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Experimental validation of analytical models for description of soft-magnetic composites in microwave range

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Abstract. Analytical models for prediction of composite properties are long established and well tested for electromagnetic properties. But there is a smaller number of experimental studies on validity of these models for composites with significant imaginary components of electromagnetic properties. Our aim was to analyze experimentally the three well known models, the Maxwell-Garnett analytical model, effective-medium theory analytical model, and Lichtenecker logarithmic model, and their ability to calculate two properties of interest. The first is calculation of the effective properties of an arbitrary composite with volume fraction *F* of the inclusions from the measured effective properties of a specific composite with known *F*. The second is calculation of the intrinsic properties of components from the measured effective properties of a composite. In this study we focused on the nonmagnetized composites with the ferromagnetic inclusions at the microwave frequencies, where large imaginary part of the permeability is present. Our results show significant differences between the analytical models for the calculated intrinsic properties with a notable failure of the Maxwell-Garnett model. On the other hand, predicting the effective properties of an arbitrary composite from the known intrinsic properties of components or from the effective properties of a specific composite is much more robust, but the accuracy still depends on the analytical model used.

Keywords: effective medium theory, analytical models, composites, permeability, microwaves

Eksperimentalna validacija analitičnih modelov za opis elektromagnetnih lastnosti mehko-magnetnih kompozitov v mikrovalovnem območju

Že več kot sto let je v uporabi vrsta analitičnih modelov, ki omogočajo izračun lastnosti kompozitnih sistemov (mešanic) sestavljeni iz dveh komponent (npr. matrica in inkluzije). Tovrstni modeli so široko uporabljeni tudi za analizo elektromagnetnih lastnosti kompozitnih materialov. Vendar pa ne obstaja veliko ekseprimentalnih študij, ki bi analizirale veljavnost teh modelov za kompozite z visoko vrednostjo imaginarne komponente elektromagnetnih lastnosti. Cilj naše raziskave je bil eksperimentalno analizirati tri uveljavljene modele, Maxwell-Garnett-ov analitični model, analitični model efektivnega medija (effective-medium theory) model, ter fenomenološki Lichtenecker-jev logaritmični model, ter njihovo sposobnost za analizo dveh specifičnih primerov: i) izračun efektivnih lastnosti kompozita s poljubnim deležem volumskega deleža ene in druge komponenete iz izmerjenih efektivnih lastnosti za specifični kompozit z znanim volumskim deležem komponent, ter ii) izračun vrednosti intrizičnih lastnosti komponent iz izmerjenih efektivnih vrednosti za dani kompozit. Osredotočili smo se na nemagnetne kompozitne materiale z feromagnetnimi vključki v mikrovalovnem območju, kjer je pristona visoka imaginarna komponenta permeabilnosti. Naši rezulati kažejo na velike razlike med naštetimi modeli pri izračunih intrizičnih lastnosti z največjimi napakami Maxwell-Garnett modela. Po drugi strani je napovedna sposobnost modelov za izračune efektivnih lastnosti za poljuben kompozit iz izmerjenih vrednosti za dani kompozit v splošnem mnogo boljša.

1 INTRODUCTION

Analytical models for prediction of composite properties are long established and well tested in diverse fields like electromagnetics, mechanics and thermodynamics [1-7]. Such models could a be very useful tool for developing and characterization of composite materials and are thus used commonly in materials research. Over the years a number of studies has been made that compare experimental results with the results of the analytical models for the composite systems where the composite inclusions have significant real and imaginary parts of the intrinsic permeability (e.g. [8] and references therein [9-16]). Such composites are becoming increasingly important as development of electronics demands new materials for operation in the microwave frequencies, where diverse relaxation and resonance effects can lead to significant imaginary components of the electromagnetic properties.

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Most of the existing studies focus on a comparison of the measured effective properties with results of the analytical models for the effective properties, where the input parameters in the models are either known or fitted. But, several studies also analyz the intrinsic properties of the magnetic composite components. These studies can be divided in two groups. In the first group, the analysis of the intrinsic parameters is limited mostly to the imaginary part of the permeability and a full analysis was is not performed. In the second group the analysis is significantly more complex with application of extended analytical models where fitting of the microstructure parameters was combined with the Landau-Lifshitz equation [e.g. 8, 13] from which the relevant intrinsic parameters are determined. These complex extended models require more detailed knowledge of the microstructure, which in some cases is not possible. On the other hand, simple analytical models offer a possibility of an easy and fast calculation of the intrinsic properties and are thus of much interest for researchers in the field of materials sciences and similar fields. For this reason, we focused on the use of simple analytical models (EMT, MG, Lichtenecker logaritmic) [2,8,17,18] and investigated experimentally the ability of these models to calculate the intrinsic properties of the components.

Further, practically no study so-far examined the ability of such analytical models to predict the effective properties of an arbitrary composite from the measured effective properties of a known composite. Such calculation is of a great practical value as commonly only a specific composite sample can be characterized and no intrinsic properties are known. Therefore, we included in our study also the analysis of the predicting strength of the above mentioned models for determining the effective properties of an arbitrary composite.

In our study we focused on the magnetic permeability of the composite materials. Our experiments were done in a microwave frequency range on a set of isotropic, nonmagnetized composites with the ferromagnetic inclusions, which are frequently used in the microwave applications. Such composites, with the magnetic inclusions and nonmagnetic matrix allowed a study on detached systems with only one component (inclusions) contributing to the effective properties. Furthermore, such composites exhibit a significant imaginary part of the permeability due to a strong permeability relaxation and/or resonance in the microwave frequency range.

We examined the analytical models with respect to the experimental results in two directions. First, the ability of the analytical models to calculate the complex intrinsic permeability of the inclusions from a measured effective permeability of the composite. Second, the predicting strength of the models to calculate the effective permeability of an arbitrary composite from the measured (effective) properties of a known composite. Further, we compared the calculated intrinsic permeability from the models with measurements on a bulk filler material and analyzed quantitatively the agreement of both results, which enabled us to test the validity of the analytical models.

2 THEORY

As shown before, for the composites with the ferromagnetic inclusions the equations of the analytical models differ with respect to the composite state, i.e. whether the composite is magnetized, demagnetized or has single domain inclusions [19]. However, since all the analyzed composites have demagnetized ferromagnetic inclusions and are multi-domain, we applied in our analysis two analytical models that are most widely used in literature, Maxwell-Garnett model and effective-medium-theory model [1,2,7,8,18], and a phenomenological model from Lichtenecker (logarithmic mixing law) [1,18,20] with no additional modifications.

For the special case of a twocomponent composite with spherical inclusions in a continuous matrix the models give the following equations, which relate the intrinsic permeability of inclusions (μ_i) and matrix (μ_m) with the effective permeability of a composite (μ_{eff}):
The Maxwell-Garnett (MG) model [1,2,7,8]

$$\frac{\mu_{eff}-\mu_m}{\mu_{eff}+2\mu_m} = F\cdot\frac{(\mu_i-\mu_m)}{\mu_i+2\mu_m}\,, \tag{1}$$

the effective-medium-theory (EMT) model [1,3,9,17]:

$$F_i\,\frac{\mu_i-\mu_{ef}}{\mu_i+2\mu_{ef}}+F_m\,\frac{\mu_m-\mu_{ef}}{\mu_m+2\mu_{ef}}=0\,, \tag{2}$$

and Lichtenecker model (logaritmic model) [1,2,18]:

$$\begin{gathered}\mu_{eff}=\mu_i^{Fi}\cdot\mu_m^{Fm}\,,\\ \log(\mu_{eff})=F_i\log(\mu_i)+F_m\log(\mu_m)\,.\end{gathered} \tag{3}$$

Here it is important to stress that both, the MG and EMT analytical models are only approximations and start with a number of assumptions [1, 2, 21, 22], the most notable being the assumption of a homogenous distribution of the inclusions and neglect the near-field effects. Further, all the models assume no distribution of the intrinsic properties or shape for inclusions in the composite. These models can be extended for the more general case of the ellipsoid inclusions where the shape factors would come into equations [7, 14, 21], however, this requires a more detailed knowledge of the composite (shape and orientation distribution), which is rarely possible.

2.1 Determining the intrinsic properties

To determine the intrinsic properties of composite inclusions μ_i, we used equations (1)-(3) of the three chosen analytical models. As the input parameters, the effective properties of a composite μ_{eff}, measured matrix permeability μ_m, and the volume fraction of the inclusions were used. From this, the intrinsic permeability μ_i was calculated for different sets of the measured composites.

2.2 Predicting streght of the models - calculating the effective properties

To analyze the ability of the models to predict the correct values of the effective properties for different volume fractions of the inclusions, we applied two approaches. In the first approach we selected a special case where the intrinsic properties of the composite components were determined with a reasonable accuracy. Then we inverted the calculations and inserted the measured intrinsic permeability of inclusions μ_i and matrix μ_m in the models to calculate the effective permeability of the composites μ_{eff} for different volume fractions. These calculated μ_{eff} were then compared to the experimental results of the effective permeability for the relevant composites.

In the second approach, we first selected and measured a reference composite with a known volume fraction of the inclusions. We inserted the effective properties of this reference composite (at a given volume fraction) into the analytical models to calculate the intrinsic permeability and inserted this calculated intermediate result again into the analytical models to finally calculate the effective permeability for different volume fractions of inclusions. These calculated effective permeabilities were compared to the experimentally determined effective permeabilities of the corresponding composites and by this the predictive strength of the models was tested.

3 MATERIALS AND METHODS

We prepared a series of measurements on the actual composites with the ferromagnetic inclusions. In the experiments we used two types of composites: i) granular composites, where grains of the NiZn ferrite are inserted in a continuous nonmagnetic matrix (wax), and ii) ceramic composites, where grains of two different soft-magnetic materials (large NiZn ferrite grains and M hexaferrite micrograins) are sintered together.

For the granular composite we used a milled NiZn ferrite powder as the inclusions in a nonmagnetic (μ_m = 1) matrix made from wax. The composite samples were prepared by manually mixing the ferrite powder (average diameter 7 μm) with a micronized wax powder (Micro-powders Inc.) (average diameter 5 μm) as a matrix, and pressing the mixture in an appropriate forming tool. Pressing the eliminated microstructure of wax gave continuous matrix. We prepared a set of samples with the inclusions volume fractions from 2 vol% to 48 vol%. Volume fraction of the inclusions was determined from a known mass ratio and density of ferrite and wax and was checked by measuring the effective density of the samples. We estimate that the absolute uncertainty of the determined volume fraction of inclusions is about 2 vol% and thus affects the samples with the low volume fractions most. The homogeneity of the inclusions distribution was obtained by thorough mixing. The ferrite grains show a characteristic polyhedron shape of the milled materials and are thus not spherical (as assumed in the analytical models), but on the other hand they do not deviate excessively as it can be seen from Fig. 1.

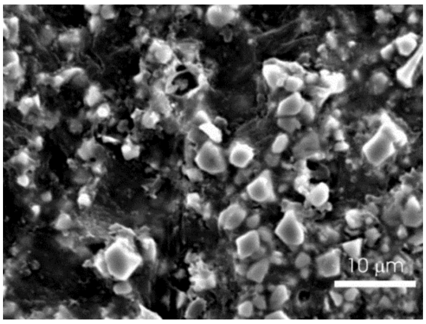

Figure 1: Scanning electron microscope image of a granular composite with the NiZn ferrite inclusions in the wax matrix. Volume fraction of the inclusions is 0.37.

The second type of the composite was a ceramic composite with the NiZn ferrite inclusions, having 50-100 μm in diameter, in a soft magnetic matrix of M hexaferrite (more details on the materials are given in [23]). We prepared samples with three NiZn ferrite volume fractions (10 vol%, 21.5 vol% and 30.5 vol%). To experimentally determine the intrinsic permeabilities of the NiZn inclusions and of the hexaferrite matrix, we also prepared bulk ceramic samples of pure NiZn ferrite and pure M hexaferrite material. Here, the volume fraction in the composites was additionally determined from the X-ray diffraction analysis [23]. Again, the uncertainty is of the order of below 2 vol%. The shape and homogeneity of the distribution of the inclusions is better in this case [23] and the spherical approximation is quite valid.

3.1 Measurements

The permeability and permittivity of a given material were obtained by measuring the S-parameters in the coaxial transmission line with an Anritsu 37369C Vector Network Analyzer for the frequencies from 500 MHz to 10 GHz. This was validated for the frequencies up to 1 GHz with measurements using impedance analyzer HP 4291A [24]. An APC-7 coaxial line was used as the sample holder and the samples had a toroidal form with the inner dimension of 3.05 mm and outer dimension 7 mm in order to fit well into coaxial sample holder. For eachgap between the sample and the sample holder a correction model was applied [24]. The measurements were made both in a short-circuited as well as in a transmission/reflection geometry and the permeability and the permittivity of the samples were calculated from the measured scattering parameters or measured impedance [24]. With such measurement set-up there are no sample demagnetization factors and the obtained permeability is the actual effective permeability of the composite material. Therefore, we used obtained permeability values for the calculations with the analytical models [20]. The measurement uncertainty was minimized by preparing and measuring two identical samples for each nominal composition, and by two consecutive measurements for each sample. We estimated the absolute uncertainty of the measured permeability to be below 2% over the measured frequency range.

3.2 Material properties

For the granular composites with a nonmagnetic (wax) the matrix measured permeabilities as a function of the frequency for different volume fractions are shown in Fig. 2.

These measurements are only a representative subset of a complete set of the composites with 14 different volume fractions of the NiZn ferrite inclusions. Only the results for this subset are presented in this work, however, a general behavior is the same for the whole set of the composites and thus the observations and conclusions are the same. The measurements show the familiar resonance character of the composite ferromagnetic materials [25-27], with the real part of the effective permeability declining with the frequency and eventually going even below 1, and also with a significant imaginary part of the effective permeability.

For the ceramic type composites, the measured permeabilities for all three composite samples, bulk matrix (M hexaferrite), and bulk inclusions (NiZn ferrite) are shown in ref. [23]. As the ferromagnetic resonance frequency of the M hexaferrite is in the millimeter wave region [23,25,26], the real part of the permeability is close to 1 and the imaginary part is relatively low. In analytical models we used the measured frequency spectrum of the bulk M hexaferrite permeability as the intrinsic value of the matrix permeability. In contrast, the magnetic permeability of the bulk NiZn ferrite shows the familiar ferromagnetic resonance character [25, 26] and is much larger than that of the matrix, so it represents the main contribution to the effective permeability of the ceramic composite.

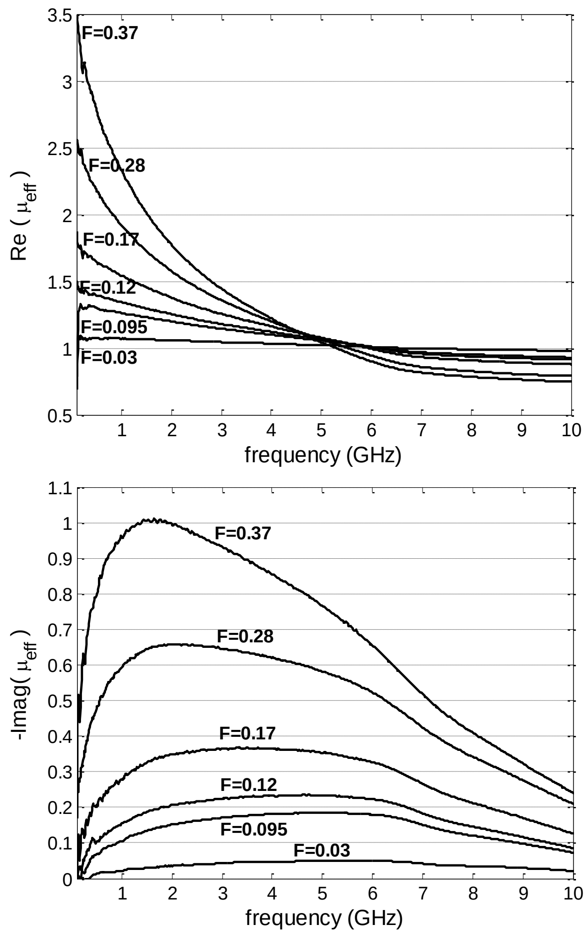

Figure 2: Frequency spectrum of the real and imaginary part of the effective permeability of several composites for different volume fractions of the NiZn ferrite inclusions in a wax matrix.

However, the measured permeability of the bulk NiZn ferrite cannot be simply used as the intrinsic permeability for all composites. Although the composition of the NiZn ferrite is the same in both types ofthe composites (with wax or ceramic matrix), there is a significant difference in the diameters. The large inclusions in the ceramic composite are much larger than the typical domain sizes in ferrite [23,28] and should basically exhibit similar properties as the bulk material. Therefore, we took bulk permeability of the NiZn ferrite as a reasonable approximation for the intrinsic permeability of the ceramic composite inclusions, while for the composites with the wax matrix, this value can be used only as an order of magnitude approximation.

This is true since for the granular type of composites (wax matrix) the average diameter of the inclusions is below 10 μm and is obtained by an additional milling of the starting NiZn ferrite material. Such particles can have significantly changed intrinsic properties [25,27] both due to the milling and due to the dimensions close to the typical single domain limit [28], and cannot be accurately presented by the bulk properties. Therefore, the bulk NiZn ferrite permeability can serve only as an order-of-magnitude guideline for the intrinsic permeability of inclusions. Due to this, we analyzed the calculated intrinsic permeability of the NiZn ferrite in a wax matrix only by comparing values obtained from different analytical models and from the composites with different volume fractions of the NiZn ferrite inclusions.

4 Results and Discussion

4.1 Calculating the intrinsic permeability of the inclusions – applicability of different analytical models

By using equations (1-3) we calculated the intrinsic permeability of inclusions μ_i from measured effective permeability μ_{eff} of the composites with wax matrix for various volume fractions of inclusions.

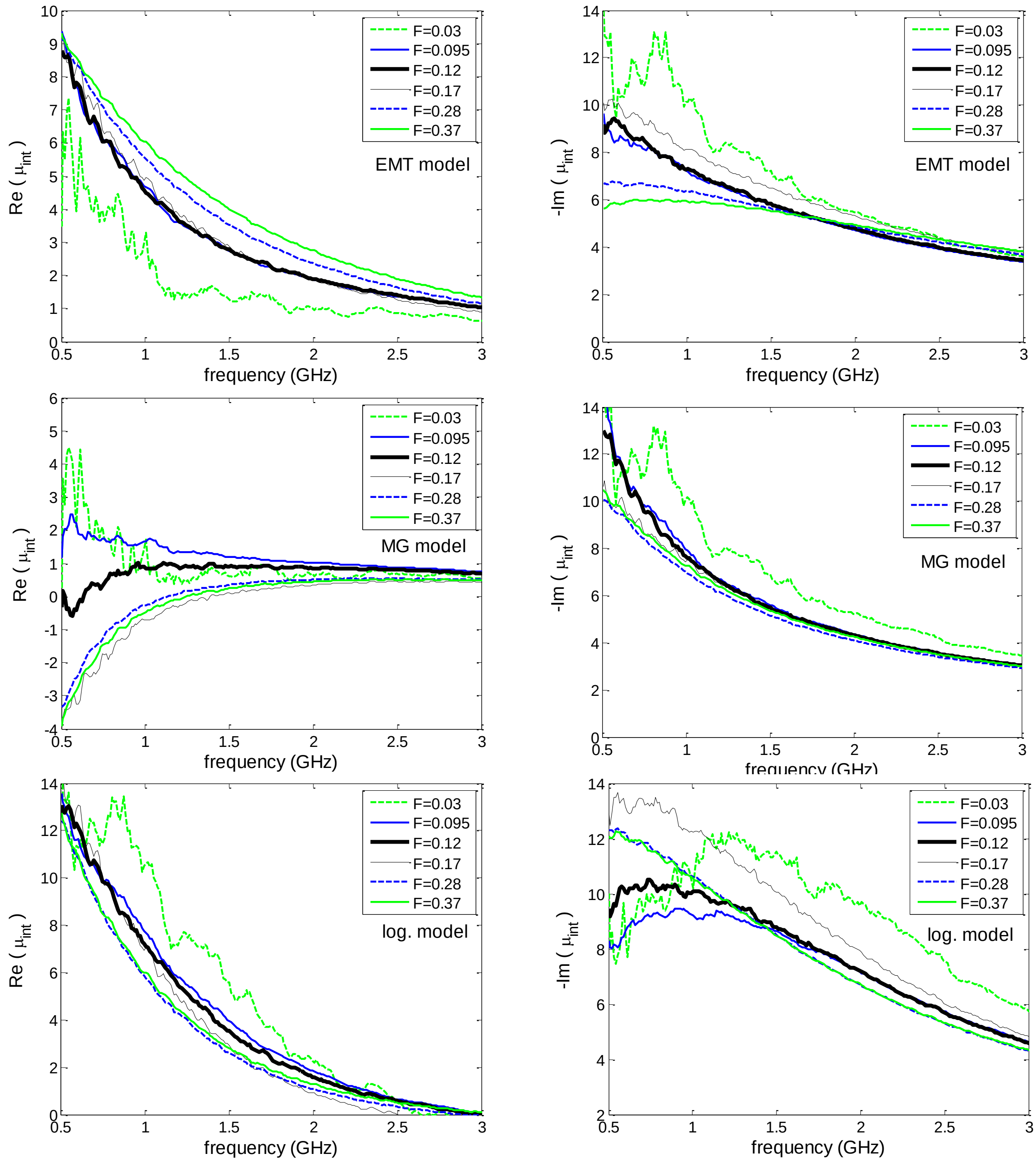

Figure 3: Real (left) and imaginary (right) intrinsic permeabilities of the NiZn ferrite inclusions, calculated with the effective-medium-theory model (EMT), Maxwell-Garnett model (MG) and logarithmic (log.) model. The measured effective permeabilities (NiZn inclusions in a wax matrix) used for calculations are shown in Fig. 2.

Fig. 3 shows the calculated curves of the inclusions intrinsic permeability for the composites with different volume fractions as calculated by all the three analytical models (Eqs. 1-3). As the inclusions in all the composites are of the same type, one expects similar calculated intrinsic properties for all volume fractions.

When analyzing the imaginary part of the intrinsic permeability (μ''_i), the MG models gives much smaller difference between the curves for different volume fractions than the EMT models, where a gradual transition in the curve shape occurs for the volume fractions above 20%. This could be explained by the increase in the number of the agglomerates as the volume fraction nears the percolation point since the agglomerated particles can have different properties to those of the isolated particles [27, 29-31]. The results of the Lichtenecker model also differ significantly with the changing volume fraction. High variation between curves for the lowest volume fraction for the all three models can be attributed to the experimental error since both, the volume fraction uncertainty and the measured signal to noise ratio are largest for such composite. Also, the intrinsic permeability is not much larger than the inverse of the demagnetization factor (μ_{int} >>3), where the calculation of intrinsic permeability from composite measurements is problematic [8]. However, there is a much more significant difference in the real part of permeability μ'_i, where the EMT and the log model give quite disperse values but all positive, whereas the MG model gives deeply negative values for several composites. Although the negative values of the real part of the permeability can occur due to the resonance, such low values usually occur only for single crystal samples and are limited to the frequency range above the ferromagnetic resonance. For our type of the NiZn ferrite the bulk permeability does not fall below zero and falls below 1 only at about 2 GHz (Fig. 4 and [23]). Such curves with deeply negative values are unphysical for the used materials and thus the MG model gives clearly incorrect results. One possible explanation for such results could be the uncertainty of the volume fraction and of the measured permeability. This is certainly true at very low frequencies (below 250 MHz) as the measurement error is very large, however, as we varied the data to analyze this effect at higher frequencies the required changes in permeability and/or the volume fraction to obtain positive real curves were way beyond the estimated uncertainties.

The exceptions to the above observations are the curves for the low volume fractions. For all the models, the curve with smallest volume fraction differs notably from other curves, but this can be expected as the measured uncertainties have the proportionally largest effect. Further, such curves for both the MG and EMT analytical models do not exhibit the dip in the negative region and are qualitatively similar. This we would expect since in the limit of a very dilute system, both analytical models should give the same results. This indicates that the observed unusual character of the MG model is related to higher volume fractions of inclusions. This agrees with the fact that MG model is an approximation where the multipole interactions are neglected and is thus valid only for the "dilute" composite systems.

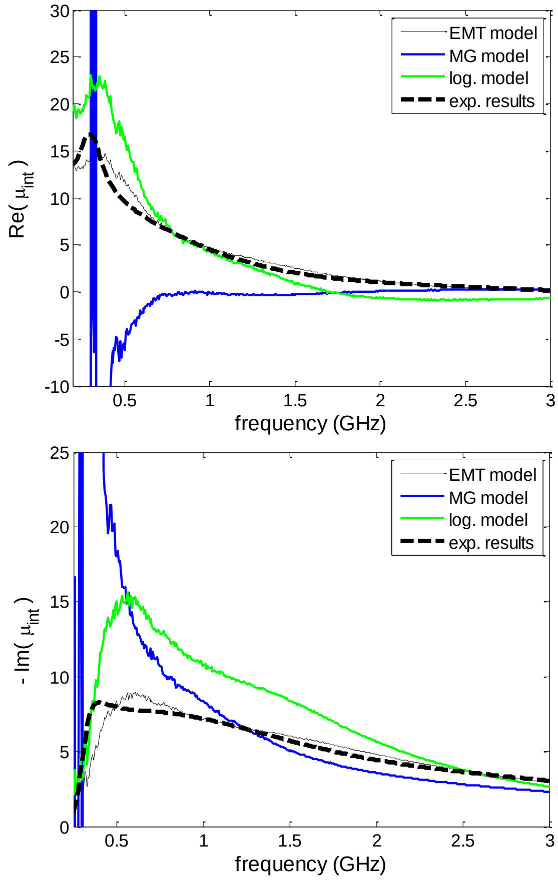

Figure 4: Comparison of the calculated intrinsic permeability of the inclusions in the ceramic composite (NiZn inclusions in the M-hexaferrite) with the experimentally determined permeability of the bulk inclusion material – pure NiZn. For the calculations we used the effective permeability of the ceramic composite with F = 0.3.

Furthermore, in Fig. 4 we analyze the second set of our measurements of the effective permeability for the ceramic composites with the NiZn inclusions in M-hexaferrite (results are given for F = 0.3). In contrast to the wax matrix composite, in the case of the ceramic composite the measured bulk permeability of the NiZn ferrite is a good approximation for the intrinsic properties of the inclusions and thus offers a possibility of a direct comparison of the actual and calculated values. With the EMT model we obtained similar results for the intrinsic permeabilities of the ceramic composites as in the case of the wax matrix, with higher values of both parts of the permeability for the lower volume fractions of inclusions (results not shown). Again, this behavior could be explained by the effect of percolation, as the domain structure and thus the permeability could be different for the isolated particles than for larger agglomerates [27, 29-31]. Obviously, there is again also an effect of the uncertainty in determining the volume fraction of the inclusions, which has a much larger impact for the lower fractions. But in general, for the ceramic composite, the EMT model calculations of the intrinsic permeability show good qualitative agreement with the experimental values for the bulk NiZn ferrite, especially at larger volume fractions (shown in Fig. 4). There is a notable difference at the frequencies below 500 MHz, however, this we can attribute to the changed domain configuration in the composite material vs. the bulk ceramic material.

The values obtained with the MG model (see Fig. 4 for *F*= 0.3) deviate considerably from the values obtained by other two models and experimental results. By comparing the curves in Fig. 4 for the both bulk ceramic and the MG calculated permeabilities, it becomes obvious that the MG model predictions for the intrinsic real permeability are very far from the bulk permeability and even ithe maginary part of the intrinsic permeability shows large overestimation. The Lichtenecker logarithmic model again gives higher values than the EMT model for both parts of the permeability, although qualitatively the frequency dependence of the permeability is similar to both EMT model results and the experimentally determined bulk (intrinsic) permeability of the NiZn ferrite inclusions. Similar results were obtained also the for ceramic composites with volume fractions $F = 0.1$ and $F = 0.2$ (results not shown).

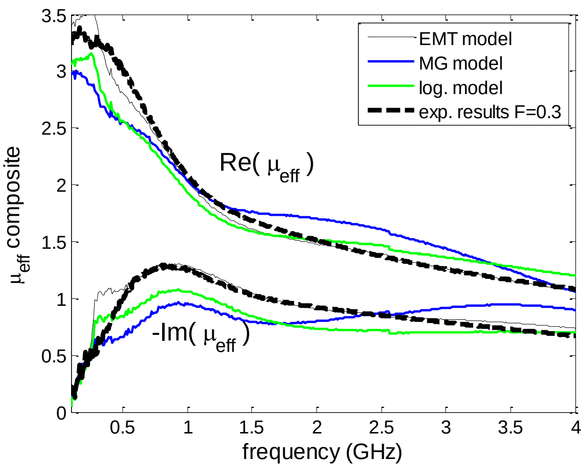

Figure 5: Measured effective permeability of a ceramic composite ($F = 0.3$) volume fraction of the NiZn ferrite is compared to the effective permeability calculated (Eqs. 1-3) from the intrinsic permeability (bulk permeabilities of the M hexaferrite and NiZn ferrite as the input parameters.

The difference between the models is evident also when calculating the effective permeability of the composite from the known intrinsic properties. Here we used as the input parameters in the models (intrinsic permeability of the matrix and the inclusions) the measured bulk properties of the barium hexaferrite and of the NiZn ferrite, as given in [23].

In Fig. 5, a very good agreement between the measurements on the real composite and the EMT results is evident, whereas both the MG and the Lichtenecker model diverge noticeably from the measurements. Results are similar also for smaller volume fractions, but with a somewhat decreased agreement for the EMT model at $F = 0.1$, which as before we attribute to the uncertainty in the determined volume fraction of the inclusions and possible effect of the microstructure.

Altogether, our results show that for determination of the intrinsic properties from the measured effective properties of a composite only the EMT analytical model gives reasonable results. On the other hand, the logarithmic model tends to give larger values and the MG model gives odd values that are very different to usual values for the soft-magnetic materials. This is observed both for the wax matrix composites and ceramic composites where a direct comparison with the measured permeability of the bulk materials of the inclusions is possible.

However, even though the EMT model gives the best overall agreement with the experimental values, a significant difference in calculated μ_i is observed between the composites with a lower and higher volume fraction of the inclusions.

Our previous numerical studies on the effects of agglomeration and percolation on the effective permeability of a composite [30, 31] showed a similar effect. Analytically calculated μ_i deviated from th enumerical models which can be explained with the fact that the agglomeration/percolation and associated near-field effects are not taken into account in the analytical models (Eqs. 1-3), while the numerical calculations were performed on realistic microstructures.

The standard analytical models like the effective-medium theory model and the Maxwell-Garnett model, assume a homogenous microstructure in the composite [1,7,32] and can thus include only a simple microstructure information such as the volume fraction and inclusions’ shape. On the other hand, there are several extended analytical models that aim at incorporating also other parameters of the microstructure and are frequently used in combination with expressions for the intrinsic permeability like the Landau-Lifshitz equation for the ferromagnetic resonance [8,13].

However, since microstructure of a real composite is in general quite complicated, it is difficult to determine experimentally the relevant parameters [32,33]. Therefore, the extended analytical models that aim to incorporate a more accurate microstructure [8,13, 34, 35] generally rely on fitting the experimental data to the model to obtain the necessary parameters.

4.2 *Effective permeability of a composite – predictive strength of the analytical models*

Another application of the analytical models is to calculate the effective properties of the composites for different volume fractions from the measured effective properties of a given composite (input composite). To examine the ability of the analytical models to correctly predict the effective properties for different volume fraction we selected a given composite for which we measured the effective permeability at some input volume fraction F_{ini}. We first calculated with a selected analytical model the intrinsic permeability of the inclusions, and then used this calculated intrinsic permeability together with an arbitrary volume fraction as input parameters in the same analytical model to calculate the effective permeabilities of the composites for arbitrary volume fractions.

The results for the wax matrix composites are shown in Fig. 6 where the composites with inclusions volume fractions F_{ini} = 37 vol% and 9.5 vol% were used as the initial input. Surprisingly, the results of the MG and log models show very good qualitative agreement with the experimental results for the effective permeability in the whole range of the volume fractions. There are deviations from the experimental results of about max. 10% that increase with a larger difference between the input and the calculated volume fractions of inclusions. The deviations can be to some extent attributed to the uncertainty of determining the actual volume fractions in the composites. On the other hand, the results of the EMT model show a notable deviation from the experimental results and only at the volume fractions close to the input composite become similar to the experimental results. Similar observations are obtained for other input composites with different volume fractions of the inclusions. A similar qualitative difference between the experimental curves and the curves calculated with the EMT model was already observed [36], nevertheless, further analysis of the results was not done.

Results of a similar analysis for the ceramic composites are presented in Fig.7.

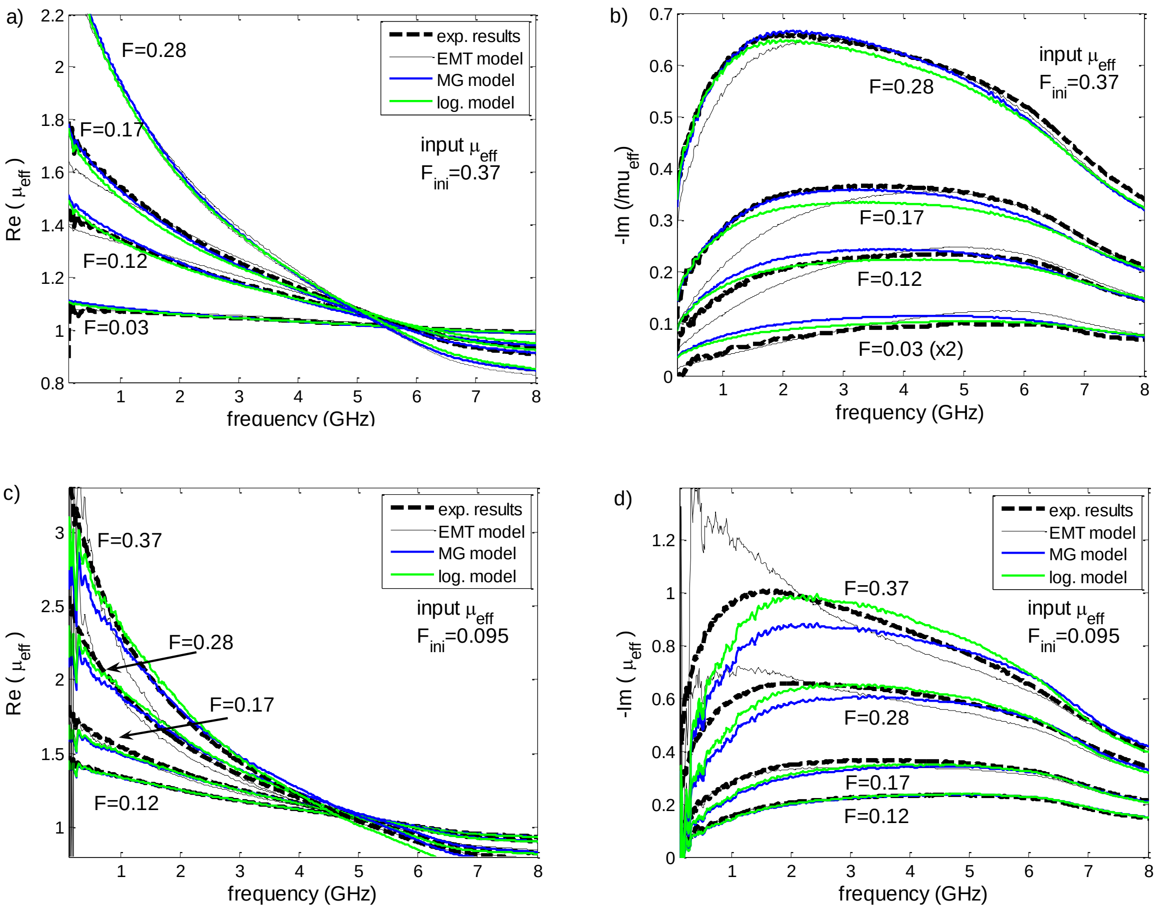

Figure 6: Comparison of the measured effective permeabilities of the wax matrix composites (dashed curves) and calculations of the effective permeabilities with the analytical models (solid curves) where the input parameters are taken from the measured reference (input) composite for F_{ini} = 0.37 (a) and (b), and for F_{ini} = 0.095 (c) and (d).

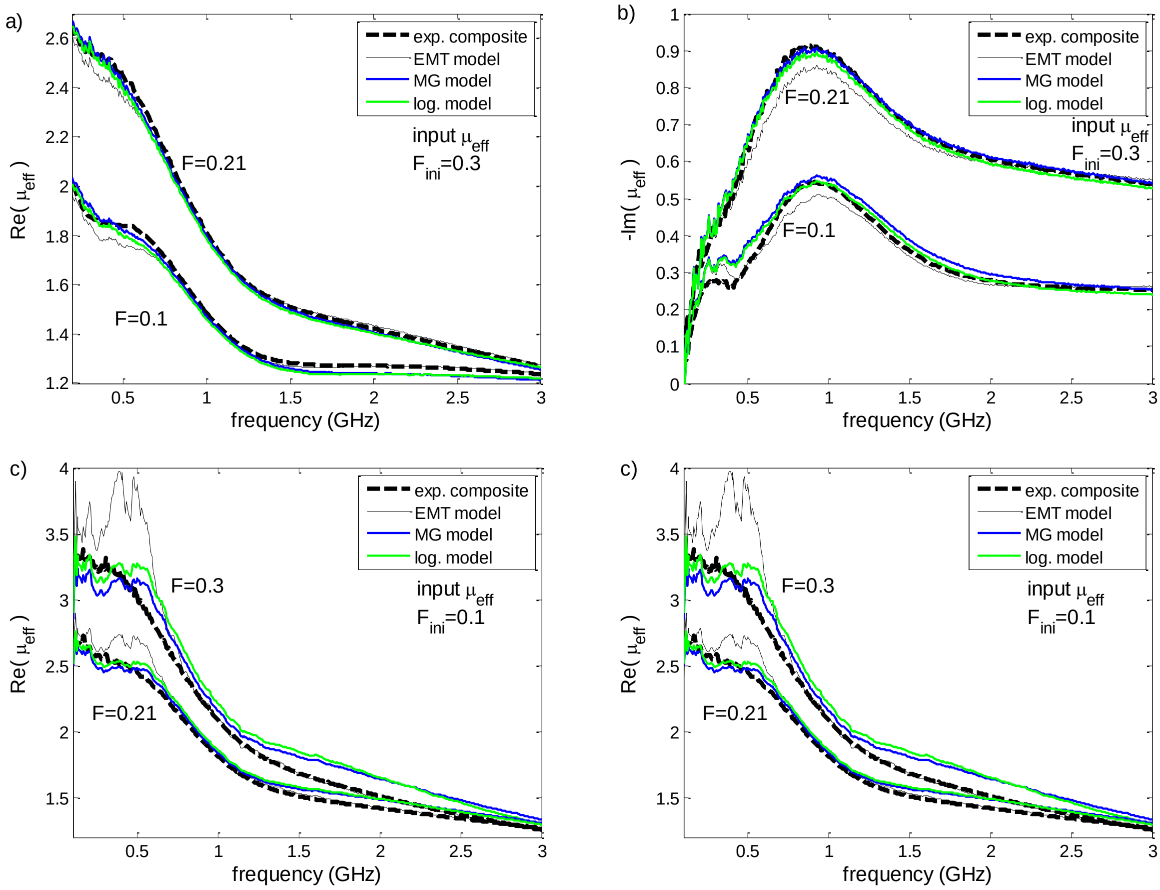

Figure 7: Comparison of the measured effective permeabilities of the ceramic composites (dashed curves) and calculations of the effective composite permeability with the analytical models (solid curves) wherethe input parameters are from the measured reference (input) composite for F_{ini}=0.3 (a) and (b), and for F_{ini} =0.1 (c) and (d).

The observations obtained for the ceramic composites are similar to the observations for the wax matrix composites. The MG and Lichtenecker models give a good qualitative agreement with the experimental results, thus demonstrating a good predictive quality. The calculations of the EMT model give a reasonable agreement with the experimental results for the input composite with a large volume fraction (F_{ini} = 0.3 and F_{ini} = 0.21, for the latter results are not shown), but are quite off the mark for the input composite with a smaller volume fraction (F_{ini} = 0.1).

5 CONCLUSIONS

Simple analytical models are very convenient for an easy and fast calculation of the intrinsic properties of a composite material. However, there is a lack of a more detailed analysis of the validity of these models either for their ability for accurate calculation of the intrinsic parameters or for their predictive strength. For this reason, our study focused on three most widely used analytical models (EMT, MG, Lichtenecker logaritmic) [2,8,18]. We used experimental measurements on the composites with the ferromagnetic inclusions to test the ability of these models to obtain the intrinsic permeability of inclusions and to predict the effective permeability of a composite with an arbitrary volume fraction of the inclusions.

To determine the intrinsic permeability of the inclusions from the measured effective properties of a composite, our results suggest that none of the analysed models gives good quantitative results. But while the EMT model, and to some extent also the Lichtenecker model, show a qualitative agreement with the experimental results for the intrinsic permeability, the MG model gives clearly erroneous results. However, this is evident only from the real part of the permeability and analyzing only the imaginary part could mask the problem with the calculated result.

Nevertheless, the analytical models can be more successfully applied to predict the effective properties of a composite with an arbitrary composition. As the actual intrinsic properties of the inclusions and matrix are usually not known, one can use a measured reference composite for the input parameters in the analytical models. In contrast to the previous case, the Maxwell-Garnett and Lichtenecker models gave good predictions of the effective properties over the whole range of the volume fractions that we examined. On the other hand, the effective-medium model gave only a poor agreement that worsened as the difference between the volume fraction of the reference and the target composite increased.

Altogether it seems that none of the analyzed simple models is able to completely describe the realistic composites with a complex microstructure. Our previous numerical studies [30,31] as well as other theoretical and experimental studies [6,8,13] showed importance of taking into account also the microstructure (agglomeration, percolation), therefore a large part of a relatively poor ability to obtain intrinsic parameters can be attributed to this. However, the predictive ability for calculation of the effective properties of especially the MG model is quite good. This is in contrast with expectations that the MG model should be accurate only at low volume fractions and suggests that for some applications the MG model is phenomenologically applicable, especially for predicting the effective properties from a known material.

Our results show that a great care has to be taken in trying to extract intrinsic properties from the composite measurements and the results obtained in such manner can have a large range of uncertainty. Predicting the effective properties based on known intrinsic properties or reference composite is much more robust and can give good results. Even though the analytical models can be valuable tools in analyzing the properties of the composite materials, one has to be aware of the limitations and choose an appropriate analytical model for the specific task.

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