ECCOMAS Congress 2016
VII European Congress on Computational Methods in Applied Sciences and Engineering
M. Papadrakakis, V. Papadopoulos, G. Stefanou, V. Plevris (eds.)
Crete Island, Greece, 5–10 June 2016

ON THE INFLUENCE OF THE INCLUSIONS' MORPHOLOGY ON THE ACCURACY OF THE PREDICTION OF REINFORCED COMPOSITES MECHANICAL BEHAVIOUR

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Keywords: Mean-field approaches, Fast Fourier Transform, Mechanical behaviour, Composites Materials, ellipsoidal and spherical reinforcements.

Abstract. *Nowadays, the prediction of mechanical behaviour of reinforced composites is a paramount problem in computational materials science. The macroscale effective coefficients are obtained from the microscale information also known as the so-called corrector problem. A numerical strategy based on integral equation known as the periodic Lippmann-Schwinger equation is used in the present study. For that purpose, several Representative Volume Elements (RVE) are generated with a mixture of spherical and ellipsoidal inclusions, by the Random Sequential Adsorption scheme or the time driven Molecular Dynamics. In this work, the influence of the inclusions morphology on the accuracy of some classical mean-field approximation methods and a full-field computational method based on numerical homogenization techniques to predict the mechanical properties of these materials is studied. For low volume fractions of inclusions, the results of the mean-field approximations and those of the Fast Fourier Transform-based (FFTb) full-field computation are very close, whatever the inclusions' morphology. For RVEs consisting of ellipsoidal or a mixture of ellipsoidal and spherical inclusions, when the inclusions volume fraction becomes higher, one observes that Lielens' model and the FFTb full-field computation give almost similar estimates. The accuracy of the computational methods depends on the shape of the inclusions and their volume fraction.*

1 INTRODUCTION

During the last decades, the micromechanics of heterogeneous materials, which is an area of very fertile researches at the boundary between physics and mechanics of materials, evolved from the so-called mean-field approaches to full-field schemes such as Finite Element Method (FEM) or more recently Fast Fourier Transforms (FFT) methods initiated by Moulinec and Suquet [1]. The aim of these approaches is to predict the effective behaviour of reinforced composite materials which is strongly influenced by the microstructure (e.g., constituents' properties, volume fractions, shapes, orientations, etc.). In such materials, inclusions can have several morphologies with different geometrical orientations. To exactly describe the effective behaviour of reinforced composite materials, a highly accurate description of the microstructures is required.

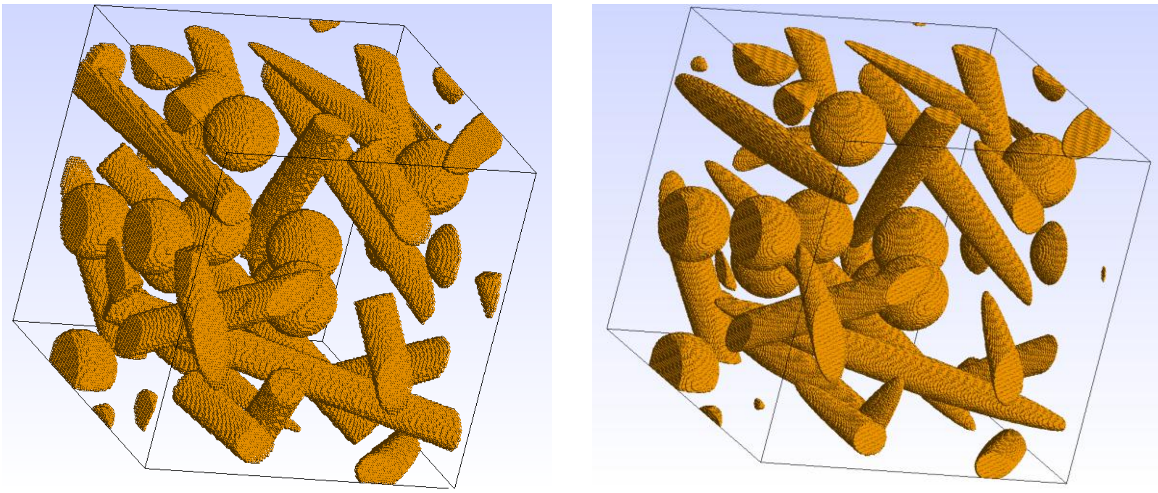
The full-field approach used in this paper is the Fast Fourier Transform-based (FFTb) homogenization technique [2] where an iterative scheme is used for computing effective properties of each given RVE (Representative Volume Element). One of the main advantages of this approach is its low time and memory consumption in comparison for example with finite elements methods. The application of the FFTb methods to the study of the overall behaviour of materials involves a preliminary step of 3D RVE generation. Another important feature of this method is that the generated RVE well approximates, at least in certain aspects, the real microstructure of the material. Mean-field homogenization methods rely on a simple statistical analysis of the microstructure.

Several works were conducted to highlight the validity domains of these mean-field micromechanical models (by comparing them to full-field approaches) when predicting the elastic properties of various RVEs with different microstructures. Most of these studies have been focused on spherical particles reinforced composites, aligned fibers reinforced composites, randomly oriented fiber reinforced composites or microstructures with aligned or randomly oriented clay platelets.

In this work, the effective material properties obtained using the numerical FFTb homogenization techniques were compared to three different analytical methods: the normalized self-consistent scheme (NSC) [3,4], the Mori–Tanaka model (MT) [5] and Lielens' model [6]. The aim of this paper is to exhibit the influence of the inclusions morphology on the accuracy of these homogenization techniques to predict the mechanical properties of reinforced composites consisting not only for spherical or ellipsoidal inclusions but also for a mixture of spherical and ellipsoidal inclusions.

2 RVE GENERATION AND FFT-BASED HOMOGENISATION

In this paper, we focus on the influence of the inclusions morphology on reinforced composite materials. In order to study the heterogeneous medium, one needs to have access to RVEs. Basically there are two main families of algorithm to design an RVE with complex microstructures. The First technique is based on the RSA method (Random Sequential Adsorption) whereas the second technique is based on MD (Molecular Dynamics). In this paper our choice goes to the latter one. Actually this approach is richer, more fruitful to design RVEs. Moreover this approach is reliable and fast enough to build RVEs. Indeed, in this technique, one can play with different kind of parameters which allow one to design RVEs with various inclusions such as spheres, cylinders, ellipsoids or a mixture of different kind of these simple geometries. The MD-based method is also powerful for higher volume fractions (of order $50 - 60\%$): MD generates a configuration in about a second whereas the RSA can get stuck. The classical RSA and the time-driven version of MD applied to generate the mixture of inclusions of spherical and cylindrical shapes will not be presented is this paper. A detailed description can be found in [7]. An example of a sample with a mixture of nonintersecting spherical and cylindrical inclusions is presented on the figure 1a. Because mean-field approaches usually describe inclusions shape by the means of ellipsoids, we introduced imperfections (see [2] for more details) on the cylinder inclusions without spoiling the efficiency of the generation algorithm, in order to obtain ellipsoidal inclusions instead of cylinder. The figure 1b shows the same RVE as in figure 1a where the extremities of cylinder inclusions are rounded in order to obtain ellipsoidal inclusions.



(a) (b)

Figure 1: Examples of 3D view of generated RVEs: (a) spherical and cylindrical inclusions, (b) spherical and ellipsoidal inclusions.

Once the RVE are generated, one seeks to find out the effective mechanical properties. For that purpose we have developed a homogenization technique based on the Lippmann-Schwinger equation, since the RVE are periodic. Actually, a mechanical homogenization problem for periodic heterogeneous media, leads to solve a microlocal problem, also known as the corrector problem, in which the corrector term is the periodic unknown. The corrector term is then a solution of an elliptic partial differential equations (PDE). From a mathematical point of view, solving a PDE returns to determine the Green function associated with the PDE. Hence, the above problem is a Fredholm integral equation leading to the Lippmann-Schwinger equation (see Moulinec and Suquet [1]) which can be solved using the FFT techniques. In the sequel, this approach has been privileged and is called the FFT-based homogenization (FFTb).

3 MEAN-FIELD HOMOGENIZATION MODELS

Because the inclusions frame of reference *Ri* attached to its main axes are not always identical to the RVE coordinate system *RI*, subscripts "*Ri*" and "*RI*" are added, for the sake of clarity, to each tensor to indicate the coordinate system in which it is expressed. One can note that the subscripts "*Ri*" and "*RI*" are not necessary when the inclusions are spherical or all aligned, because in this case *Ri* and *RI* are the same. For randomly oriented inclusions reinforced composites, the geometrical orientation of each inclusion is described by three Euler angles $(\varphi_1,\phi,\varphi_2)$. The transition between the local coordinate system *Ri* of the inclusion and the RVE system *RI* is made by these three Euler angles in the Bunge convention [8] where the transformation matrix is given by the following equation (1). This matrix is applied, for a given rank four tensor K, as showed in equation (2).

$$m(\varphi_1,\phi,\varphi_2)=\begin{bmatrix} cos\,\varphi_1\,cos\,\varphi_2-sin\,\varphi_1\,sin\,\varphi_2\,cos\,\phi & -cos\,\varphi_1\,sin\,\varphi_2-sin\,\varphi_1\,cos\,\varphi_2\,cos\,\phi & sin\,\varphi_1\,sin\,\phi \\ sin\,\varphi_1\,cos\,\varphi_2+cos\,\varphi_1\,sin\,\varphi_2\,cos\,\phi & -sin\,\varphi_1\,sin\,\varphi_2+cos\,\varphi_1\,cos\,\varphi_2\,cos\,\phi & -cos\,\varphi_1\,sin\,\phi \\ sin\,\varphi_2\,sin\,\phi & cos\,\varphi_2\,sin\,\phi & cos\,\phi \end{bmatrix} \quad (1)$$

$$_{RI}K_{ijkl}=m_{im}m_{jn}m_{ko}m_{lp}\ {}_{Ri}K_{mnop} \quad (2)$$

The homogenized stiffness tensor C of a composite consisting of n different types of inclusions (in terms of shape and geometrical orientation) in the RVE coordinate system RI is:

$$C=\left\langle {}_{RI}C\ :{}_{RI}A\right\rangle_{RVE}=f_m\ {}_{RI}C_m:{}_{RI}A_m+\sum_{i=1}^{n}f_i\ {}_{RI}C_i:{}_{RI}A_i \quad (3)$$

where f_i, C_i and A_i denote respectively the volume fraction, the stiffness tensor and the strain-localization tensor of inclusions exhibiting the same shape and the same geometrical orientation. f_m, C_m and A_m denote respectively the volume fraction, the stiffness tensor and the strain-localization tensor of the matrix. $A:B$ denotes the double scalar product using the Einstein summation convention. $\langle\ \rangle_{RVE}$ stands for the volume average over the whole RVE (matrix + inclusions). From the average strain theorem, one can obtain:

$$C={}_{RI}C_m+\sum_{i=1}^{n}f_i\left({}_{RI}C_i-{}_{RI}C_m\right):{}_{RI}A_i \quad (4)$$

This is the main relation used to calculate the homogenized stiffness tensor. The strain-localization tensor A_i in this relation differs from one model to another. In the normalized self-consistent [3,4], Mori-Tanaka [5] and Lielens' [6] models, the strain-localization tensors are expressed respectively as follows:

$$_{Ri}A_i^{NSC}=\left[I+{}_{Ri}E:\left({}_{Ri}C_i-{}_{Ri}C\right)\right]^{-1}:\left\langle\left[I+{}_{Ri}E:\left({}_{Ri}C_i-{}_{Ri}C\right)\right]^{-1}\right\rangle_{RVE}^{-1} \quad (5)$$

$$_{Ri}A_i^{MT}=\left[I+{}_{Ri}E_m:\left({}_{Ri}C_i-{}_{Ri}C_m\right)\right]^{-1}:\left\langle\left[I+{}_{Ri}E_m:\left({}_{Ri}C_i-{}_{Ri}C_m\right)\right]^{-1}\right\rangle_{RVE}^{-1} \quad (6)$$

$$_{Ri}A_i^{Lielens}=\left\{(1-f)\left[{}_{Ri}A_i^{lower}\right]^{-1}+f\left[{}_{Ri}A_i^{upper}\right]^{-1}\right\}^{-1}:\left\langle\left\{(1-f)\left[{}_{Ri}A_i^{lower}\right]^{-1}+f\left[{}_{Ri}A_i^{upper}\right]^{-1}\right\}^{-1}\right\rangle_{RVE}^{-1} \quad (7)$$

E is the Morris' tensor [9], which represents the interaction between an inclusion with a given morphology and the homogeneous equivalent medium. In the case of an ellipsoidal inclusion whose principal axes lengths are $\{2a_1,2a_2,2a_3\}$, it is written in the coordinate system of the inclusion Ri as follows:

$$_{Ri}E_{ijkl}=\frac{1}{4\pi}\int_0^{\pi}sin\theta d\theta\int_0^{2\pi}\gamma_{ikjl}d\phi={}_{Ri}S_{ijkl}^{Esh}\ {}_{Ri}C_{ijkl}^{-1} \quad (8)$$

where $\gamma_{ikjl}=K_{ik}^{-1}(\xi)\xi_j\xi_l$, $K_{jp}(\xi)={}_{Ri}C_{ijpl}\xi_i\xi_l$, $\xi_1=\dfrac{sin\,\theta\,cos\,\phi}{a_1}$, $\xi_2=\dfrac{sin\,\theta\,sin\,\phi}{a_2}$, $\xi_3=\dfrac{cos\,\theta}{a_3}$.

S^{Esh} is the Eshelby tensor; a_1, a_2, a_3 are used to describe the inclusion shape. In the Mori-

Tanaka model (equation (6)), the Morris' tensor E_m is computed by using the matrix properties C_m as the infinite media, instead of C. In equation (7) related to Lielens' model, f is an interpolating factor which depends on the inclusions volume fraction [6,10]. For a set of inclusions with the same shape (ellipsoid, sphere,…) having a volume fraction f_{shape}, the interpolating factor is given by:

$$f = \frac{1}{2} f_{shape}\left(1 + f_{shape}\right) \tag{9}$$

In our study, for the RVEs exhibiting inclusions with two different shapes (ellipsoid and sphere), an interpolating factor is considered for each shape according to their volume fraction. A_i^{lower} and A_i^{upper} in equation (7) are given by:

$$_{Ri}A_i^{lower} = \left[I + {}_{Ri}E_m : \left({}_{Ri}C_i - {}_{Ri}C_m\right)\right]^{-1} \tag{10}$$

$$_{Ri}A_i^{upper} = \left[I + {}_{Ri}E_i : \left({}_{Ri}C_m - {}_{Ri}C_i\right)\right]^{-1} \tag{11}$$

Note that in these relations, the Morris' tensors E_m and E_i are computed by using the matrix and the inclusion properties respectively as the infinite media.

4 RESULTS AND DISCUSSION

In this part, several RVEs exhibiting different morphologies have been studied. The accuracy of homogenization models was evaluated for composites made of an isotropic matrix reinforced with isotropic spherical and/or ellipsoidal particles. Effective properties predicted by mean-field homogenization analytical models have then been compared to those obtained numerically (by FFT-based homogenization technique) in order to rigorously validate the investigated models and to highlight the influence of the inclusion volume fraction on their accuracy and their sensitivity to the inclusions morphology. The effective bulk, shear and Young's moduli were computed for each RVE using the corresponding homogenized stiffness tensor C. For the sake of simplicity, only the effective shear μ_{eff} and bulk κ_{eff} moduli were presented in this paper. In the following, the Poisson ratio is assumed to be ν = 1/3 for all phases (inclusions and matrix).

4.1 Influence of spherical inclusions on the accuracy of the models

Many works were conducted to highlight the validity domains of these mean-field micromechanical models (by comparing them to full-field approaches) when predicting the elastic properties of composites reinforced with spherical particles [11,12]. Consequently, our study is not concerned with this kind of RVEs. However, some simulations were made in order to confirm if our results are in accordance with theirs. For inclusion volume fraction of about 10%, predictions of μ_{eff} and κ_{eff} by the mean-field approaches are almost satisfactory. Around 30% of inclusions, mean-field models are accurate for low contrast and deviate from the FFTb solution for high contrasts. When the inclusions volume fraction reaches 50%, the same behaviour was observed. Figure 2 shows the normalized effective shear μ_{eff} and bulk κ_{eff} moduli evolutions as function of the Young's modulus contrast E_i/E_m (ratio between the Young's modulus E_i of the inclusion and E_m of the matrix) in this case. The predictions of Mori-Tanaka and normalized self-consistent models diverge very rapidly when the contrasts increase, as observed in [11]. The Mori-Tanaka model underestimates the accurate solution (supposed to be FFTb results) while the normalized self-consistent model overestimates it. For low contrasts, (up to 50 for the bulk modulus and up to 20 for the shear modulus), Lielens' model is close to the FFTb, with a relative deviation from the FFTb less than 10%.

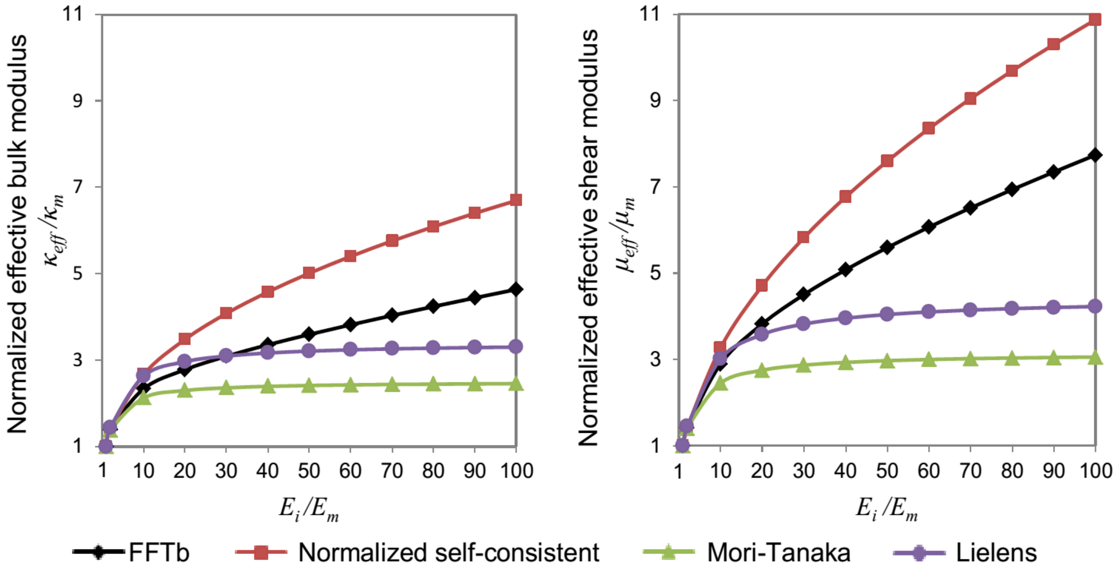


Figure 2: Normalized effective shear and bulk moduli as function of the Young's modulus contrast for a RVE consisting of spherical inclusions with a volume fraction of 50%.

4.2 Influence of ellipsoidal inclusions on the accuracy of the models

The aim of this part is to highlight the accuracy of the mean-field approaches while predicting the mechanical behaviour of these kinds of composites made of randomly oriented isotropic ellipsoidal inclusions distributed into an isotropic matrix. To do this, six RVEs containing respectively 4, 8, 12, 16, 20 and 30% (in volume) of ellipsoidal inclusions have been investigated. Each inclusion contains 10 ellipsoidal inclusions randomly distributed. This distribution differs from one RVE to another. For this study, the aspect ratio of the ellipsoids is taken equal to 5 ($a_1/a_2 = 5$, $a_2 = a_3$) for all inclusions.

Using the RVEs' parameters given by the RSA and MD-based generation algorithms, we computed the homogenized properties of each RVE. The figure 3 shows the evolution of the normalized effective shear modulus as function of the Young's modulus contrast for some of the RVEs investigated. The effective bulk modulus results are not showed for the sake of clarity and especially because the relative deviations from the FFTb observed in this case are less than those obtained for the effective shear modulus. In the case of high volume fraction of ellipsoidal inclusions, the normalized self-consistent model cannot be reasonably used. Actually, in that case, the computation time increases drastically and the algorithm involves a high memory consumption due to the iterative resolution. For all these reasons, this model was not presented in this part. Figure 3 shows that Lielens' model is close to FFTb. The mean-field analytical models are more sensitive for ellipsoidal inclusions' volume fraction. Up to about 20% of inclusions, all the models deliver accurate estimates of the homogenized properties regardless of contrasts. The relative deviations from the FFTb model observed in this case do not exceed 4% for Lielens' model and 6% for Mori-Tanaka model. For a volume fraction of 30%, the models deliver accurate estimates only for low contrasts ($E_i / E_m \leq 20$).

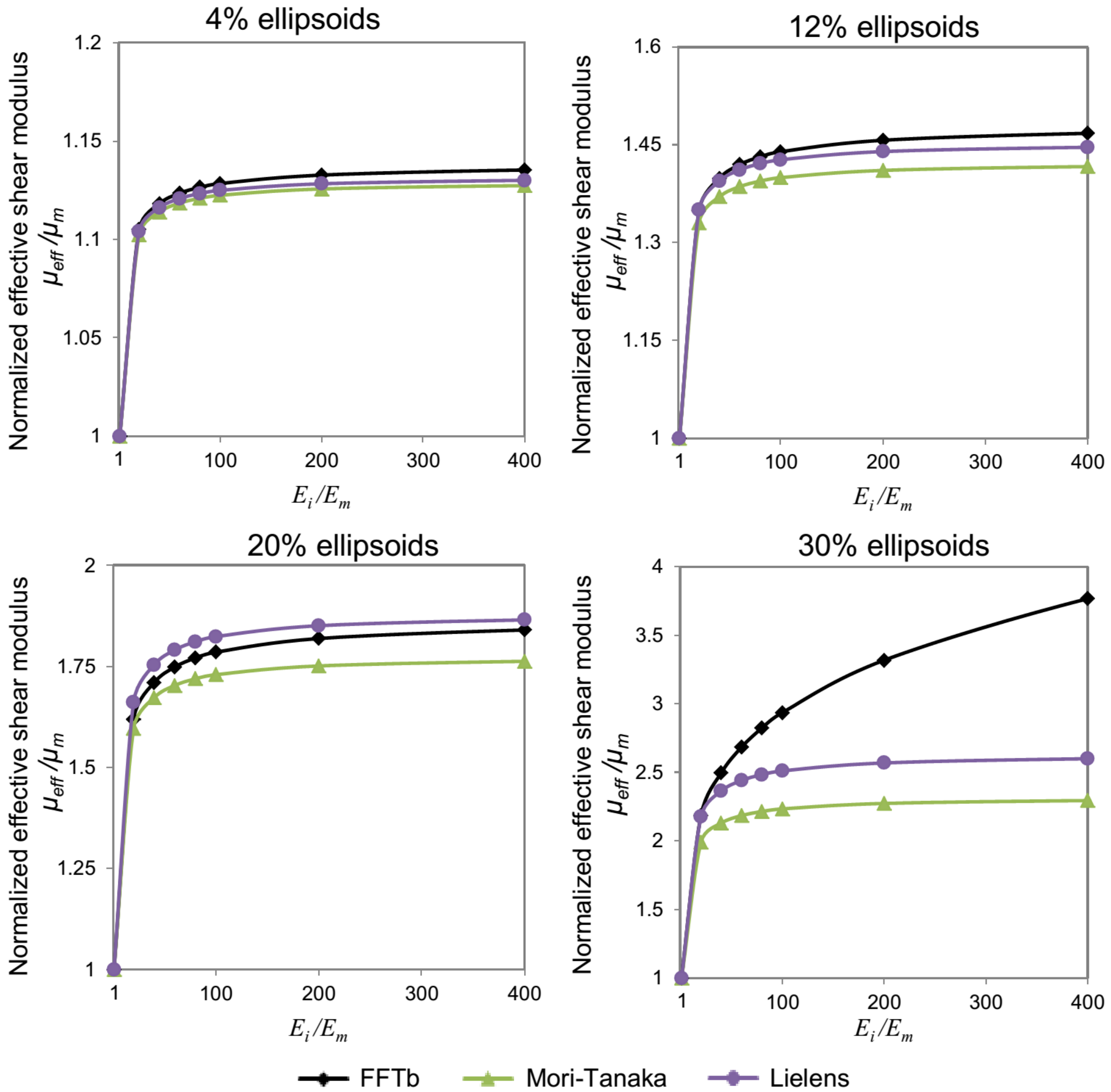


Figure 3: Normalized effective shear modulus as function of the Young's modulus contrast for RVEs consisting of ellipsoidal inclusions with a volume fraction of 4, 12, 20 and 30%, respectively. In each RVE, 10 ellipsoidal inclusions exhibiting different orientations were considered. The aspect ratio of the ellipsoids is a_1/a_2=5.

4.3 Influence of a mixture of spherical an ellipsoidal inclusions on the accuracy of the models

Now, we consider two RVEs containing a mixture of spherical and ellipsoidal inclusions. For the two RVEs, the volume fraction of the spherical and ellipsoidal inclusions is set at 5% and 6.7% respectively. For one RVE, the ellipsoids were oriented in only 2 directions different from the main directions of the RVE. It is clear that the macroscopic behaviour will be strictly anisotropic in this case. The aspect ratio of the ellipsoids is fixed to 5. For the other RVE, 10 different geometrical orientations were considered for the ellipsoidal inclusions. The aspect ratio of the ellipsoids for this RVE is set at 10. Figure 4 shows the normalized effective shear and bulk moduli evolutions related to the contrast of these RVEs. The models are in good agreement for the shear and bulk moduli in each case. Figure 4 shows that Lielens and Mori-Tanaka models deliver, for the volume fractions investigated, the most accurate predictions when the RVE contains a mixture of spherical and ellipsoidal inclusions, regardless of the macroscopic behaviour being isotropic or not. Although the normalized self-consistent model deviates from the other models, the maximal relative deviation from the FFTb results observed for this later do not exceed 5%. One can also notice that the mean-field models are sensitive to the ellipsoids aspect ratio because the discrepancies observed are more important in the RVE where the ellipsoids exhibits the great aspect ratio.

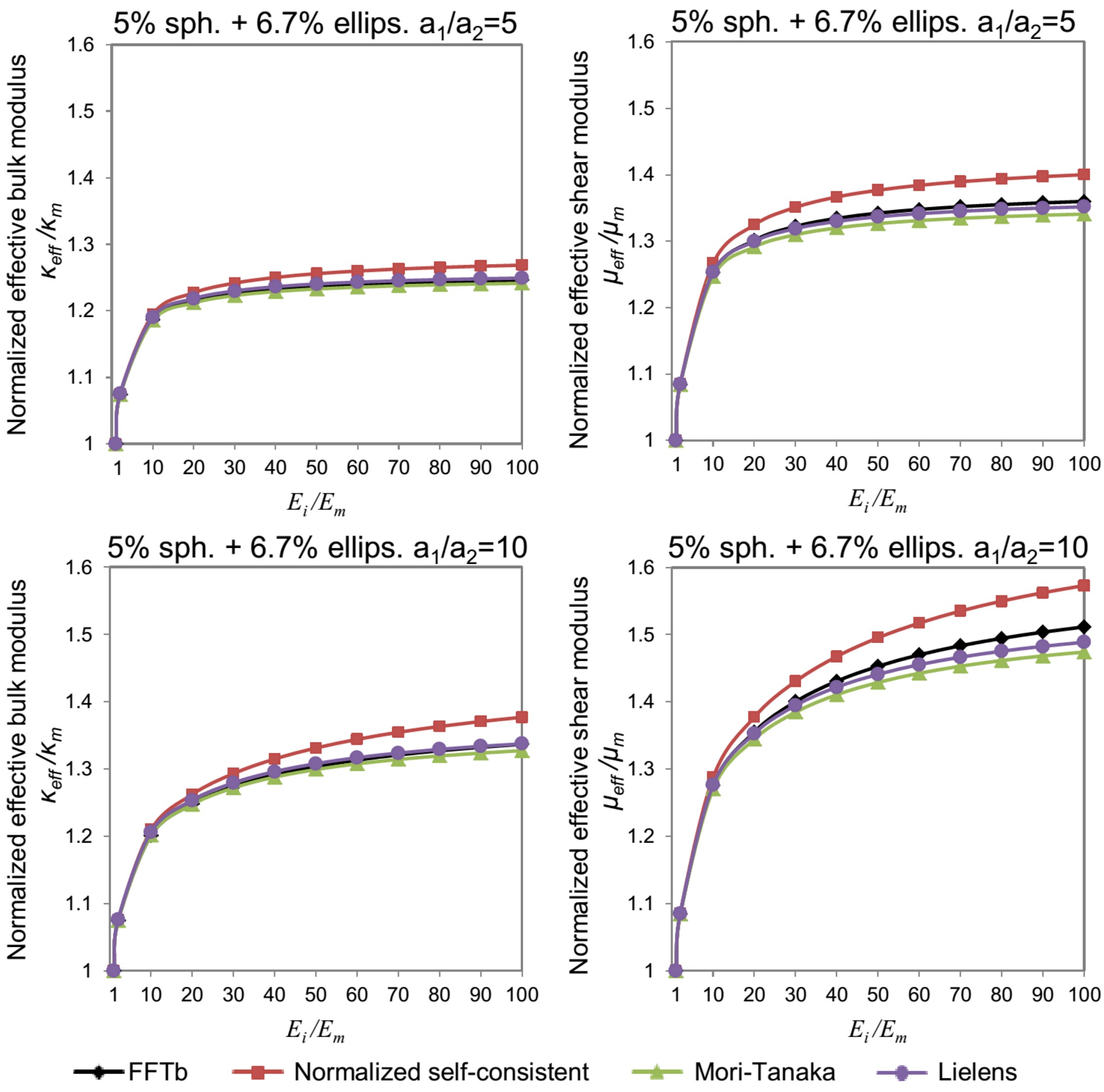


Figure 4: Normalized effective shear and bulk moduli as function of the Young's modulus contrast for 2 RVEs consisting of a mixture of spherical (5%) and ellipsoidal (6.7%) inclusions. The ellipsoidal inclusions ($a_1/a_2=5$) of the first RVE exhibit 2 different geometrical orientations. In the second RVE, the ellipsoidal inclusions ($a_1/a_2=10$) exhibit 10 different geometrical orientations.

5 CONCLUSIONS

The study highlights the influence of the inclusions morphology on the accuracy and discrepancy of some classical mean-field approximation methods and a full-field computational method based on numerical homogenization techniques to predict the mechanical properties of these materials. Several Representative Volume Elements containing spherical, ellipsoidal inclusions and a mixture of both were studied. For low volume fractions of inclusions, the results of the mean-field approximations and those obtained with the Fast Fourier Transform-based (FFTb) full-field computation are very close regardless to the inclusions morphology.

RVEs consisting of ellipsoidal or a mixture of ellipsoidal and spherical inclusions, when the inclusions volume fraction becomes higher, one observes that Lielens' model and the FFTb full-field computation give almost similar estimates. The sensitivity of the computational methods, consequently their accuracy depends on the shape of the inclusions and their volume fraction. The ellipsoids aspect ratio has also some influence on the accuracy of the models but this one is less than the influence of the volume fraction. For microstructures with a mixture of ellipsoidal and spherical inclusions, Lielens' and Mori-Tanaka models could be a good alternative to the FFTb model when the total inclusions vol-ume fraction is about 12%. In this case, the normalized self-consistent model could also be an alternative to the FFTb model with an error less than 5%.

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