Numerical testing of homogenization formulas efficiency for magnetic composite materials

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Abstract. Magnetic composite materials are used for multiple applications. The macroscopic behavior of the material is influenced by its microscopic properties. Sometimes it is difficult to model the complex structure of the composite because the properties differ at the microscopic scale. Some simplifying assumptions are often needed, one of them being the homogenization of the material. In this paper two different composite materials were analyzed using a finite element software (COMSOL Multiphysics©). Both materials have a non-magnetic matrix with inclusions made of paramagnetic material for one of them, and ferromagnetic material for the other. The 3D numerical simulations were made for different particle concentrations and for different applied fields. The homogenization was implemented using two different formulas: Maxwell Garnett and Bruggeman. Numerical comparison was made between the magnetic properties of non-homogeneous materials and homogeneous ones showing each formula’s efficiency for different cases.

1. Introduction

Starting from the middle of the 20th century the composite materials experienced an enormous development in multiple areas of the industry. Magnetic composite materials can be used in various applications, such as electromagnetic shielding, electrical machines, biomedical applications (for example, magnetic resonance imaging contrast enhancement or magnetic drug delivery), magnetic sensors [1] ÷ [5] and many other branches of the industry.

In the past years more and more scientists and research companies started developing new composite materials capable of improving the properties of classical materials. Magnetic composite materials can be described as a combination of minimum two materials: the matrix and the inclusions. The large number of combinations between the matrix and inclusions, imposes an important study of the structure and properties of the created composite. The continuous research and development of the composite materials leads to complex structures that cannot be analyzed easily. The solution in these cases is to make some simplifying assumptions. For example, all the material inclusions are considered being exactly the same from the geometrically point of view and perfectly embedded in the matrix. The other assumption, that was used successfully, is the homogenization technique. The idea behind this technique is to replace the complex structure of the real material with a homogeneous one that is able to describe correctly all the material properties (e.g. material conductivity, permeability, permittivity). The real material properties differ at the microscopic scale and it is important to know all these properties in order to describe correctly the material. By using the homogenization technique the medium becomes homogeneous, the properties are the same in each point and the macroscopic rules
and formulas can be applied with success. One of the immediate results of using the homogenization is that the time needed to obtain the desire solutions is reduced.

Multiple homogenization techniques exist in technical literature. Some of them are based on analytical formulas (such as Maxwell Garnett, Bruggeman, Clausius-Mossotti), others based on different computational techniques (e.g. finite element method or finite difference method) [6] ÷ [14].

The object of this paper is to analyze, from the numerical point of view, the efficiency of Maxwell Garnett and Bruggeman homogeneisation formulas, implemented in a three dimensional software, for two different materials.

Maxwell Garnett homogenization rule [15] is one of the first techniques used to describe the permeability of composite materials. It can be successfully used in describing materials with spherical and ellipsoidal inclusions but it is not limited to these applications alone. The initial formula was developed for describing the material permittivity, but can be used with good results for describing the permeability of the composite material. The Maxwell Garnett homogenization formula is [16]:

\[
\frac{\mu_i - \mu_o}{\mu_i + 2\mu_o} = f \cdot \frac{\mu_i - \mu_o}{\mu + 2\mu_o}
\]

where indexes \(o, m\) and \(i\) indicate the homogeneous, matrix and inclusion magnetic permeability, and \(f\) is the volume fraction of the inclusions in the matrix.

Bruggeman extended the previous model by taking into consideration the interaction between particles. The Bruggeman homogenization formula is [15]:

\[
(1 - f) \cdot \frac{\mu_i - \mu_o}{\mu_i + 2\mu_o} + f \cdot \frac{\mu_i - \mu_o}{\mu + 2\mu_o} = 0
\]

where indexes \(o, m\) and \(i\) indicate the homogeneous, matrix and inclusion magnetic permeability, and \(f\) is the ratio between the inclusion and the matrix volume.

By performing the numerical comparison between the magnetic properties of the real composite materials and homogeneous ones each formula’s efficiency can be pointed for different concentrations (\(f\) values).

2. Investigated Materials
Two different composite materials were considered. The first one, Material A, is a composite material developed for biomedical applications, which has a non-magnetic matrix (cellulose) with inclusions made of paramagnetic material. The second one, Material B, is a soft magnetic composite material, also with a non-magnetic matrix, but with inclusions made of ferromagnetic material.

Figure 1. Magnetic characteristic for the pure powder for material A
The magnetic characteristics of the two materials samples were measured using the Vibrating Sample Magnetometer (VSM) device – the sample been a thin disk with 5 mm diameter. The characteristics for the spherical inclusions (the mean particle radius is approximately 20 nm) were obtained from the VSM results (figure 1) and they were used in the numerical approach in order to create a model which is very close to the real material.

3. Numerical approach
Using the finite element software COMSOL Multiphysics © [16], 3D models were made for both materials for the static case. A cube was chosen to model the composite material (figure 2). The inclusions were considered spherical in shape (inclusion radius = 20 nm) and were distributed randomly.

Figure 2. The 3D model used for the real material

Five different particle concentrations were considered: 50%, 40%, 30%, 20% and 9%. For each concentration, the number of inclusions was calculated – respectively 27, 22, 17, 11 and 5. Using the boundary conditions, a uniform magnetic field was applied in the x direction (figure 3). A refined mesh was chosen with approximately 100,000 finite elements for the composite material with the highest number of inclusions.

Figure 3. Magnetic flux density arrow plot

The matrix magnetic permeability is $\mu_m=1$ for both materials. The permeability of the inclusions was chosen from the $B=f(H)$ curve of the real material. Thus, for material A, five relative permeability values were chosen, each one corresponding to one different point from the $B$-$H$ characteristic: $\mu_{r_{P1}} = 4.22$, $\mu_{r_{P2}} = 3.34$, $\mu_{r_{P3}} = 2.24$, $\mu_{r_{P4}} = 1.44$, $\mu_{r_{P5}} = 1.05$. For material B, three magnetic permeability
values were considered: $\mu_r_{P1} = 6225.13$, $\mu_r_{P2} = 589.65$, $\mu_r_{P3} = 99.76$. Each permeability value imposes different values for the boundary conditions, which means different values for the applied magnetic field.

Numerical simulations were made for different particle concentrations: 9%, 30% and 50% for material A and 50%, 40%, 30%, 20% and 9% for material B. For each concentration three simulations were performed: the composite material and two homogeneous materials (where the material has the same relative permeability, in one case computed with Maxwell Garnett formula, and in the other case using Bruggeman). The results obtained were compared and different conclusions were made.

4. Results and discussions

The first step in the analysis is to compute the homogeneous permeability. Material A was the first analyzed material (figure 4). Because the values of the inclusions relative permeability are not very high and the difference between the relative permeability of the matrix and inclusions is also small, it is expected that the two formulas are efficient in the analysis.

![Figure 4. Magnetic field line plot for a model with 50% particle concentration](image)

The magnetization was computed for all three cases (particle concentration is 9%, 30%, respectively 50%) and for five different values of the applied magnetic field ($H_1 = 23.86$ kA/m, $H_2 = 47.81$ kA/m, $H_3 = 71.66$ kA/m, $H_4 = 119.39$ kA/m, $H_5 = 453.65$ kA/m – see figure 1). Figure 5 presents the variation of the magnetization for three different filler factors $f = 0.09, f = 0.3$ and $f = 0.5$, for $\mu_r_{P3} = 2.24$ and $B=0.3$T. It is easy to observe that the magnetization values are approximately the same regardless of the homogenization formula.

![Figure 5. Magnetization variation for $\mu_r_{P3} = 2.24$ and for three different concentrations (9%, 30%, 50%)](image)
In order to point better the difference between the obtained results, the relative error was calculated.

\[
err = \left( \frac{M_{\text{composite}} - M_{\text{homogeneous}}}{M_{\text{composite}}} \right) \cdot 100 \% \quad (3)
\]

Table 1 presents the relative errors between the computed composite material magnetization and the homogenized one. It can be observed that the errors are bigger with the increase of the difference between the two permeability values. Also, the errors obtained using Bruggeman formula are bigger than the ones using Maxwell Garnett.

**Table 1.** Relative error between the magnetization values

|        | \( f = 0.09 \) | \( f = 0.3 \) | \( f = 0.5 \) |
|--------|----------------|----------------|----------------|
| M. G.  | M. G. Brugg.   | M. G. Brugg.   | M. G. Brugg.   |
| P1     | 2.17%          | 6.56%          | 2.73%          |
| P2     | 1.87%          | 4.98%          | 2.31%          |
| P3     | 1.25%          | 2.66%          | 1.79%          |
| P4     | 0.77%          | 1.05%          | 0.67%          |
| P5     | 0.85%          | 0.85%          | 1.29%          |

There are two possible explications for these errors: one is the fact that if the value of \( f \) is relatively high (0.5 in this case) errors appear because the interactions between inclusions are high and the second one is the fact that performed analysis is made at a microscopic scale which brings some complications because the physical laws change at this scale.

However the differences between the values are not very high so, for this kind of composite materials, the two homogeneous formulas can be used in a numerical simulation of the real material behavior, for simple and also complex structures. The homogeneous results can approximate in acceptable limits the real results.

The second material that is analyzed in this paper is material B. Like in the previous case the material magnetization was computed for the composite material and for the homogeneous one. Three points were chosen: \( P1 - H=89.41 \, \text{A/m}, \, \mu_r_{p1} = 6225.13; \, P2 - H=2161.89 \, \text{A/m}, \, \mu_r_{p2} = 589.65; \, P3 - H=15296.81 \, \text{A/m}, \, \mu_r_{p3} = 99.76 \) from the \( B=f(H) \) magnetic characteristic obtained using the VSM device (figure 6).

![Figure 6. Magnetic permeability graph for material B](image)

Like in the previous case an homogeneous field was applied in the \( x \) direction. Figure 7 presents the magnetic flux density plot for point P3 and for \( f = 0.3 \). Even thought the imposed field is homogeneous, one can observe that, the presence of inclusions disturbs the flux density values inside the plot.
Figure 7. Magnetic flux density plot for a model with 30% particle concentration

For each permeability value, five different inclusion concentrations were considered - 50%, 40%, 30%, 20% and 9%. In the next three figures the magnetization variation versus the concentrations for each permeability value in part are presented.

Figure 8. Magnetization variation for $\mu_r P_1$ versus different inclusions concentrations

Figure 9. Magnetization variation for $\mu_r P_2$ versus different inclusions concentrations
It is easy to observe that the Maxwell Garnett formula is more efficient for modeling the composite material. The relative error is less than 5% between the obtained results with Maxwell Garnett and the real material. Also, from the three figures one can easily observe that the Maxwell Garnett formula gives smaller results than Bruggeman. The difference is due to the fact that Maxwell Garnett does not take into consideration the inclusions interaction. In this case, because the configuration is not very complex, the impact of the fact that the local material behavior is not taken into account is not very big. But the impact of the concentration value is significant, the two formulas having a higher error for a bigger $f$ value - the errors increase approximately two times. The errors for Bruggeman formula are ten times higher than the ones for Maxwell Garnett. Another factor that influences the errors is the fact that in this second case the difference between the permeability of the matrix and the inclusions is bigger than in the first case. This fact perturbs the accuracy of the results obtained using Bruggeman formula. In conclusion, for this case the Maxwell Garnett formula is also more efficient in showing the sample magnetic behavior.

5. Conclusion

From the simulations that were carried out one can observe that the results obtained using the Maxwell Garnett homogenization formula are closer to the ones obtained for the composite material. Although both formulas produce good results for many configurations and are successfully used in order to confirm the expected results in some cases, they are not so precise if the inclusion concentrations are too high or the configuration is too complex. It is important for each desired case to take into consideration the problem hypothesis (inclusions concentration, difference between matrix and inclusions magnetic permeability etc.) in order to be able to make some predictions on the formula accuracy for that specific case. In conclusion, for this analysis, the Maxwell Garnett formula was more efficient for the two considered materials than Bruggeman formula.

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