inclusions, their main axes may be all aligned, or dispersed statistically according to some distribution law. Inclusions may have any orientation with equal probability in 3D or 2D space, or may fall within some spatial angle, forming a structure with a certain rate of order.

A composite is an inhomogeneous structure. However, it can be characterized by effective electromagnetic parameters, obtained through averaging, or homogenization (Von Hippel, 1995), if sizes of inhomogeneities are much smaller than the wavelength. Any existing homogenization theory tries to ascribe effective parameters to a mixture of different phases, providing mixing rules (Sihvola, 1999). A mixing rule is an analytical formulation that describes an effective parameter as a function of frequency and concentration. Hence, it is very desirable that effective permittivity and permeability of a composite would be governed by the same mixing rule. For example, it has been recently experimentally observed that μ and ε of composites with multi-domain magnetic inclusions may be governed by analogous (dual) mixing rules (Rozanov et al., 2009).

There is currently a multitude of mixing rules in the literature based on different homogenization theories and approaches. They are applicable to different types of mixtures, rate of generalization, and depend on which parameters are homogenized, and what their limits are. This is not aimed at a review of all existing homogenization theories and models. A good review of existing mixing rules can be found in the current literature (Landauer, 1978; Sihvola, 1999; Diaz et al., 1998) and references therein.

There are a few most widely used mixing rules, which are briefly reviewed herein. The first is the Maxwell Garnett (MG) formulation (Maxwell Garnett, 1904, 1906), which results in the lowest estimate of the effective parameter. The MG rule in terms of the relative dielectric or magnetic susceptibilities, expressed through the corresponding dielectric and magnetic contrasts between inclusion and host materials

$$\alpha = \left\{ \begin{array}{c} \varepsilon_{incl} \, / \, \varepsilon_{host} - 1 \\ \mu_{incl} - 1 \end{array} \right\} \tag{1}$$

and effective parameters of the mixture and the host

$$\beta = \left\{ \begin{array}{c} \varepsilon_{eff} / \varepsilon_{host} - 1 \\ \mu_{eff} - 1 \end{array} \right\}, \tag{2}$$

can be written as

$$\frac{\beta}{1+n\beta} = \frac{p\alpha}{1+n\alpha}. \tag{3}$$

The parameter p is the volume concentration of inclusions in the composite. The parameter n describes effective depolarization or demagnetization factors, often called „form factors" of inclusions (Landau & Lifshitz, 1960). The form factor for calculating permittivity and permeability should be the same. This shape factor is $n=1/3$ for the particular case of spherical inclusions.

The second widely used theory is the Bruggeman symmetric rule (BSR), which is also known as the effective medium approximation (EMA) (Bruggeman, 1935, 1936), as is

$$\frac{p(\alpha-\beta)}{\beta+1+n(\alpha-\beta)} - (1-p)\frac{\beta}{\beta+1-n\beta} = 0. \tag{4}$$

For the permittivity of a metal-dielectric mixture, the BSR is conventionally considered the most suitable theory, because it allows for predicting the percolation threshold p_c (McLachlan, 1990). The latter is the concentration at which the composite turns from a non-conducting to a conducting state, and where the real part of effective permittivity of the mixture tends to infinity. As follows from (4), the percolation threshold should be equal to the average form factor, $p_c = n$. However, even for a mixture of spherical particles with $n=1/3$, p_c may vary over a wide range, since the percolation threshold is associated with interactions between inclusions and matrix. It has been experimentally shown, for example, that in composites filled with carbon black inclusions of almost identical spherical shape, but with different polymer host materials, the percolation threshold varied from about 5 to 50% volume fraction of inclusions (Miyasaka et al., 1982). The experimental data do not agree with the BSR prediction of $p_c \approx 30\%$. Moreover, even for a composite containing carbonyl iron powder (CIP) with almost perfect spherical inclusions, the experimentally obtained effective form factor both for permittivity and permeability may significantly deviate from 1/3 (Osipov et al., 2002). This means that the BSR may not be applicable for predicting properties of magnetic composites.

The third important mixing rule is the the Bruggeman asymmetric rule (BAR), or 1/3-power rule (Bruggeman, 1935; Hanai, 1960; Neelakanta, 1995)

$$(\beta+1)^{\frac{1}{3}} \cdot (1-p) = \frac{\alpha-\beta}{\alpha}. \tag{5}$$

This rule is fit to the composites of randomly distributed spherical inclusions, and can be applied to both dilute and dense mixtures.

The Landau-Lifshitz-Looyenga (LLL) formula (Landau & Lifshitz, 1960; Looyenga, 1965; Dube, 1970),

$$(\beta+1)^{1/3} - 1 = p\left\{(\alpha+1)^{\frac{1}{3}} - 1\right\}, \tag{6}$$

plays a special part among the mixing rules. The LLL formula is rigorous at low inclusion-matrix contrast $\alpha \ll 1$, and, hence, at high frequencies ($\omega \to \infty$). Thus any mixing rule should converge to the LLL limit at the high frequencies. This is important, because predicting frequency dependences of the material parameters must include the high-frequency region. For example, it can be shown that at $n = p_c = 1/3$, the BSR satisfies the LLL high-frequency limit. However, the parameters p_c and n are of a different physical nature: the percolation concentration p_c describes collective properties of inclusions, while shape factor n is an individual characteristic of an inclusion related to its depolarization or demagnetization at the boundary surface. This means that in a physically meaningful mixing formula these two parameters should be uncorrelated "free" fitting parameters for any particular composite. Such splitting of p_c and n as two independent physical properties can be found in a formulation proposed by Odelevsky (Odelevsky, 1951) for calculating static effective permittivity of a metal-dielectric mixture,

$$\varepsilon_{static} = 1 + \frac{p \cdot p_c}{n(p_c - p)} . \tag{7}$$

The formula to describe static magnetic properties of composites containing ferromagnetic inclusions in a dielectric host is analogous to (7)

$$\mu_{static} = 1 + \frac{p \cdot p_c \cdot (\mu_i - 1)}{n(p_c - p)(\mu_i - 1) + p_c} . \tag{8}$$

Odelevsky's formulas (7) and (8) fit experimentally obtained static data (Rozanov et al., 2009), and it can be used for finding intrinsic permeability and permittivity of inclusions (without any host material). This is very important, since in majority of cases these values are not known and cannot be found by any other methods, especially, intrinsic permeability of magnetic inclusions, which is different from the bulk materials. The latter significantly depends on the way of crushing the bulk magnetic material and the crush parameter ξ, which is a ratio of an average gap between magnetic particles in the crushed bulk material to an average size of a magnetic particle, or grain (Tsutaoka, 2003). At low frequencies ($\omega \to 0$), Odelevsky's formulas (7) and (8) converge to the corresponding MG formula (3) for $p_c = 1$. When $n = p_c = 1/3$, formula (7) for effective permittivity becomes equivalent to the BSR (4), but formula (8) for effective permeability never turns to (4). Moreover, both (7) and (8) do not satisfy the LLL limit at high frequencies ($\omega \to \infty$). For this reason, Odelevsky's theory is applicable only for static parameters.

The mixing rules (7) and (8) are formulated for a quasi-static assumption, which means that the inclusion size is much less than the shortest wavelength in the composite. Hence the frequency dependence of the effective material parameters of the mixture is associated with frequency dispersion in its ingredients. If the host martix is almost non-dispersive, the frequency dependence of the mixture will be totally determined by the frequency responses of the inclusions.

There are also complex mixing rules, which are based on cluster theories, e.g., (Sheng, 1980; Hui & Stroud, 1986; Musal et al., 1988; Doyle & Jacobs, 1990). They describe different groups of inclusions (clusters), using different mixing rules depending on their applicability. Then separate clusters are mixed using some other mixing rule at the higher level of homogenization. In some cases, these cluster theories provide good agreement with

experiment, but due to their complexity, choice of partial mixing rules is a matter of art rather than science, and there are no general recommendations that could work for any type of a composite.

Any mixing rule should describe effective permittivity and permeability using the same mathematical formulation due to the duality of ε and μ. However, existence of numerous mixing rules in the present-day literature indicates that there is no unique rule that is be applicable for describing simultaneously dielectric and magnetic properties of magneto-dielectric composites over a wide frequency range. Researchers make attempts to model effective properties for particular cases of mixtures, and any mixing theory has its applicability limitations. Thus, for an important case of composites filled with ferromagnetic metal powders, currently there is no standard unified and experimentally validated mixing rule to calculate dependences both of frequency and concentration.

2.2 A new mixing rule based on a spectral function approach

Alternative to mixing rules, properties of composites can be considered in terms of the Bergman–Milton (BM) *spectral function theory* (Bergman, 1978; Stroud et al., 1986; Bergman & Stroud, 1992; Milton, 2001). A spectral function is an unambiguous and universal characteristic of a composite (Bergman & Stroud, 1992). It describes the statistical distribution of inclusions with respect to their form factors. The spectral function can take into account variations of the effective form factor due to interactions within the mixture, as well as the spread of inclusion form factors. This is an important and favorable feature of the BM theory, as opposed to the existing mixing rules and effective medium theory. The effective normalized susceptibility of the composite can be found as

$$\beta = \int_0^1 \frac{\alpha B(n)}{1+n\alpha} dn, \tag{9}$$

where $B(n)$ is the spectral function. The sums rules for a spectral function (Bergman, 1978; Fuchs, 1978),

$$\int_0^1 B(n)dn = 1 \quad \text{and} \quad \int_0^1 nB(n)dn = \frac{p(1-p)}{D}, \tag{10}$$

provide an agreement with the LLL theory. The parameter D is the dimensionality of the composite, e.g., D=2 for the case of infinite cylinders, and D=3 for arbitrary-shaped inclusions of a bulk isotropic composite.

All the known mixing rules are particular cases of the general BM theory with their own spectral functions. Examples of spectral functions for a few different mixing theories are presented in Fig. 4. For convenience of plotting, the functions $nB(n)$ are presented instead of just $B(n)$. The calculations are made for the mean form factor of $\hat{n}$=1/3, and the volume concentration of inclusions of p=0.25. Each spectral function "peak", or continuous region, corresponds to some frequency dispersion range.

The spectral function of the MG theory is the δ−function, concentrated at the average form factor $\hat{n}$, which is around 1/3,

$$B(n) = p \cdot \delta\{n - (1-p)\hat{n}\}. \tag{11}$$

The spectral function for the effective medium BSR is

$$B(n) = \begin{cases} \dfrac{D\sqrt{(n-n_1)(n_2-n)}}{4\pi}, & \text{for } n_1 < n < n_2 \\ 0, & \text{otherwise} \end{cases}. \qquad (12)$$

In (12), n_1 and n_2 are the range of the possible form factors of inclusions, and D is the composite dimensionality. The BSR spectral function is a semi-circle in terms of $nB(n)$. It can be seen that the BAR has an asymmetric spectral function $nB(n)$ with a higher spectral density at a lower form factors n. Sheng's cluster theory (Sheng, 1980) results in a few separate regions of the $nB(n)$ function. However, there is no physically meaningful explanation, why the distribution with respect to form factors is not continuous in the cluster theory.

Though the BM spectral function approach is general for taking into account physically existing spread of inclusion parameters, it is not widely applied to analyze actual experimental data. The main reason for this is that obtaining the spectral function is not a straightforward procedure. Equation (9) is the integral relation for the intrinsic and effective parameters, and a simple algebraic representation is not always possible.

One of the most practically useful approaches that apply the notion of the BM spectral function is the Ghosh−Fuchs formulation (Ghosh & Fuchs, 1988),

$$B(n) = \begin{cases} C(n-n_1)^{1-A}(n_2-n)^{B}, & \text{for } n_1 < n < n_2 \\ 0, & \text{otherwise} \end{cases}. \qquad (13)$$

In this theory, the spectral function is taken the same as in the BSR, but with five fitting parameters: A, B, C, n_1, and n_2. C, n_1, and n_2 determine the amplitude, position, and the "peak" width of the spectral function. As a result, the width of the spectral function may vary over some range to take into account the actual spread of the inclusion form factors. The critical exponents A and B are introduced to get agreement with percolation theory. If $A=B=1/2$ in (13), and $C = D/(4\pi)$, then the spectral function will be the same as in the BSR (12).
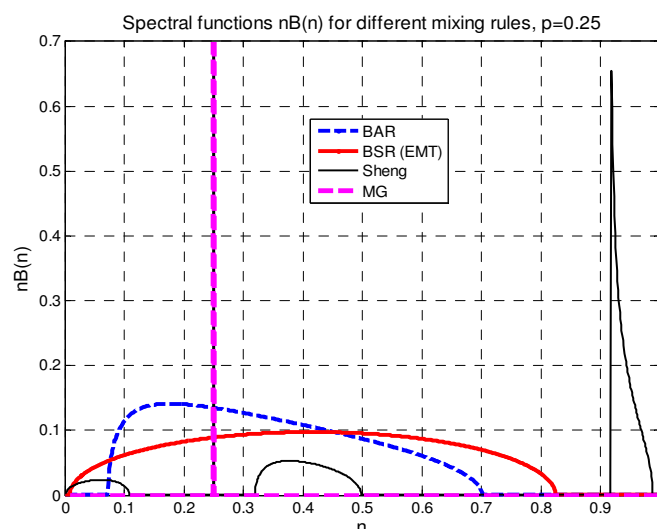

Fig. 4. Spectral functions $nB(n)$ for the BAR, BSR, Sheng's theory and MG mixing rule

The formulation (13), in contrast to the other mixing rules, agrees very well with experiments for both permittivity and permeability of composites containing ferromagnetic inclusions (Rozanov et al., 2009). However, (13) is not convenient for practical use because of cumbersome integral representation for the spectral function, and the necessity of using many (five) fitting parameters. Hence, it is appealing to have a simplified formalism dealing with algebraic operations.

The first step in the development of a simplified model is to set the critical exponents $A=B=1/2$ in the spectral function (13). This assumption is consistent with the BSR, but is different from the classical percolation theory. The reason is that the classical percolation behavior of material parameters is observed only in soot-filled (carbon black) polymers. As for the other materials, it is difficult to make them with concentration close to percolation, and even if it is possible, frequency dependence of permittivity may differ significantly from predicted by the percolation theory. One of the reasons may be imperfect contacts between particles in conducting clusters, since cluster conductivity depends mainly on these contacts, but not geometry of clusters, as the percolation theory assumes. Hence, it is reasonable to exclude the critical indices different from 1/2 from further consideration. This assumption would allow for further representing the formulation (13) in a simple algebraic form, analogous to the BSR. Also, the scaling coefficient is set $C=1$, as follows from the sums rule (10). It is also important that two limits - the LLL at high frequencies

$$\omega \to \infty, \ \alpha \ll 1: \ \ \beta = p\alpha + \frac{p(1-p)\alpha^2}{D}, \tag{14}$$

and Odelevsky's static case will be satisfied,

$$\omega \to 0, \ \alpha \gg 1: \ \ \beta = 1 + \frac{p}{n} \cdot \frac{p_c}{p_c - p}. \tag{15}$$

Since the spectral function chosen is a particular case of the BSR formulation, which is basically a quadratic equation, as follows from (4), the effective medium solution can be written as

$$\beta = Q\left(R + \alpha \pm \sqrt{(\alpha - S)^2 - T}\right), \tag{16}$$

where Q, R, S, and T are unknown coefficients, but they can be found uniquely using the limits (14) and (15). These coefficients depend on the physical parameters: composite dimensionality, inclusion concentration, average form factor, and the percolation threshold. In the BSR and in (16), the percolation threshold is determined by switching the sign ($\pm$) in the solution of the quadratic equation for α_n effective parameter. This is a consequence of the material passivity, i.e., energy can dissipate, but not generate in the material.

Then the resultant equation corresponding to the new mixing rule can be written as

$$(1-p)p_c\beta^2 - D(p\alpha - \beta)\left[p(p_c + n\beta) - np_c\beta\right] = 0. \tag{17}$$

A consequence of this new formulation is that only two fitting parameters ($n = \hat{n}$ and p_c) are used to approximate the spectral function, as opposed to five fitting parameters in the formulation (13). These fitting parameters can be found from the concentration dependence

of permittivity of the composite. The limiting case for this formulation coincides with the LLL mixing rule, and provides a unique equation for the effective material constant (either magnetic or dielectric susceptibility) as a function of inclusion concentration, percolation threshold, and dimensionality.

The correctness of (16) can be checked for a few particular cases. If concentration of inclusions $p=1$, then (16) leads to the result $\beta = \alpha$. If the average form factor is taken as $\hat{n} = 1/3$, and the percolation threshold is not taken into account, *i.e.*, allowed to be $p_c=1$, then (16) converts to the MG formulation (3). If $\hat{n} = 1/3$, but the percolation threshold is $p_c=1/3$, then (16) transforms to (4), which is the Bruggeman effective medium theory, or the BSR.

The proposed above formulation is valid for only crumb-like inclusions in the composite, since it should satisfy the inequalities

$$1/4 < p_c < 1 \quad \text{and} \quad \sqrt{4/3} - 1 \approx 0.154 < n < 1/3 . \tag{18}$$

The results of applying the above simplified algebraic formulation to reconstruct the experimentally obtained permeability of a polymer-based CIP composite with different volume concentrations of CIP are shown in Fig. 5 (Rozanov et al., 2009). As is seen from Fig. 5, the reconstruction of experimental results well achieved over the frequency range from 10 MHz to 3 GHz. The CIP inclusions had arbitrary crumb-like shape with a mean size of inclusions around 60 μm. Fitting according to the proposed model is provided at each frequency point.
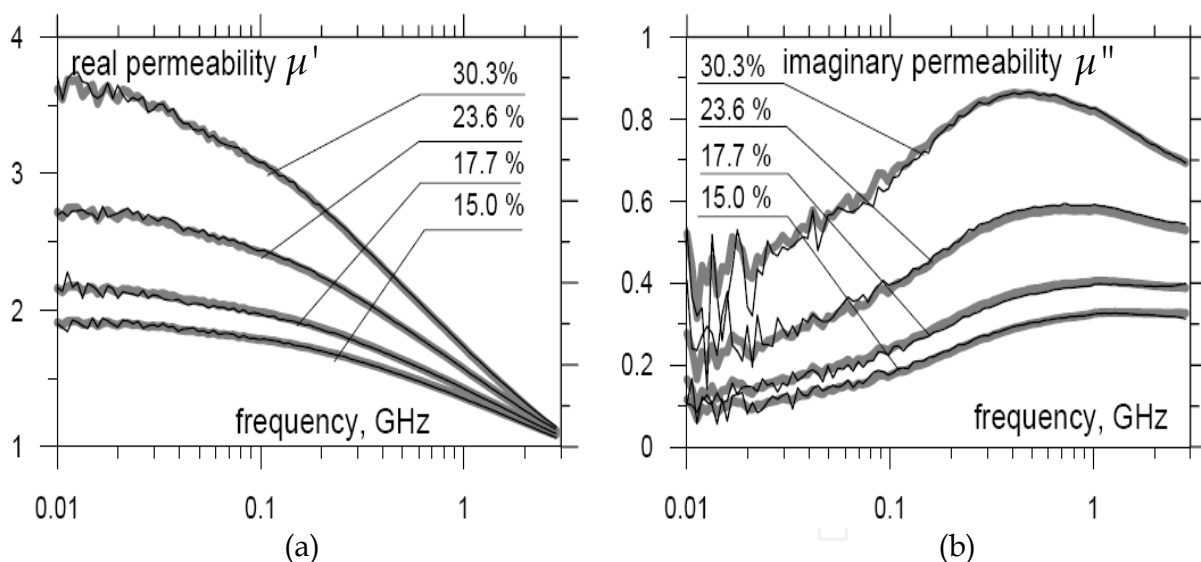

Fig. 5. Experimental (heavy grey) and modeled (thin black) curves for (a) real part of the permeability μ' and (b) imaginary part of permeability μ'' of polymer-based CIP composite with different volume concentrations of CIP (Rozanov et al, 2009)

To reconstruct the experimental data, it is important to retrieve the intrinsic permeability of inclusions. The spectral function as the distribution of inclusions with respect to varying form factor is presented in Fig. 6 (a). As the volume concentration of inclusions increases, the spectral function broadens, and this is evidence of strong cooperative phenomena, such as cluster formation. On the other hand, the spectral function has a significant width even at

the lowest volume fraction under study (15%), which means that the spread in the shapes of powder particles significantly impacts the measured permeability.
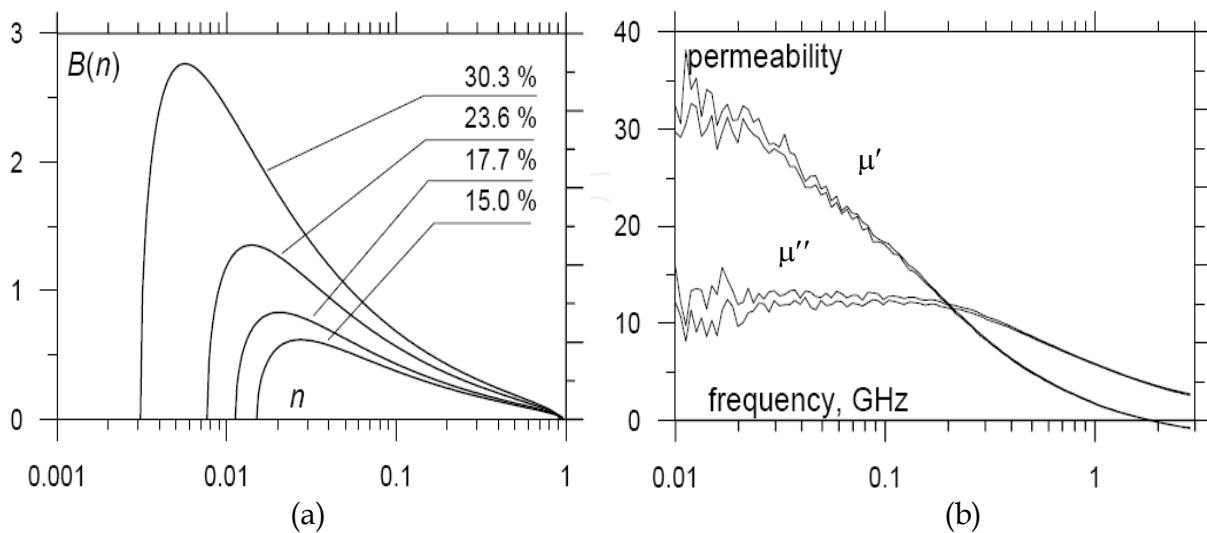

Fig. 6. Restoration of (a) spectral functions for different concentrations of CIP inclusions in the mixture (Rozanov et al, 2009), and (b) intrinsic perbeability of CIP inclusions

Fig. 6 (b) shows the frequency dependence of the intrinsic permeability obtained from different concentrations. It is seen that the intrinsic permeability obtained from different concentrations is almost identical. The reconstructed static permeability value for inclusions is $\mu_{static} \approx 30$, while the Odelevsky formula results in about $\mu_{static} \approx 17$. An additional check for the obtained result is made based on Snoek's constant of the inclusions (Snoek, 1948). This value is known to depend on the composition only, basically, on the magnetic saturation $4\pi M_s$, according to Snoek's law (Snoek, 1948; Liu, Y. et al., 2006),

$$\left(\mu_{static}-1\right)\cdot f_{res}=\frac{2}{3}\cdot\gamma\cdot 4\pi M_s, \tag{19}$$

where the gyromagnetic ratio $\gamma \approx 2.8$ GHz/kOe for iron and the majority of ferromagnetics, and f_{res} is the resonance frequency. From the microwave performance, Snoek's constant is estimated as the product of the static permeability and the ferromagnetic resonance frequency ($S = \mu_{static} \cdot f_{res}$). At the ferromagnetic resonance frequency, the real part of permeability is unity, and the imaginary goes through zero. As is seen in Fig. 6(b), it is approximately 1.3 GHz, so Snoek's constant is about $S \approx 39$ GHz, which is in a good agreement with the reference value for iron ($4\pi M_s = 21.5$ kG=2.15 T), so $S \approx 40$ GHz (Liu, Y. et al., 2006). This confirms that the static permeability of inclusions is predicted correctly, while Odelevsky's formula underestimates this value.

2.3 New mixing rule based on modified Bruggeman asymmetric rule

The proposed above theory predicts the behavior of composites containing nearly spherical inclusions. However, composites filled with magnetic needles or flakes are of great practical interest due to the possibility of achieving much higher permeability than the composites containing crumb-like particles with the same inclusion filling factor. A scanning electron microscope (SEM) picture for a powder with iron-type flakes is shown in Fig. 7.

Also, magnetic needles or flakes are favorable for the design of thin sheet materials. The formulation proposed herein is based on the Modified Bruggeman Asymmetric Rule (MBAR), which in terms of permeability is written as (Bruggeman, 1935)

$$\frac{\mu_i - \mu_{eff}}{\mu_i - \mu_{host}} = \left(1 - p\right)\left(\frac{\mu_{eff}}{\mu_{host}}\right)^{\frac{1}{3}}. \tag{20}$$

However, demagnetization factors in this formulation are missing. At the same time, it is known that the Polder-van Santen (PVS) formula, which is a consequence of the Bruggeman effective medium theory, or BSR, may account for shape factors (n_k, $k = 1,2,3$) of ellipsoidal inclusions (Polder & van Santen, 1946),

$$\mu_{eff} = \mu_{host} + \frac{p(\mu_i - \mu_{host})}{3}\sum_{k=1}^{3}\frac{\mu_{eff}}{\mu_{eff} + n_k(\mu_i - \mu_{eff})}. \tag{21}$$
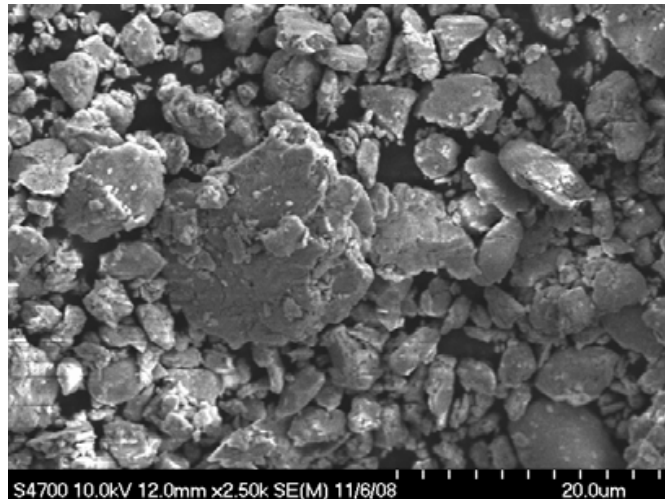
Fig. 7. SEM picture of the magnetic powder

A flake with the disk-like shape, whose aspect ratio is $u = d/h$, where d is the diameter and h is the height of the disk, can be approximated as an oblate spheroid with the axial form factor being approximately (Landau & Lifshitz, 1960)

$$n_3 \approx 1 - \frac{1}{\sqrt{1 + u^2}}, \tag{22}$$

while the other two depolarization factors are assumed to be equal, $n_1 = n_2 = 0.5(1 - n_3)$.
For high inclusion-host permeability contrast $\mu_i/\mu_{host} \gg 10$, and for volume loading of inclusions greater than ~30%, the resultant μ_{eff}, calculated using (21), is significantly higher than the values predicted by (20), especially for the low-frequency region. This happens even for an aspect ratio of inclusions close to 1 and the corresponding form factors close to 1/3, as in the spherical case. The reason is the high-frequency nature of the BSR, analogous to the LLL, and not taking into account form factors in the BAR.
The objective is to modify the BAR for permeability (20) in such a way that it would be possible to apply it to flakes. It is appealing to retrieve the correction factor which depends

on both concentration p and aspect ratio u of inclusions from (21), as a coefficient of proportionality between spherical inclusions and flakes as

$$\mu_{flake}^{PVS}(p,u) = F(p,u) \cdot \mu_{sphere}^{PVS}(u). \tag{23}$$

and then apply the same correction coefficient $F(p,u)$ to (20) as

$$\mu_{flake}^{BAR}(p,u) = F(p,u) \cdot \mu_{sphere}^{BAR}(u). \tag{24}$$

Effective permeability curves $\mu_{flake}^{PVS}(u)$ as functions of the aspect ratio u for varying inclusion concentration p are shown in Fig. 8. These curves are calculated for magnetic inclusions with intrinsic static permeability of inclusions $\mu_i = 630$.
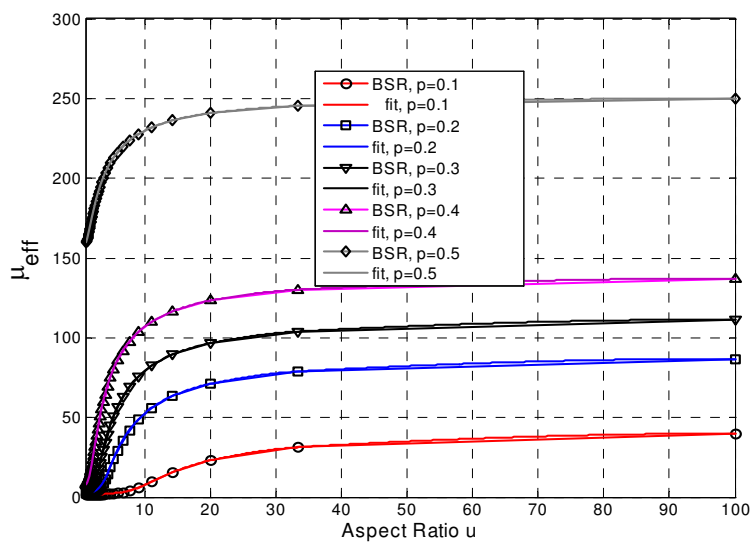
Fig. 8. Effective permeability calculated using BSR (EMT – effective medium theory) and corresponding curve-fitting to retrieve the function (p,u)

The closed-form expression for the correction factor $F(p,u)$ is obtained by analyzing numerous dependencies as those in Fig. 8, and can be well approximated by the function

$$F(p,u) = 1 + \left\{ G \cdot \tanh^2(u)(1 - H \cdot \operatorname{sech}^2(u)) / \mu_{sphere}^{PVS}(p) \right\}, \tag{25}$$

where G and H are the fitting parameters.

Fig. 9 shows the effective permeability of the mixture calculated using the modified BAR when the static permeability of inclusions is $\mu_i = 630$, the resonance frequency in the magnetic Debye curve $f_{res} = 880$ MHz, and $p = 0.25$. As aspect ratio of the plate lets increases, static permeability increases, and the loss peak shifts slightly to lower frequencies. Fig. 10 shows an example of reconstruction of experimentally obtained and curve-fitted with a sum of Debye terms (this curve-fitting will be discussed in the next Section) permeability and permittivity data for a composite containing magnetic alloy flakes with an average aspect ratio $u = 25$, volume fraction $p = 0.5$, $4\pi M_s = 4000$ G, $\mu_{static}^{bulk} = 2100$, and the crush parameter $\xi = \frac{\langle \delta \rangle_{gap}}{\langle d \rangle_{grain}} = 10^{-3}$. The frequency characteristic of the permeability, modeled using the MBAR, is approximated by the skewed Cole-Davidson dependence with exponents $s = 1.01$ and $t = 0.6$ as

$$\mu = 1 + \frac{\mu_{static} - 1}{\left(1 + (j\omega\tau)^t\right)^s} . \tag{26}$$

rather than a single-term Debye dielectric curve. The simmilar formula is used for the permittivity,

$$\varepsilon = \varepsilon_\infty + \frac{\varepsilon_{static} - \varepsilon_\infty}{\left(1 + (j\omega\tau)^t\right)^s} - \frac{j\sigma_e}{\omega\varepsilon_0} . \tag{27}$$

It is important to mention that μ_{static} in (26) is different from μ_{static}^{bulk}. μ_{static} is for the crushed magnetic material,

$$\mu_{static} = \frac{\mu_{static}^{bulk}}{1 + \xi\mu_{static}^{bulk}} . \tag{28}$$
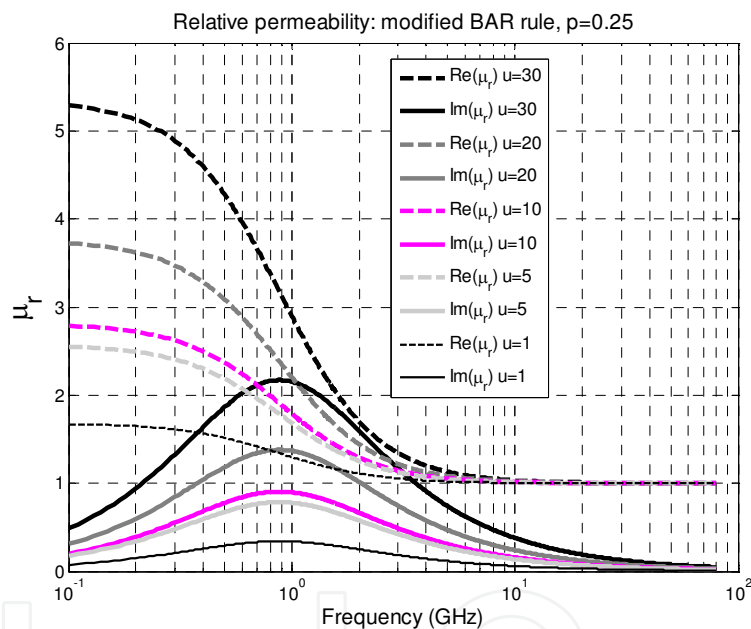
Fig. 9. Effective permeability calculated using the modified BAR, p=0.25

When modeling permittivity using the PVS equation (21), the conductivity of inclusions is $\sigma_i = 10^4$ S/m; the parameters of the silicon-based polymer are taken as $\varepsilon_{static}^{host} = 3.32$, $\varepsilon_\infty^{host} = 2.82$, and $\sigma_{host} = 5.5 \cdot 10^{-4}$ S/m. Cole-Davidson dependence exponents are s=1.01 and t=0.6. This input data gives the closest approximation to the measured results.

Another example is the reconstruction of the complex permeability function for two given volume loadings of flake-like magnetic alloy inclusions (15% and 25%), and prediction of frequency behavior of a composite with 30% volume fraction of the same inclusions. The results are shown in Fig. 11. In this case, the aspect ratio of flakes is u=110, $4\pi M_s$=9179 G, μ_{static}^{bulk}=2000, and the crush parameter $\xi = 10^{-3}$. The Cole-Davidson exponents are s=1.01 and t=0.65.

The proposed modified Bruggeman asymmetric rule allows for realistic predicting the frequency behavior of composites, if intrinsic constitutive parameters of inclusion and host

materials, their volume fractions, and morphology of the composite (shape and size of inclusions, their orientation, including statistical distribution of orientations, homogeneity and formation of clusters, *etc.*) are known *a priori*.
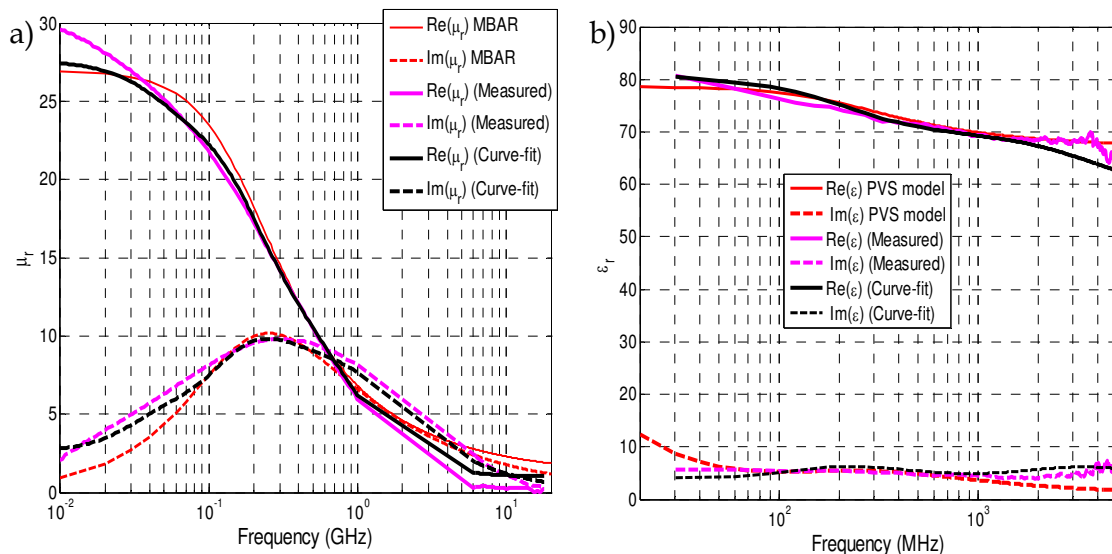

Fig. 10. Measured, curve-fitted (28), (29), and modeled (27), (28) data for (a) permeability and (b) permittivity for the test composite material
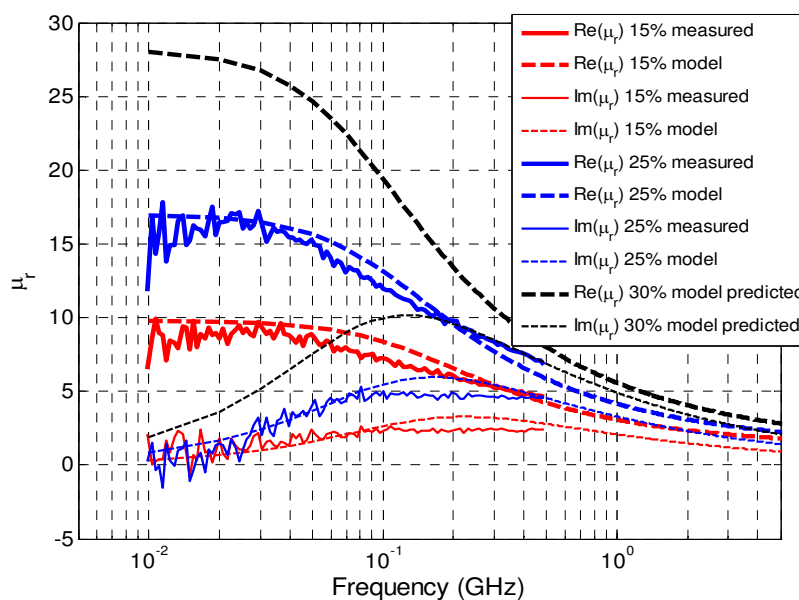

Fig. 11. Measured and reconstructed permeability for two test materials and predicted data for the third material with the different volume loading of inclusions

3. Material extraction procedure and curve-fitting for numerical modelling

A procedure for extracting complex permittivity and permeability of materials from measurements based on a travelling-wave transmission-line technique (single-ended and differential microstrip and stripline) has been applied. It includes an accurate and efficient

curve-fitting for approximating frequency dependencies of both permittivity and permeability by a series of rational-fractional functions. RF and microwave material frequency characteristics in the majority of cases can be accurately approximated by a series of non-resonance Debye and resonant Lorentzian terms, which have poles of the first and the second order, respectively, as

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_{k=1}^{N} \frac{\chi_{ek}}{1 + j\omega\tau_{ek}} + \sum_{i=1}^{M} \frac{\chi_{ei}\omega_{0i}^2}{\omega^2 - \omega_{vi}^2 + j\omega\Delta\omega_{ei}} - j\frac{\sigma_e}{\omega\varepsilon_0}, \tag{29}$$

$$\mu(\omega) = 1 + \sum_{k=1}^{N} \frac{\chi_{mk}}{1 + j\omega\tau_{mk}} + \sum_{i=1}^{M} \frac{\chi_{mi}\omega_{0i}^2}{\omega^2 - \omega_{vi}^2 + j\omega\Delta\omega_{mi}}. \tag{30}$$

The last term in equation (29) allows for taking into account the d.c. (ohmic) conductivity.

The curve-fitting procedure employs Legendre polynomials and regression analysis. The flowchart of this curve-fitting procedure is shown in Fig. 12. The measured data is interpolated to smooth curves using Legendre polynomials. Then a non-linear least-mean-square regression optimization procedure is used to approximate these curves to the series (25) and (26). For $\mu(\omega)$ and $\varepsilon(\omega)$, only Debye terms are sufficient to realize the curves, as is shown herein.

Measured μ and ε

Extracting Legendre coefficients $\quad P_n(x) = \frac{1}{2^n n!}\left(\frac{d}{dx}\right)^n \left(x^2 - 1\right)^n$

Smooth continuous curves with Legendre polynomial model

Non-linear least mean squares regression $\quad b_1 = \frac{\sum(X_i - \bar{X})(Y_i - \bar{Y})}{\sum(X_i - \bar{X})^2} \quad b_0 = \bar{Y} - b_1\bar{X}$

Regression curve fit to Debye terms

Approximating material parameters

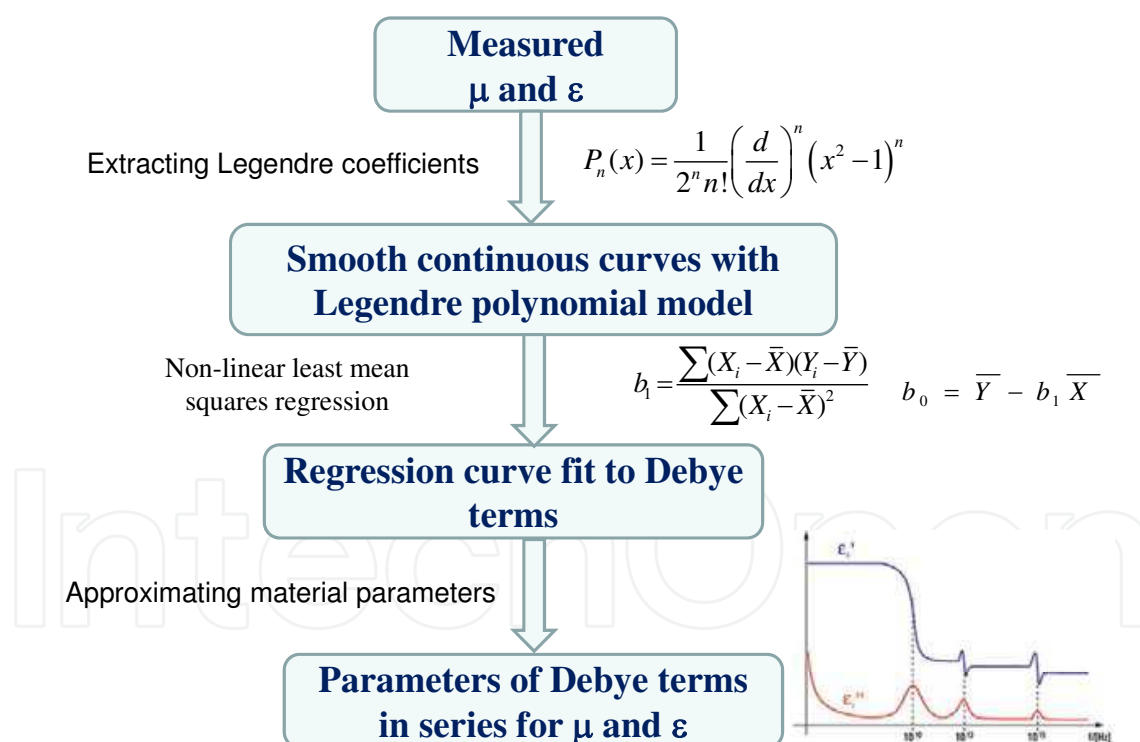**Parameters of Debye terms in series for μ and ε**

Fig. 12. Flowchart of the curve-fitting procedure

It is important that the curve-fitting procedure ensures satisfying the Kramers-Krönig causality relations, which is necessary for using these characteristics in further numerical and analytical modeling. Moreover, the proposed curve-fitting allows for restoration of the missing measurement data in some frequency intervals. For example, Fig. 10 shows the curve-fitted frequency dependences along with the measured permittivity and permeability

of the test composite material. The curve-fitting algorithm based on the Legendre polynomials and non-linear regression optimization is friendly and converges rapidly. Curve-fitting corrects non-causality of the measured responses. However, as all optimization procedures, it needs some initial guess about the Debye data to start search for the actual Debye parameters, fitting the continuous functions. These continuous functions are obtained from the series of interpolating the measured data at the discrete frequency points and smoothening using the series of the Legendre polynomials.

The curve-fitted frequency dependences of permittivity and permeability are convenient for time-domain electromagnetic numerical simulations, in particular, finite-difference time-domain method (FDTD). The advantage of time-domain techniques is that they efficiently provide broadband responses of the modeled structures. FDTD code can be used for wideband simulations of complex geometries containing frequency-dispersive materials, including a subcellular feature for modeling thin layers of absorbing materials. Both bulk cells and thin-layer sub-cells in this algorithm can include curve-fitted frequency characteristics for both permittivity and permeability.

The numerical simulations have been experimentally tested for a number of structures, such as a single-ended microstrip line, a differential microstrip line, a monopole antenna comprised of a cable driven against a ground plane, and a rectangular cavity. An example with the microstrip line covered with the test absorbing sheet is presented herein. The experimental and the FDTD model setups are shown in Fig. 13. Measured and FDTD modeled amplitude and phase of the input impedance of the open-circuited microstrip line covered with a thin sheet of absorbing material with the modeled frequency characteristics as shown in Fig. 10, is presented in Fig. 14. Good agreement with experiment validates the modeling approach.

Experimental Setup   **Numerical Model Setup (EZ-FDTD)**
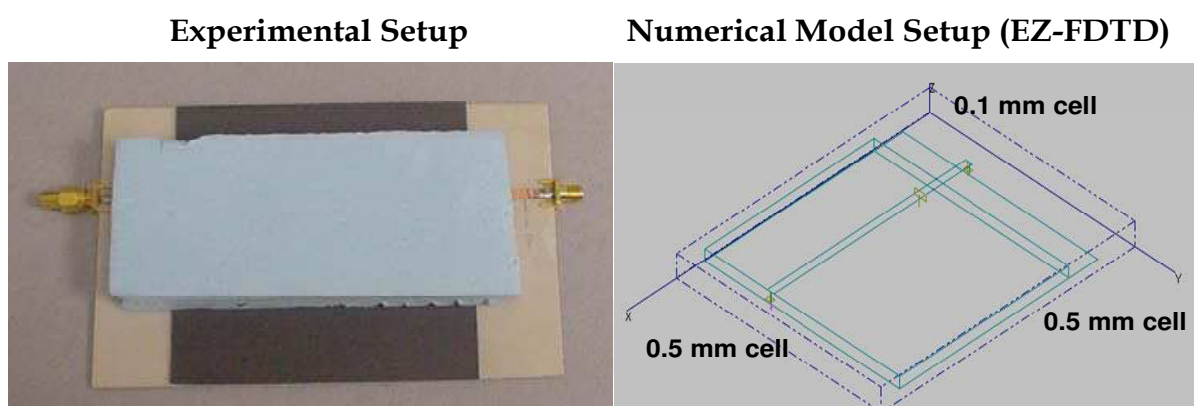
Parameters of the structure

- Length of the board is 14.7 cm.
- Trace width is 3.5 mm. FR-4-type dielectric (ε_r=3.53; $\tan\delta$=0.001), height = 1 mm.
- Characteristic impedance = 50 Ω. Absorbing sheet: 10 mm x 10 mm x 0.5 mm.

Fig. 13. Single-ended microstrip line on a printed circuit board coated with an absorbing composite layer. The thick top overlay material with $\varepsilon_r \approx 1$ is used for pressing the aborbing sheet flat
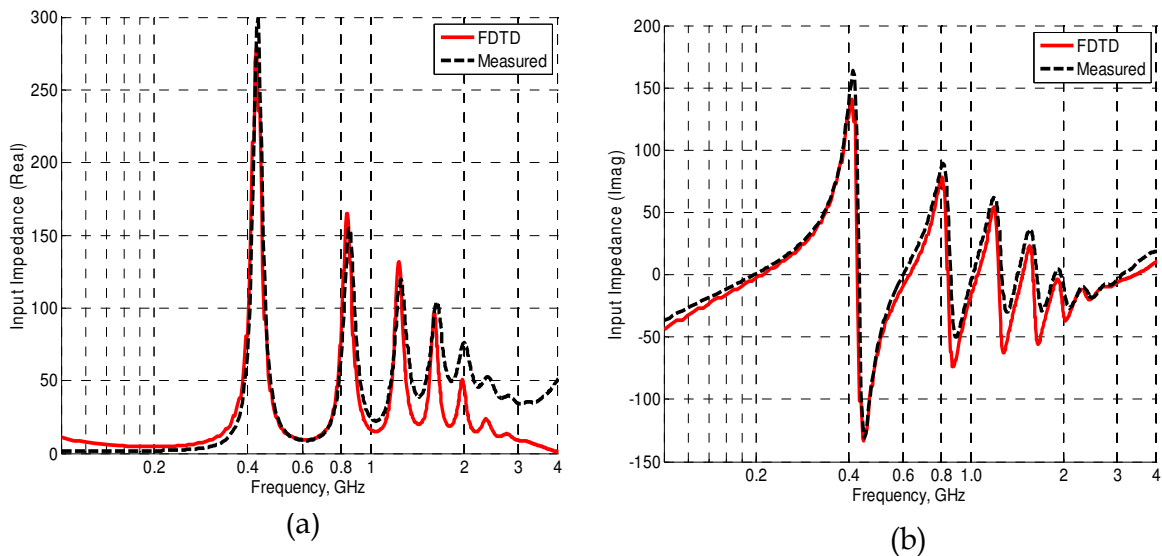
Fig. 14. Measured and FDTD modeled input impedance (in Ohms) of an open-circuit single-ended microstrip transmission line: (a) real part; (b) imaginary part

4. Conclusion

A methodology for engineering of new materials with desirable frequency characteristics and other physical, mechanical, and environmental properties, and their analytical and numerical modeling and experimental testing in different practical scenarios is presented in this work. The proposed methodology may be applied to design absorbers that would mitigate radiation from heatsinks, spurious radiation from chips and other active circuit components, suppress parasitic resonances within enclosures, and reduce radiation from cable structures. It is an effective approach for evaluating whether a material could be a successful candidate for mitigating unwanted radiation and coupling, and to optimize dielectric and magnetic properties of materials and their ingredients in composites, as well as geometries for particular practical problems.

5. Acknowledgment

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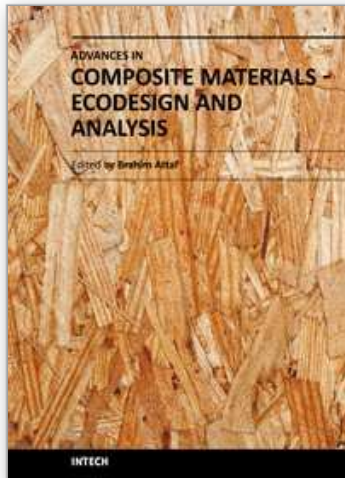
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Edited by Dr. Brahim Attaf

By adopting the principles of sustainable design and cleaner production, this important book opens a new challenge in the world of composite materials and explores the achieved advancements of specialists in their respective areas of research and innovation. Contributions coming from both spaces of academia and industry were so diversified that the 28 chapters composing the book have been grouped into the following main parts: sustainable materials and ecodesign aspects, composite materials and curing processes, modelling and testing, strength of adhesive joints, characterization and thermal behaviour, all of which provides an invaluable overview of this fascinating subject area. Results achieved from theoretical, numerical and experimental investigations can help designers, manufacturers and suppliers involved with high-tech composite materials to boost competitiveness and innovation productivity.

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