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DESIGN OF PARTICULATE-REINFORCED COMPOSITE MATERIALS

Aleksander Muc, Marek Barski

Institute of Machine Design, Cracow University of Technology, Kraków, Poland

olekmuc@mech.pk.edu.pl

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Abstract. A microstructure-based model is developed to study the effective anisotropic properties (magnetic, dielectric or thermal) of two phase particle-filled composites. The Green's function technique and the effective field method are used to derive theoretically the homogenized (averaged) properties for a representative volume element containing isolated inclusion and an infinite, chain-structured particles. Those results are compared with the FE approximations conducted for the assumed representative volume element. In addition, as a special case, the Maxwell–Garnett model is retrieved when particle interactions are not taken into consideration. We shall also give some information on the optimal design of the effective anisotropic properties taking into account the shape of magnetic particles.

1. Introduction

Temperature, magnetic and electric fields in composite materials are of interest in many engineering applications. In order to use them effectively in modern constructions it is necessary to predict the effective homogenized properties. Theoretically, for particle filled, two phase composites, their homogenized, effective physical properties may be derived in the similar manner in the linear case. The methods for searching for effective magnetic permeabilities, dielectric permittivities or thermal conductivities are analogous and they may be calculated using the same approach. Although, the similarity of those three fields have been noticed a long time ago the models for the prediction of the effective composite properties are build separately for each class of problems and in addition with the use of various simplified hypothesis (assumptions).

It is worth to point out that the analysis of an incompressible viscous fluid flow through a porous medium can be described by the analogous equations as the mentioned above for the two phase composites. On the macroscopic scale, flow through the porous material is governed by Darcy's law having the permeability tensor K_H, so that the analogous methods to the discussed herein may be also successfully applied to the fluid flow problems.

There are different homogenization approaches and they can be divided into three classes, i.e.: direct, indirect and variational methods. Direct methods are based on volume average of field quantities and they can be performed by a numerical procedure, usually FEM or BEM. Indirect homogenization follows the idea of the equivalent inclusion method based on Eshelby's eigenstrain solution. In this area different variants of solutions are developed and they may be divided into: the self-consistent schemes, the Mori-Tanaka method and the differential method. The variational approach can give upper and lower bounds of the effective properties. Monographs describing in details different homogenization methods have been written by Mura [1], Nemat-Nasser and Hori [2], Qin and Yang [3]. The review presented by Wang and Pan [4] first examines the issues, difficulties and challenges in

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prediction of material behaviors by summarizing and critiquing the existing major analytical approaches dealing with material property modeling.

The model of Maxwell and Garnett [5,6] was one of the first to describe the effective permittivity of composites containing randomly dispersed spherical particles. Hashin and Shtrikman [7] used a variational approach to determine the upper and lower bounds of the effective magnetic permeability of multiphase materials. To investigate the effect of the interaction between particles, Fu et al. [8] presented an analytic approach to derive the explicit effective permittivity in a series form for composites containing spherical particles. For periodically distributed composites, McPhedran and McKenzie [9] and McKenzie et al. [10] extended a method devised by Rayleigh [11] to calculate the conductivity of simple cubic, body-centered cubic, and face-centered cubic lattices of composites containing conducting spheres. Doyle [12] and Lam [13] presented their models for composites with cubic lattices of particles. White [14] introduced a T-matrix solution based on a unit cell for general periodically distributed composites. Chen et al. [15] obtained the electric field distribution numerically in a Legendre series for one chain embedded in an infinite medium. Yin and Sun [16] derived effective magnetic properties for a chain-like structure considering a single column of particles embedded in an infinite medium. Sareni et al. [17,18] applied the boundary integral method to solve effective permittivity for random and periodic composites. A broad review of the above mentioned approaches and methods with the particular discussion of the results is given in the monograph Kanaun and Levin [19]. In addition to experimental and theoretical approaches, computer simulations have been adopted more and more frequently to study the effective properties of composites. Effective dielectric constants of two-phase composite dielectrics have been estimated numerically by Wu et al. [20]. Krakovsky and Myroshnychenko [21] used the finite element method to compute the effective permittivity for two-dimensional random composites. Numerical approach to the evaluation of the effective properties for MR fluids is demonstrated in Ref [22], however the analysis and results deal only with the prediction of magnetic permeability in one direction only. Although materials with directional features are common, most previous work has focused on the isotropic cases, except some studies attempted to bring this property-direction dependence into general formulation. For instance for MR fluids some magnetic properties, such as the saturation magnetization and the crystalline anisotropy, are intrinsic and depend mainly on the chemical composition and the crystalline symmetry of the material. On the other hand, extrinsic properties, such as remanence, coercivity and permeability, depend largely on the structure of the material Yin and Sun [16] or on the shape of reinforced particles [23]. Through analytical predictions and numerical modeling, certain optimization approaches and design schemes for novel materials could be resulted for engineering applications, and in turn the new observations and experiences from the practice would accelerate the development of new theories and methodologies.

Smart materials, by definition, have some properties which can be altered or tuned using an external field. Examples include materials that exhibit ferroelectricity, pyroelectricity, piezoelectricity, a shape memory effect, electrostriction, magnetostriction. electrochromism, photomagnetism and photochromism. Most of these materials tend to be used in their solid state, i.e. in a polycrystalline or a single crystal form as bulk materials or thin films deposited on appropriate substrates. In general they form a special class of two-phase composite materials. When an external field is applied, the particles become polarised and are thereby arranged into chains or clusters. The chains can further aggregate into columns, when the composite material exhibits a solid-like mechanical behaviour. Therefore, the effective properties of smart materials vary in time, starting from a random state of particles being in a viscous fluid and finishing in a composite solid being a chain-like structure.

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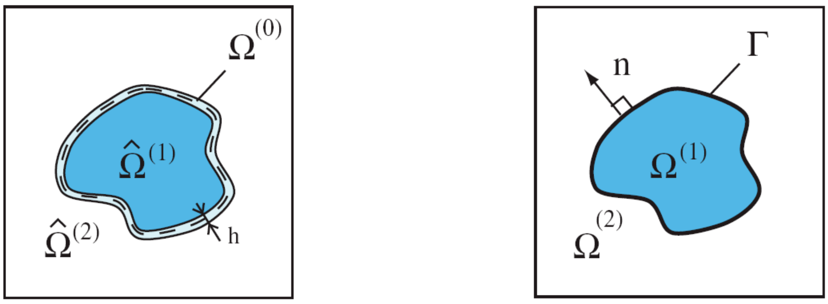
In order to demonstrate the theoretical limitations this study firstly aims to present the possibility of theoretical predictions of effective properties for smart materials in the 3D approach. The presented theoretical approach is based on the use of the effective field method. Having in mind the possible applications of two phase composites as smart materials two separate problems are discussed in details, i.e. the case of isolated inclusions and chain-like structures. Then, the numerical method of homogenization is proposed and the results are compared with theoretical ones. The suggested scheme of numerical homogenization is applied to optimize the effective properties varying the shape of inclusions. As it is reported in the literature the behaviour of aggregated particles has a great influence on the appropriate modeling of the smart material deformation as it is strained and especially in view of its yield stress value. For such a class of composites the present analysis is an introduction to the global FE modeling of rheological deformations.

2. The Effective Field Method

First of all, let us note that in engineering practice particles embedded in a matrix are usually coated by an additional material (an interface) between constituents (Fig. 1) in order to enhance various properties of composites, i.e. thermal, magnetic or dielectric. Surfactants are added to alleviate the settling problem. Thus, a particulate composite is made of three different phases. Such a class of problems is analysed for instance by Kamiński [24]. Since it is very difficult to estimate physical properties of an interface our analysis is limited to the considerations of two phase material demonstrated in Fig. 1b.



a) particles with interfaces b) two phase composites

Fig. 1. Fields in a homogeneous medium with inclusions

Now, consider a homogeneous medium with properties described by a tensor $C^0_{\alpha\beta}$. The medium contains the region $\Omega^{(1)}$ (inhomogeneity) with another property tensor $C_{\alpha\beta}$. The intensity E(x) and the flux D(x) of the field in the medium are described by the following system of differential equations:

$$\nabla_\alpha D_\alpha(x) = -q(x),\ D_\alpha(x) = C_{\alpha\beta}(x)E_\beta(x),\ \mathrm{rot}_{\alpha\beta}E_\beta(x) = 0 \quad (1)$$

where q(x) is a scalar density of the field sources. The third equation is satisfied automatically if the field $E_\alpha(x)$ is the gradient of a scalar function φ(x) called as the potential of the field:

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$$E_\alpha(x) = \nabla_\alpha \varphi(x) \tag{2}$$

The physical meaning of the potential φ(x) depends on the problem under considerations. It means the temperature field for heat transfer problems, the potential of the electric (magnetic) field for the electrostatics (magnetostatics, rsp.) fields.
The tensor $C_{\alpha\beta}$ represents the local properties of the inclusion – Fig.1b and is defined as the sum:

$$C_{\alpha\beta}(x) = C^0_{\alpha\beta} + C^1_{\alpha\beta}(x) \tag{3}$$

Thus, if we consider a homogeneous medium with the property tensor C^0 containing a set of inclusions with the property tensor C, the system of differential equations (1), (2) may be reduced to integral equations for the fields E(x) inside the inclusions, i.e.:

$$E_\alpha(x) + \int_{\Omega^1_{(k)}} K_{\alpha\beta}(x - x')C^1_{\beta\mu}(x')E_\mu(x')V(x')dx' = E^0_\alpha(x) \tag{4}$$

where V(x') is characteristic function for the kth inclusion that is equal to one if the variable x' belongs to the region $\Omega^1_{(k)}$ occupied by the kth inclusion and to zero if x' does not belong to the domain $\Omega^1_{(k)}$ (see Fig. 1). Here E_0 is the external field in the medium without the inclusion ($C^1(x)=0$) by the action of the same sources of the field. The kernel K(x) is determined in the classical manner:

$$K(x) = -\nabla_\alpha \nabla_\beta G(x) \tag{5}$$

where G(x) is the Green function for the infinite homogeneous medium with the property tensor C^0. The Green function satisfies the equation:

$$\nabla_\alpha C^0_{\alpha\beta} \nabla_\beta G(x) = -\delta(x) \tag{6}$$

The equivalence between the relations (4) and (1)-(3) can be proved under two additional assumptions:

- with the use of the potential $\varphi(x)$ the solution of eqn (1) can be decomposed as follows: $\varphi(x) = \varphi^0(x) + \varphi^1(x)$, where $\varphi^0(x)$ is the potential in the medium without the inclusion, and $\varphi^1(x)$ is the perturbation of the potential due to the presence of the inclusion that tends to zero when $|x| \to \infty$,
- the potential $\varphi^0(x)$ satisfies the relation analogous to eqn (1), i.e.: $\nabla_\alpha C^0_{\alpha\beta} \nabla_\beta \varphi^0(x)$ = -q(x).

The solution of this equation for an arbitrary anisotropic medium takes the following form:

$$G(x) = \frac{1}{4\pi r(x)}, r(x) = \sqrt{\det C^0 x_\alpha B^0_{\alpha\beta} x_\beta}, B^0 = (C^0)^{-1} \tag{7}$$

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The simplest version of the effective field method is based on the hypothesis that the local external field $E^*_\beta(x)$ that acts on each inclusion is the same for all inclusions, i.e. the field $E_\alpha(x)$ inside each inclusion can be presented in the following way:

$$E_\alpha(x) = \Lambda^E_{\alpha\beta}(x)E^*_\beta(x) \tag{8}$$

where the tensor $\Lambda^E_{\alpha\beta}(x)$ is determined from the solution of the problem for isolated inclusion in the medium with the property tensor C^0 by the action of the field $E^*_\beta(x)$ and for ellipsoidal inclusions takes the following form:

$$\Lambda^E_{\alpha\beta}(x) = \left(\delta_{\alpha\beta} + A^0_{\alpha\lambda}C^1_{\lambda\beta}\right)^{-1} \tag{9}$$

where

$$A^0_{\alpha\beta} = A^0_1 e^1_\alpha e^1_\beta + A^0_2 e^2_\alpha e^2_\beta + A^0_3 e^3_\alpha e^3_\beta,\ A^0_n = \frac{a_1 a_2 a_3}{2c_0}\int_0^\infty \frac{d\sigma}{\left(a_n^2+\sigma\right)\sqrt{\left(a_1^2+\sigma\right)\left(a_2^2+\sigma\right)\left(a_3^2+\sigma\right)}},\ n = 1,2,3 \tag{10}$$

a_1, a_2, a_3 are the semiaxes of an ellipsoid, and e^n_α are the unit vectors of the ellipsoid principal axes, the orientation of which is given by the normal *m*. For a spheroid inclusion with the semiaxes $a_1=a_2=a$, a_3 the tensor A^0 takes the following form:

$$A^0_{\alpha\beta} = A^0_1\theta_{\alpha\beta} + A^0_3 m_\alpha m_\beta,\ \theta_{\alpha\beta} = \delta_{\alpha\beta} - m_\alpha m_\beta,\ A^0_1 = \frac{1}{c_0}f_0(\gamma),\ A^0_3 = \frac{1}{c_0}(1\text{-}2f_0(\gamma)),\ \gamma = \frac{a}{a_3} > 1,$$

$$f_0(\gamma) = \frac{1-g}{2\left(1-\gamma^2\right)},\ g = \frac{\gamma^2}{\sqrt{\gamma^2-1}}\operatorname{arctg}\left(\sqrt{\gamma^2-1}\right) \tag{11}$$

The effective local exciting field $E^*_\beta(x)$ acting on the k-th inclusion is the sum of the external field $E^0_\beta(x)$ applied to the medium and the field induced by others surrounding inclusions. The field $E_\beta(x)$ inside the isolated k-th inclusion being in the background matrix and caused by the action of the field $E^*_\beta(x)$ is defined by eqn (4) where $E^0_\beta(x)$ is replaced by $E^*_\beta(x)$. The field induced in the region $\Omega^1_{(k)}$ of the k-th inclusion by all surrounding inclusions can be also represented with the use of eqn (4) in the following form:

$$E^*_\alpha(x) = E^0_\alpha(x) - \int_{\Omega^1_{(i)}} K_{\alpha\beta}(x-x')C^1_{\beta\mu}(x')\Lambda^E_{\mu\lambda}(x)V(x;x')E^*_\lambda(x')dx' \tag{12}$$

$$V(x;x') = \sum_{i\neq k} V_i(x') \ \text{ when } x \in \Omega^1_{(k)} \tag{13}$$

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Averaging the equation (12), assuming that the field $E^0_\alpha(x)$ is fixed in the problem. i.e.:

$$\left\langle E^0_\alpha(x)\right\rangle = E^0_\alpha \tag{14}$$

and identifying the conditional mean $\left\langle E^*_\alpha(x)\middle| x\right\rangle$ with the effective field E^*_α we obtain finally the following relation:

$$E^*_\alpha = E^0_\alpha - p\int K_{\alpha\beta}(x-x')P^0_{\mu\lambda}\Psi(x-x')E^*_\lambda dx' \text{ where } P^0_{\beta\lambda} = \frac{1}{\langle V_i\rangle}\left\langle \int_{\Omega^1_{(i)}} C^1_{\beta\mu}(x)\Lambda^E_{\mu\lambda}(x)dx \right\rangle \tag{15}$$

The symbol $\langle . \rangle$ denotes the averaging, and the function Ψ is defined as follows:

$$\Psi(x,x') = \frac{\langle V(x,x')|x\rangle}{\langle V(x)\rangle} \text{ where } \langle f(x)\,|\,x\rangle = \frac{\langle f(x)V(x)\rangle}{\langle V(x)\rangle} \tag{16}$$

p is the volume concentration of inclusions and it is equal to $\langle V(x)\rangle$. The symbol $\langle \cdot|x\rangle$ denotes the averaging over the realizations of the random set of inclusions by the condition $x \in V$. The solution of equation (15) can be expressed in the following way:

$$E^*_\alpha = E^0_\alpha\left(\delta_{\alpha\beta} + pK^\Psi_{\alpha\lambda}P^0_{\lambda\beta}\right)^{-1}, K^\Psi_{\alpha\lambda} = \int_{\Omega^1_{(i)}} K_{\alpha\beta}(x-x')\Psi(x-x')dx' \tag{17}$$

Multiplying Eqn (4) by the property tensor C^0 and using the definition (1b) of the flux tensor D(x) (for the inclusion and the medium) after a set of transformation of the result it is possible to derive the average of the flux tensor, i.e.:

$$\left\langle D_\alpha(x)\right\rangle = \left[C^0_{\alpha\beta} + P^0_{\alpha\lambda}\left(\delta_{\lambda\beta} + pK^\Psi_{\lambda\mu}P^0_{\mu\beta}\right)^{-1}\right]E^0_\alpha \tag{18}$$

On the other hand, using Eqs (1b), (3) the tensor of the effective (homogenized) properties of the composite may be defined as follows:

$$\left\langle D_\alpha(x)\right\rangle = C^*_{\alpha\beta}\left\langle E_\beta(x)\right\rangle, C^*_{\alpha\beta} = \left\langle C_{\alpha\beta}(x)\right\rangle \tag{19}$$

Combining eqs (18), (19) and (14) one can find that the tensor of the effective properties of two-phase composite is represented by the relation:

$$C^*_{\alpha\beta} = \left\langle C_{\alpha\beta}(x)\right\rangle = \left[C^0_{\alpha\beta} + pP^0_{\alpha\lambda}\left(\delta_{\lambda\beta} + pK^\Psi_{\lambda\mu}P^0_{\mu\beta}\right)^{-1}\right] \tag{20}$$

Taking into account eqs (12), (15) and assuming that the tensor $C^1_{\beta\mu}$ is independent on the x variable the above relation takes the following form:

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$$C^{*}_{\alpha\beta} = \langle C_{\alpha\beta}(x) \rangle = \left[C^{0}_{\alpha\beta} + pC^{1}_{\alpha\beta} \left\{ \delta_{\lambda\beta} + \left(A^{0}_{\lambda\mu} + pK^{\Psi}_{\lambda\mu} \right) C^{1}_{\mu\beta} \right\}^{-1} \right] \qquad (21)$$

If the ratio γ has the same order as the inclusion aspect ratio $K^{\Psi}_{\lambda\mu} = -A^{0}_{\lambda\mu}$ and the above equation is further reduced. Thus, as it may be easily observed the two-phase composite has isotropic, transversely-isotropic or anisotropic properties and they are directly dependent on the form of the operator $A^{0}_{\lambda\mu}$ - under the assumption that both the matrix and the inclusion have isotropic properties.

2.1. Isolated inclusions

Let the matrix material be isotropic ($C^{0}_{\alpha\beta} = c_0 \delta_{\alpha\beta}$) and the isotropic inclusions be ellipsoids with the same sizes randomly oriented in space. Neglecting the pair interaction between inclusions the analysis is reduced to the consideration of a single inclusion embedded in an infinite medium. Thus, the effective properties of the two-phase composite can be easily computed from the relation (21) with the use of the definition (11). For instance, in the case of spherical inclusions (so-called spherical symmetry) the operator $A^{0}_{\lambda\mu}$ takes the following form:

$$A^{0}_{\lambda\mu} = \frac{1}{3c_0} \delta_{\lambda\mu} \qquad (22)$$

the homogenized isotropic properties (21) coincide with the well-known Maxwell-Garnett formula [5,6].

2.2 Chain of inclusions

Using the above methodology the effective properties of two-phase composites may be also found for materials with regular lattices of identical inclusions. In this case the function $\Psi(x, x')$ in eqn (16) depends on the difference x-x'. From that definition one can find that:

$$\Psi(x) = \frac{\langle V(x', x'+x) | x' \rangle}{\langle V(x) \rangle} = \frac{\langle V(x', x'+x) V(x') \rangle}{\langle V(x) \rangle^2} \qquad (23)$$

For the i-th inclusion being in the regular lattice the integral:

$$\int V_i(x') V_i(x'+x) dx' = \frac{4}{3} \pi a_1 a_2 a_3 J(a),\ J(a) = \begin{cases} (1 - .5|x/a|^2)(1 + |x/a|/4), & |x/a| \le 2 \\ 0, & |x/a| > 2 \end{cases} \qquad (24)$$

Is the volume of the intersection of two identical ellipsoids with the center separated by the vector x. Using the above definition the numerator in eqn (23) can be written as follows:

$$\langle V(x', x'+x) V(x') \rangle = p \sum_{m} J(x - m) \qquad (25)$$

where m is the vector of the lattice composed by the centers of inclusions. Thus, the function $\Psi(x)$ can be expressed as follows:

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$$\Psi(x) = \frac{1}{p}\sum_m{}' J(x-m) \tag{26}$$

where the prime over the sum symbol denotes the exclusion of the term with m=0. Inserting the above results into the relation (17) one can find that:

$$K^{\Psi} = \int K\ (x-x')\Psi(x-x')dx' = -A^0 + \sum_m{}' \, p.v.\int K(x)\left[\frac{1}{p}J(x-m)-1\right]dx \tag{27}$$

and the symbols *p.v.* mean that the integral is understood in the sense of the Cauchy principal value. Finally, with the use of eqs (21) and (27) one can evaluate the effective properties of the composite with a regular lattice.

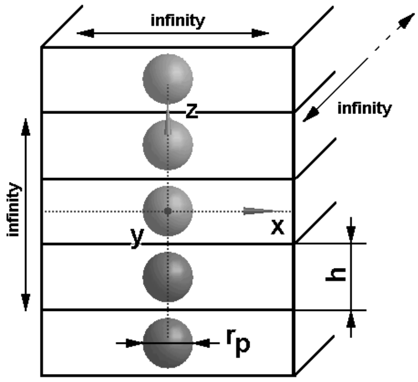

Fig.2 Chain of spherical inclusions

It is worth to emphasize that the above definition of the effective mechanical properties does not take into account the physical interaction between particles but the geometrical form of assumed elementary cells only. The analytical results of integration in eqn (27) can be obtained for specific forms of regular lattices only. For instance such a formula can be derived for an infinite chain of spherical particles where the representative volume cell has the form of a cuboid having infinite length in two directions – Fig. 2. In this case the effective properties can be expressed in the following form:

$$C^*_{ii} = \left[C^0 + pC^1\left\{1+(1-p)C^1A^0 + p\eta^1C^1\right\}^{-1}\right],\ i=1,2$$

$$C^*_{33} = \left[C^0 + pC^1\left\{1+(1-p)C^1A^0 + p\eta^3C^1\right\}^{-1}\right] \tag{28}$$

$$\eta^1 = 2\rho_0^3\sum_{m=1}^{\infty}1/m^3\ ,\ \eta^3 = -2\eta^1\ ,\ \rho_0 = r_p/h$$

assuming the isotropic properties of the matrix and inclusions, and the operator A^0 is described by eqn (22).

3. Numerical Homogenization Strategy

All known analytical methods are valid under certain limitations and particular geometries or classes of structures. For metamaterials comprising conducting and possibly

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resonant elements, and for which the periodicity is not necessarily negligible relative to the free-space wavelength, analytical homogenization techniques are unreliable or not applicable. Our intent here is to justify a numerically based homogenization scheme based on eqn. (1), in which the local fields computed for one unit cell of a periodic structure are averaged to yield a set of macroscopic fields. Once having computed the macroscopic fields, we can then determine the constitutive relationships between the macroscopic fields, arriving at the effective electromagnetic parameters. We will see that there will be virtually no restrictions on the contents of the unit cell, nor will the unit cell necessarily need to be small in comparison with the wavelength. They have proven useful in many situations, e.g. low volume fraction of homogeneous spherical or ellipsoidal inclusions in a homogeneous host material, but they fail if the volume fraction is too high or if the inclusions are not spheres or ellipsoids. Then, a more accurate homogenization procedure has to be used, which includes all contributions of the interaction between the reinforcing particles.

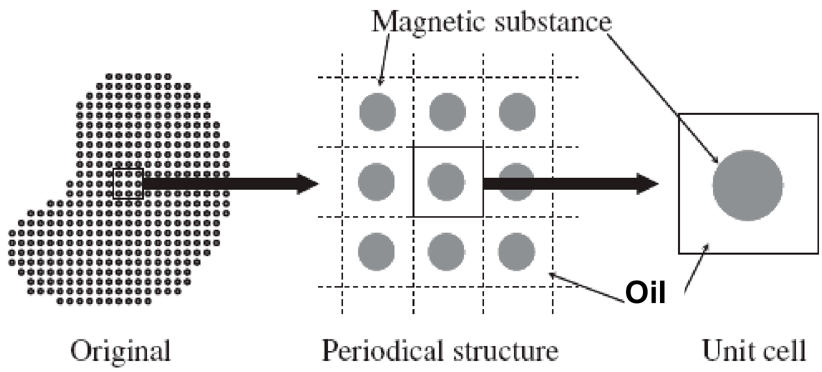

Figure 3. Definition of a unit cell

For two-phase composites a typical homogenization situation is depicted in Fig. 3. It shows a 2D model of the composite material, which includes matrix with inclusions. In the homogenization method, the structure of the two-phase composite materials is assumed to be periodic, and the unit cell, which is the minimum volume to represent the overall statistics, is defined. Here, it is assumed that one particle inclusion is located in the center of the cell. The unit cell is regarded as a homogeneous substance with the effective properties. The effective property is defined on the basis of energy balance in the unit cell (Fig.3): it is assumed that the original cell and the homogenized cell include equivalent energy when both unit cells are immersed in equivalent external field.

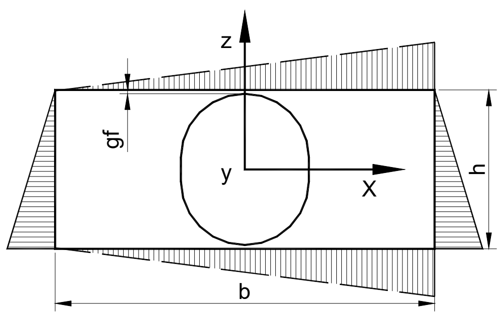

Figure 4 The geometry of the representative unit cell and of the boundary conditions – 2D problem.

In an actual estimation, the solution of the Laplace equation obtained with the use of eqs (1), (2) is computed by FEM. In this analysis, the potential φ is unknown, and assuming the applied field is unidirectional at least in the cell, the boundary conditions are set as:

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$$\varphi = 0 \text{ at the bottom } x = 0, \ \varphi = E_y x \text{ at the top } x = b/2 \tag{29}$$

The identical boundary conditions are formulated at each parallel boundaries of the cell shown in Fig.4 for 2D case. Let us note that the above type of boundary conditions satisfies periodicity of boundary conditions for arbitrary type of regular lattices as well as for the chain of inclusions and for a single inclusion.

Similarly as previously for the effective field method, the FE analysis is based on the averaging method that is carried out for the representative volume element (RVE) having the volume denoted by the symbol Ω_{RVE}. Thus, it is obvious that the results, understood in the sense of the average property tensor, are directly dependant on the dimensions and form of the RVE. It is worth to point out that even for the 2D two phase periodic composites the RVE may be of an arbitrary form, not necessarily rectangular as it shown in Fig. 3. Since in our numerical analysis we intend to give an information about variations of the property tensor components with respect to the volume fraction p we define it in the following way (see Fig.4):

$$p = \frac{\pi r_p^2}{2br_p\left(1 + g_f / r_p\right)} \text{ for 2D and } p = \frac{2\pi r_p^3}{3b^2 r_p\left(1 + g_f / r_p\right)} \text{ for 3D} \tag{30}$$

where it is assumed that the RVE has a square cross section in the y direction. Thus, for the prescribed volume fraction p the RVE is completely defined by the set of two parameters (geometrical ratios), i.e.: g_f/r_p and b/r_p for both 2D and 3D cases. Let us note that for the constant volume fraction p and the constant interparticle distance g_f the geometrical dimensions of the representative cell (i.e. b and h) are uniquely determined

For the selected RVE (Fig.4) and the selected boundary conditions in the form (29) (the unidirectional external field) the average intensity and flux of the field are defined by:

$$\left\langle E_\alpha \right\rangle = \sum_{m=1}^{TN} E_{\alpha m}, \left\langle D_\alpha \right\rangle = \sum_{m=1}^{TN} D_{\alpha m} \tag{31}$$

where TN denotes the total number of nodes in the FE mesh. Using the above relations it is possible to compute four components of the average intensity and flux of the field for two types of boundary conditions demonstrated in Fig.4 (the 2D problem) or nine for the 3D analysis. We do not know in advance how many nonzero components in the property matrix $C^1_{\alpha\beta}$ occur. Therefore, for the linear problem all components of the property matrix $C^1_{\alpha\beta}$ (nine for the 3D problem) can be derived directly from eqs (1),(2) where the appropriate components of the vectors **E** and **D** are replaced by their average values evaluated in the local cell (RVE).

For the non-linear problem (understood in the sense of the non-linear relation (1)) the components of the property matrix are computed by the comparison of the magnetic energy of the homogenized and of the original (Fig. 4) unit cell combined with the Newton-Raphson method. The energy is represented as follows:

$$U = \int_{\Omega_{RVE}} \int_{\mathbf{D}} \mathbf{E} \mathrm{d}\mathbf{D} \, \mathrm{d}\Omega \tag{32}$$

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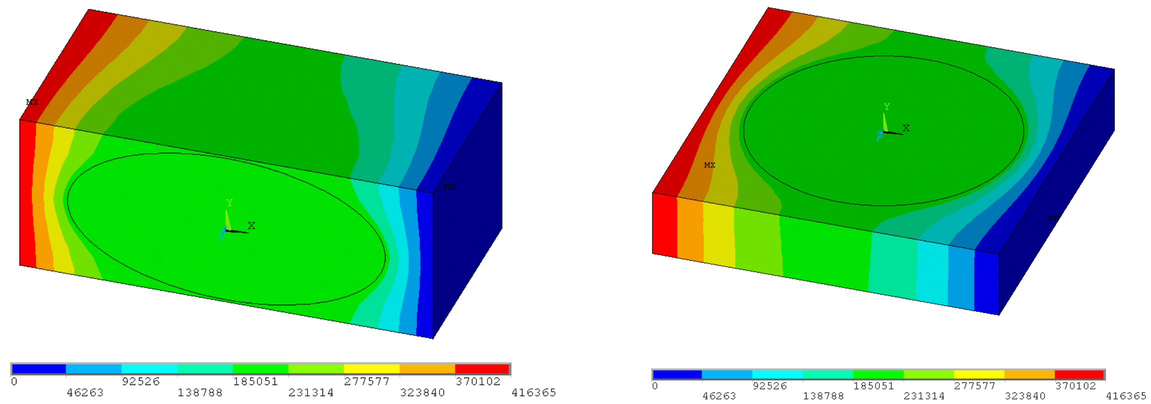
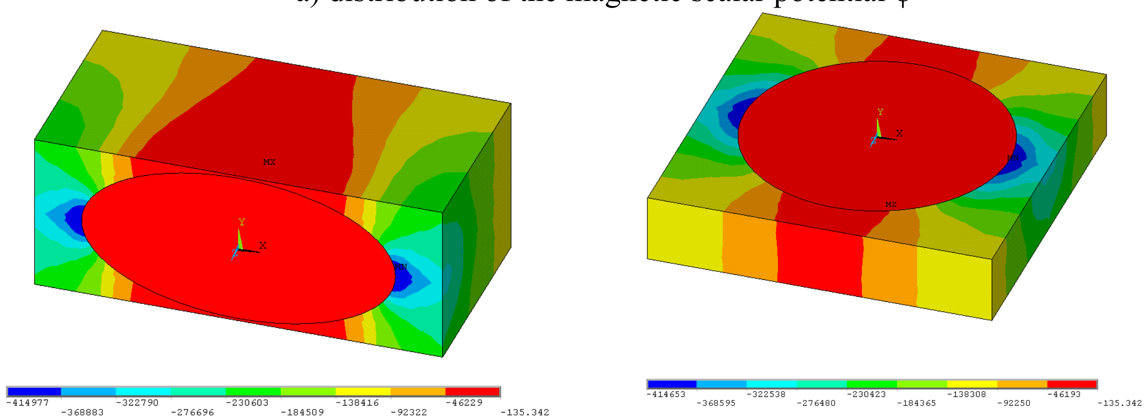
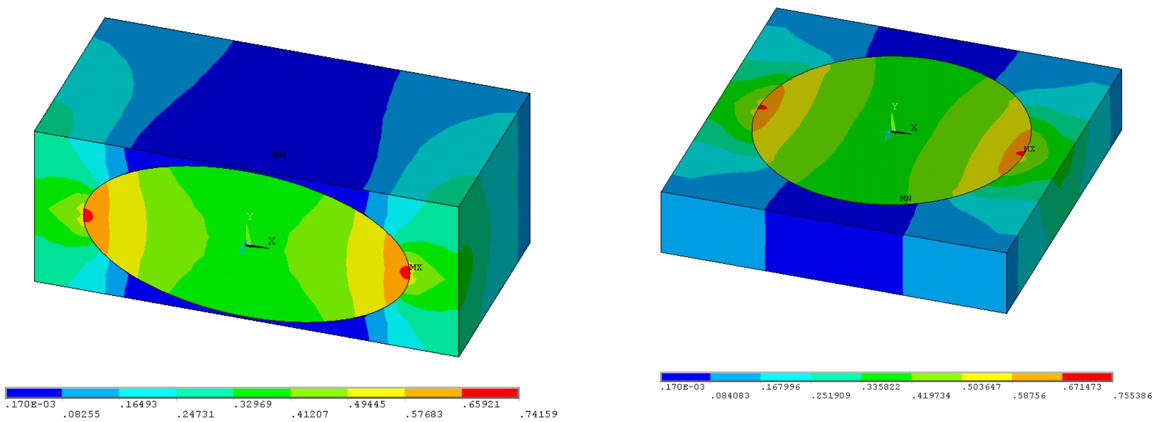
However, it is necessary to know in advance the **D-E** characteristic curve for the inclusions. Commonly, the inclusion is modeled as isotropic material but for the unit cell the local property matrix can possess anisotropic properties as it is shown for example in section 2.



a) distribution of the magnetic scalar potential φ



b) distribution of the magnetic field intensity



c) distribution of the magnetic flux density

Figure 5. The local distributions of magnetic fields for the boundary conditions (29) – spheroids $a_x/a_y = 2$, $p = 0.3$, $r_p/h = 0.485$, $\mu_0=1$, $\mu_p=2000$.

Figure 5 represents the example the FE analysis conducted for two-phase ferromagnetic composites made of phases having magnetic properties – the magnetostatic problem. The plots demonstrate the distribution of the magnetic flux density, the magnetic field and the potential φ as the external magnetic field is applied at the y direction – the

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boundary conditions (3c). As it may be seen the variations of the potential inside the local cell (Fig. 5a) result in non-homogeneous distributions of magnetic fields in all directions. The average values are evaluated by adding the values of each finite elements in the elementary cell.

For two-phase composites the effective property tensor has analogous properties to e.g. the tension modulus in elasticity: if the behavior is identical in three perpendicular directions, then it is isotropic. This conclusion points out a limitation of the use of constant second order tensors for the description of behavior. Indeed, for e.g. magnetism many experimental observations reveal that cubic single crystals are not magnetically isotropic (see for instance [25] for iron and nickel or [26] for Terfenol-D). In fact in the experiments the chains of particles do not have to be aligned in the direction of the external field, namely E_y. We have some more compact aggregates. The microscopic analysis on the structure of the two-phase composites revealed that there were aggregates forming rather than chains of spheres that can be approximated by ellipsoids, stripes or cylinders. Therefore, it is interesting to verify the correctness of the introduced FE model in the cases when the external field is rotated with respect to coordinates defining RVE in order to consider the relationship between the orientation of inclusions and to explore symmetries in the constitutive relations (1) and their relevance to the homogenized composite medium. In the case of a generalized anisotropic structure for which principal axis of external fields and the unit cell do not coincide the property tensor should satisfy the classical transformation rules of the second rank tensors. To interpret and investigate those effects let us analyze the form of the property (permeability) matrix for the 3D ring structure shown in Fig. 6. The geometry of unit cells is defined in the cylindrical coordinate system but the external field is directed along the line joining the centre of the central spherical inclusion and the centre of the ring curvature.

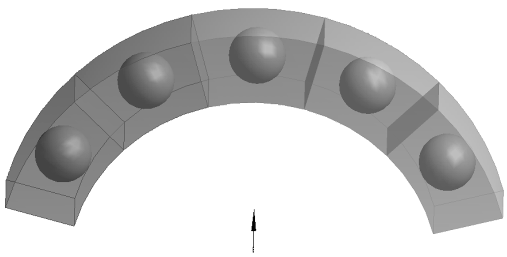

Figure 6 The system of 5 ferromagnetic particles – the external field is applied at the x, y or z direction.

The assumed form of the system of the unit cells reflects the situation as the external field is not always parallel to the cell edges . It may occur for instance for clusters of inclusion. Figure 7 shows the assumed boundary conditions and the values of the permeability matrix terms (the magnetostatic problem). The terms $\mu_{\alpha\beta}$ ($\alpha \neq \beta$) are not equal to zero what means that it may be for instance the origin of clusters and of the inclusion aggregation at the beginning of magnetization. On the other hand, it may be easily verified that the terms of the property matrix satisfy the classical transformation rule:

$$\langle \mathrm{C} \rangle^{Transf} = \mathrm{M} \langle \mathrm{C} \rangle \mathrm{M}^{\mathrm{T}}, \text{ where } M = \begin{bmatrix} \cos\theta & \sin\theta & 0 \\ -\sin\theta & \cos\theta & 0 \\ 0 & 0 & 1 \end{bmatrix} \tag{33}$$

and θ denotes the angle of rotation which is equal to 14.11^0.

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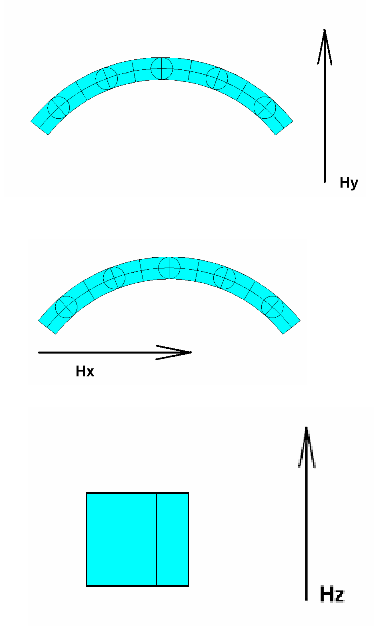

the central cell

$$\{\mu\}_{\text{eff}} = \begin{bmatrix} 2.18155 & 0 & 4.04284\cdot10^{-15} \\ 4.42751\cdot10^{-13} & 4.64986 & 4.83967\cdot10^{-14} \\ 3.63850\cdot10^{-14} & 0 & 4.75623 \end{bmatrix}$$

the intermediate cell

$$\{\mu\}_{\text{eff}} = \begin{bmatrix} 2.32835 & 0.58379 & 2.19868\cdot10^{-14} \\ 0.58380 & 4.50307 & 8.14158\cdot10^{-14} \\ 0 & 0 & 4.75623 \end{bmatrix}$$

the outermost cell

$$\{\mu\}_{\text{eff}} = \begin{bmatrix} 2.73351 & 1.02822 & 7.87749\cdot10^{-14} \\ 1.03033 & 4.09986 & 7.54181\cdot10^{-15} \\ 0 & 0 & 4.75623 \end{bmatrix}$$

Figure 7 Boundary conditions and the local (transformed off-axis) values of the permeability matrix.

4. Numerical results

The homogenization method is applied to various 2D and 3D test problems in order to evaluate the distributions of the terms of the property matrix and to compare theoretical predictions with numerical ones that take into account the finite dimensions of the unit cell. In the test problems, a sample two-phase composite material composed of an isotropic matrix and inclusions having the following material properties: $c_p=1$ and $c_f=2000$. The analysis is conducted for 2D and 3D unit cells to test the capability and limitations of the proposed model. Similarly as previously the numerical model corresponds to the analysis of magnetorheological fluids.

4.1 2D Problems

Let us consider a single circular particle surrounded by a nonmagnetic carrier fluid – the planar problem Fig.4. This is a typical homogenization problem analyzed for the MR fluids – see e.g. Simon *et al.* [27] for magnetorheological fluids. However, on the contrary to the cited work we compute the four (the planar problem) permeability matrix coefficients. The off-axis terms are equal to zero, and two others are plotted in Fig. 8 for various volume fractions.

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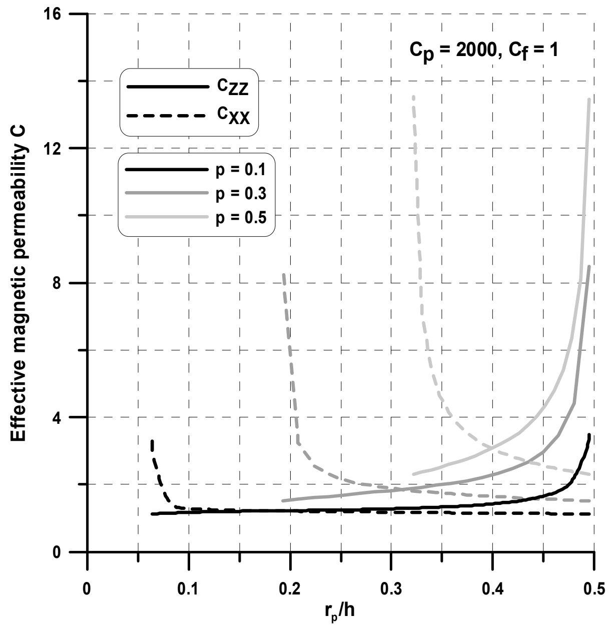


Figure 8 Variations of the effective permeability at two perpendicular directions with particle radius.

As it may be seen the effective permeability c_{zz} decreases as the interparticle distance increases and it is the highest for the highest volume fraction p=0.3. The decrease of the effective permeability c_{xx} is associated with the increase of the effective permeability c_{zz}. Let us note that for the constant volume fraction p the variations of the ratio r_p/h (or g_f/r_p) results in the change of the ratio b/r_p – see eqn (30). Using the single unit cell (Fig.4) one can observe that for the external magnetic field having the non-zero component H_y only, the chains of ferromagnetic particles are completely isolated since there is no interaction at the x direction. In fact, the experiments demonstrate evidently that they form clusters of different shapes - see Bossis *et al.* [28].

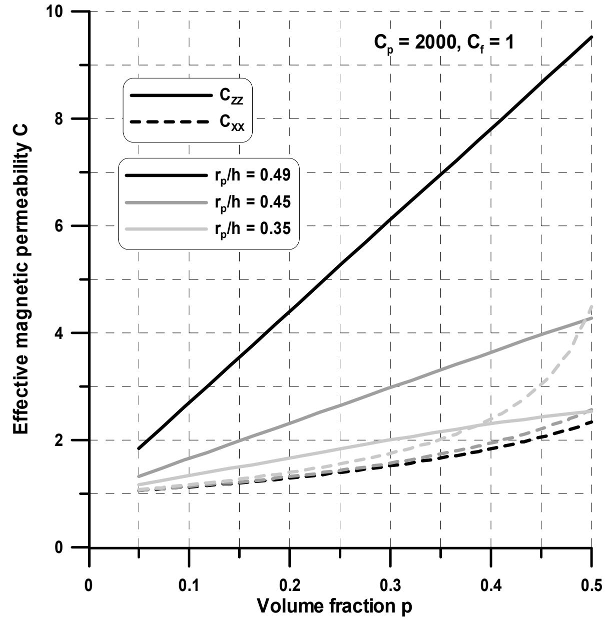


Figure 9 Variations of the effective permeability at two perpendicular directions with volume fractions

3.2 3D Problems

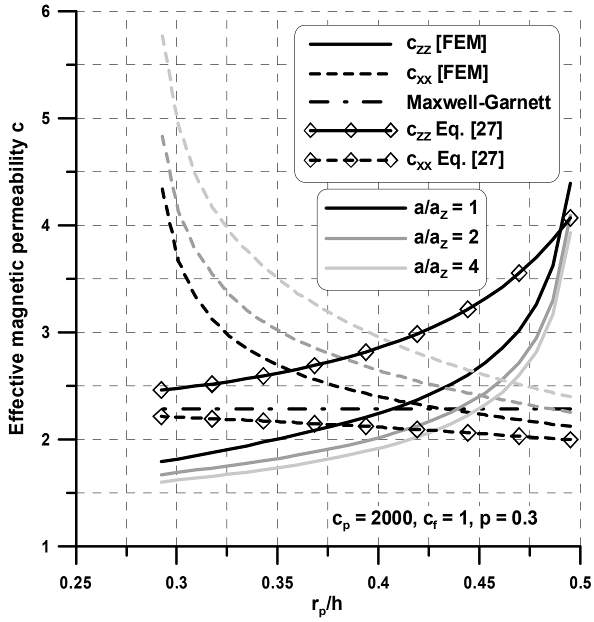


Figure 10 Variations of the permeability coefficients with interparticle vertical distance r_p/h (transversely-isotropic body)

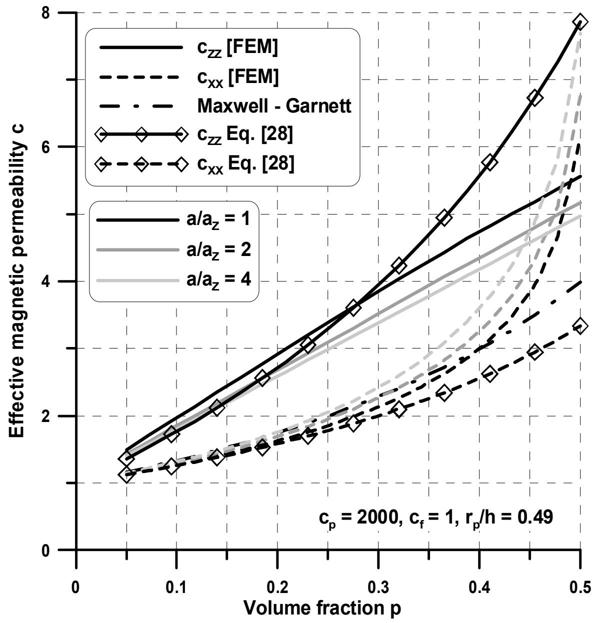


Figure 11 Variations of the permeability coefficients with volume fraction p (transversely-isotropic body)

Now, 3D Laplace equation (2) have been solved for 3D unit cell using FE package. The results for the variations of the property tensor components are demonstrated in Figs 10 and 11 taking into account the assumption (30) what leads directly to the transversely isotropic properties of composites (i.e. $c_{xx}=c_{yy}$). The plots are drawn both for spherical ($a=a_x=a_y=a_z$) and spheroid ($a=a_x=a_y\neq a_z$) inclusions. For spherical inclusions the distributions of the homogenized properties for the 3D case are similar to those for the 2D case (see Figs 8 and 9). However, assuming the identical geometric ratios of the RVE, for the 3D case the averaged values in the x direction are higher than those evaluated for the planar case, whereas the values in the z direction are almost identical. The shape of inclusions has a significant influence on the averaged values. For the increasing γ parameter the effective property c_{zz} decreases, and c_{xx} increases since the x axis corresponds to the longer axis of spheroids. Figs 10 and 11 show that the proposed model provides an transversely isotropic effective permeability, whereas the Maxwell–Garnett model gives an isotropic one. The Maxwell–Garnett model always yields the same estimates for any r_p/h because it is insensitive to microstructure. Thus the Maxwell–Garnett model cannot be used for these composites because, even when the overall volume fraction is very small, the distance between particles of the same chain is small so that particle interactions cannot be disregarded. For the low volume fractions the Maxwell-Garnett gives a very good estimations in the x directions only – Fig.11. For spherical inclusions, as it may be observed in Figs 10 and 11, the effective field model gives much better approximations of the effective values evaluated with the use of the FE model than the Maxwell-Garnett model. However, the effectiveness of the effective field model decreases for the high particle interactions ($r_p/h<0.1$) and for the high volume fractions ($p>0.25$). It is obvious that theoretical estimations, i.e. with the use of the Maxwell-Garnett model and the effective field model have limited applications in the comparison with the FE model since the first corresponds to the random (quasi-isotropic) structure of reinforced particles, and the second to the chain-like structure, i.e. in two directions the dimensions of the RVE tend to infinity.

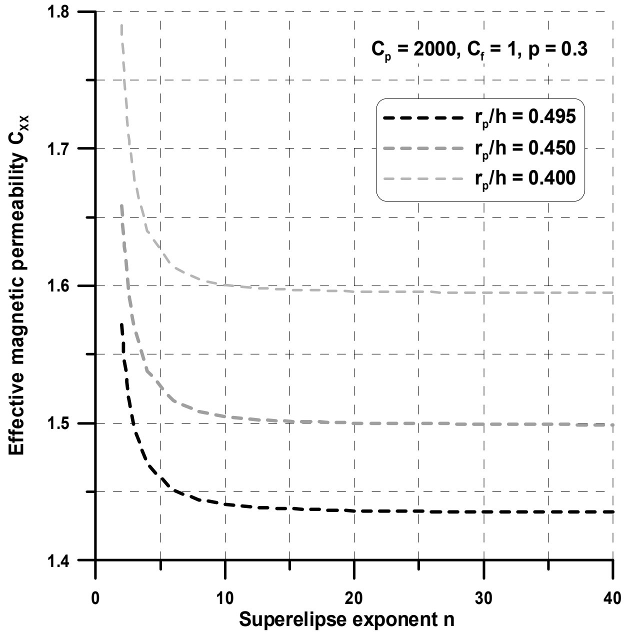
5. Optimal Design

In theoretical and numerical analysis it is commonly assumed that the reinforcement particle has an ideal spherical form. However, as it is demonstrated in Figs 10 and 11 the shape of the particle can affect significantly the effective material properties. Therefore, development of optimized multifunctional composite materials becomes of great interest from technological and theoretical viewpoints to all engineering fields. This section designs such materials computationally using the method of parametric optimization. In particular, two-dimensional periodic two phase composite materials are optimized for the optimal effective properties. To analyse the effects of different shapes for simplicity it is assumed that the particle is modelled as a superellipse (Fig.4) having the following form:

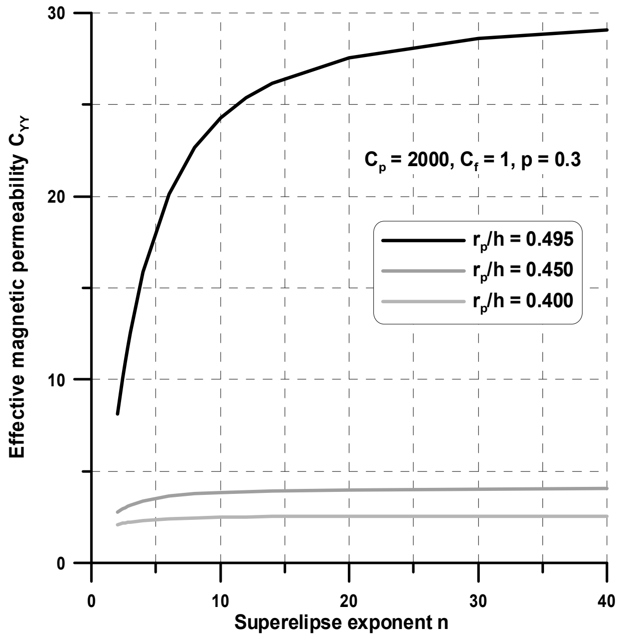
$$\left(\frac{x}{a}\right)^n + \left(\frac{y}{b}\right)^n = 1 \tag{34}$$

where a and b denote the superellipse semi-axis, and n is a parameter greater than 1. Having the constant volume fraction in the RVE and varying the n value one can observe the change of the terms in the effective property matrix.

For higher values of the n parameter ($n>10$) the shape resembles a rectangular and in this case both components of the effective permeabilities, i.e. c_{yy} and c_{xx} reach their optimal values – see Figs 12 and 13. However, the optimal values of the effective properties are strongly dependent on the values of the geometrical ratios r_p/h and the volume fractions p. Let us note that maximal value of the term c_{yy} is much higher than those plotted previously in the section 4, and the values of c_{yy} are much lower. Therefore, it seems to be reasonable to conclude that the optimal rectangular form of the particles can prevent the aggregation of them in ellipsoids or cylinders instead of linear chains and in this sense the theoretical effective field model may be applicable in the estimations of the effective properties. It is worth to mention also that the obtained optimal designs resembles completely those obtained by Guest and Prevost [29] for fluid transportation problem (the Darcy law). They concluded that the Schwartz P minimal surface is believed to be the maximum permeability structure in the 3D case. However, the authors of the cited paper assumed in advance the isotropic properties of the permeability matrix.



a) the x direction



b) the y direction

Figure 12 Variations of the effective properties with the particle shape – the constant volume fraction p

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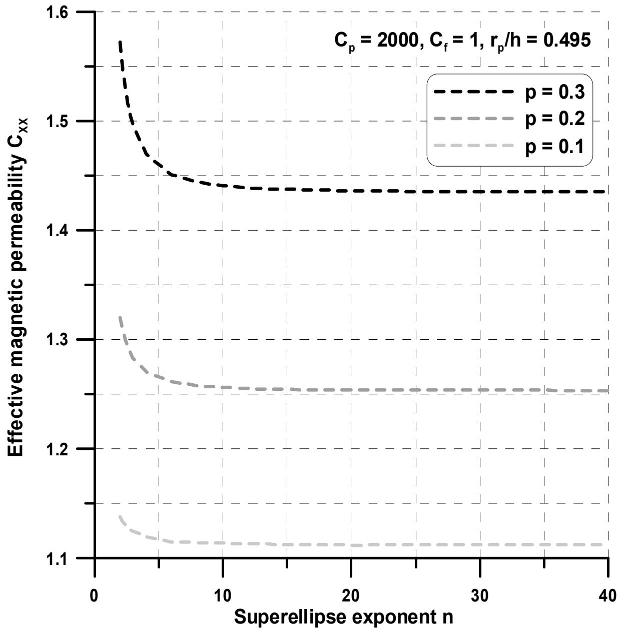
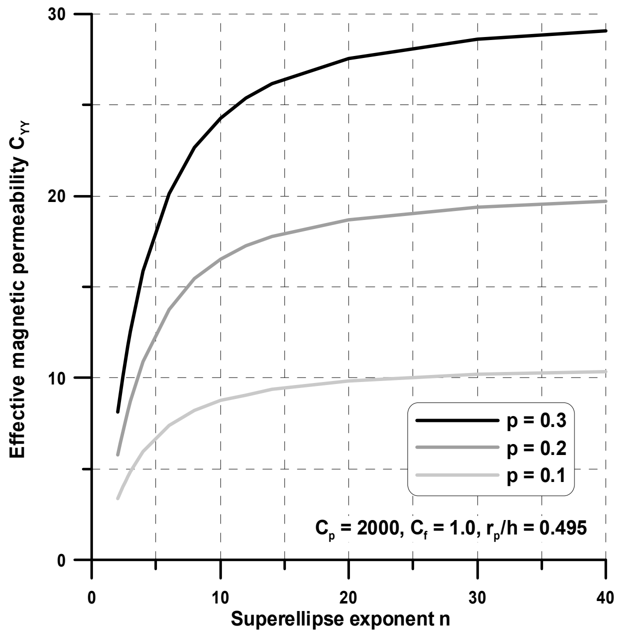


a) the x direction



b) the y direction

Figure 13 Variations of the effective properties with the particle shape – the constant interparticle distance r_p/h

6. Concluding Remarks

Properties of a heterogeneous medium (two phase composites) made of inclusions distributed in a locally periodic way in a matrix have been derived and studied. A uniform test

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external field is applied on the boundary of the composite, and then the averaged fields of the particles and matrix are derived by the Green's function technique and then compared with FE results based on the numerical homogenization technique. An anisotropic effective property tensor is further provided. The effective property tensor of the composite medium is symmetric, positive definite, generally anisotropic, and depend on the microstructure both for 2D and 3D cases. The proposed method can be successfully applied to the analysis of the non-linear problems, taking into account the non-linearity of the characteristic curves (e.g. **B-H)**. From these models it is found that the averaged property tensor components are strongly dependent on the dimensionless interparticle distance and the volume fraction.

This paper proposes also a shape optimization methodology for designing multifunctional two phase composite material optimized for tensor property components. For the 2D problems the optimal shape resembles rectangular with rounded edges. It is verified in this study that optimal design based on the finite element analysis is a valid method for the output improvement of constructions.

It is important to emphasize that the underlying methodology of homogenization and optimization is quite general and can be applied to the design of composite materials.

Conflict of interest

No conflict of interest.

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