PREDICTION OF EFFECTIVE THERMAL CONDUCTIVITY OF HETEROGENEOUS RANDOM MULTI-PHASE COMPOSITES

Kamel FEDAOUI[1], Salah MADANI[2], Toufik KANIT[3]

The numerical homogenization technique is used in order to compute the effective thermal conductivity ETC of heterogeneous random multi-phase composites. Two types of microstructures are considered: microstructure with random distribution of identical non-overlapping 3-phases inclusions and microstructure with non-overlapping 4-phases inclusions, based on the Poisson process. Two boundary conditions are applied on the representative volume element, RVE, of microstructures, for thermal modeling by Finite Element Method. The aim of the work was to examine how spatial distribution and particles volume fraction influences the ETC of the composite material. The results were compared to various analytical models.

Keywords: multi-phase heterogeneous composite, thermal conductivity, numerical simulation.

1. Introduction

The growing demand for smaller, lighter and faster machines and electronics has created a need for new materials. In addition, industry has a growing need to adapt the properties of materials, including thermal conductivity, to desired applications. Composites are often used to fill these needs. Composites are a mixture of two or more types of materials (polymer, metal, ceramic, etc.) that form a new material with properties that are a combination of the components. Many study about the determination and the prediction of thermo-elastic properties were developed and published in the case of heterogeneous material [1, 2, and 3]. Conductive composites are often formed by the addition of thermally conductive particles to a matrix [4]. In the early years, analytical estimates for the effective thermal properties were developed and published in the case of composite material. For example, Progelhof [5] give a review on methods for predicting the thermal conductivity of heterogeneous materials. Woodside [6] and Dobson [7] developed two analytical models for multiphase material. Numerical homogenization technic is widely used in the analysis of composite

[1] Phd., Laboratoire de Mécanique de Structures & Matériaux, LAMSM, Université de Batna 2, Algeria, e-mail: fedaouik@yahoo.fr
[2] Prof., Laboratoire de Mécanique de Structures & Matériaux, LAMSM, Université de Batna 2, Algeria, e-mail: pr.madani.salah@gmail.com
[3] Prof., Laboratoire de Mécanique de Structures & Matériaux, LAMSM, Université de Batna 2, Algeria, e-mail: pr.kanit.salah@gmail.com

materials in order to obtain the effective thermal properties. Many studies have investigated the effect of the addition of single type of particles to increase the thermal conductivity of matrix [8, 9]. The effects of shape, orientation and distribution of the inclusion pores on the conductivity were studied [10-14]. Various conductive particles can be used to increase composites thermal conductivities. Numerous numerical and theoretical predictions have been proposed in the literature to study the thermal properties of composite materials [15, 16]. The problem for most of analytical models is the estimation of thermophysical properties based on the knowledge of particles volume fraction and properties of each component of the mixture only, with possible shape factor and thermal contact resistance at the matrix-inclusions interface. They are not generally well suited for highly loaded composite materials and for randomly distributed inclusions. The influence of particles' size and volume fraction was taken into account in numerical simulations. The results of composites ETC were analysed and compared with various analytical models.

2. Computational thermal homogenization

In this section, all elements and notations of numerical homogenization necessary to determine the effective thermal conductivity, using the methodology explained by [16] based on the FEM, are carried out.

2.1. Microstructures generating and thermal conductivity of phases

This work concerns the prediction of *ETC* of multi-phases composite materials by *3D* numerical simulation. Simulations are performed using finite element method coupled with a homogenization method. The generation algorithm of random position of particles is developed under Mathematica. Matlab code use the generated position to generate an input file for the Comsol code, see figure 1. To simplify the calculations, simulations were performed for a Representative Volume Element (RVE) randomly filled with spherical particles. The algorithm generates the positions of the inclusion for a specified volume fraction, in a square region (matrix). It ensures that the particle do not intersect by judging whether the newly generated particle intersects the particles that have been created. If there is no intersection, it means that the newly generated particle satisfies the condition and is accepted, and on the contrary, is rejected. The automated generation process will not stop until the volume fraction of particles is satisfied. The morphology and technique of *3D* microstructures generating is presented in this section. For each studied microstructure, many configurations with different populations of inclusion are investigated. Each microstructure contains one population of non-overlapped inclusions, randomly distributed in a continuous matrix. It should be noted that there is no contact between neighboring inclusions of the dispersed phase. Fig. 2 shows an example of different used microstructures containing a random distribution and random orientation of inclusions for different volume fractions. Different thermal conductivities are attributed to the inclusions phases, in order to predict the effective thermal conductivity of composites. Table 1 represents the thermal properties of each phase used for numerical computations. ϕ_i, R_i are respectively the volume fraction and the size of the i th phase. ϕ is the volume fraction of the polymer matrix. Table 1 represents the thermal properties of each phase used for numerical computations.

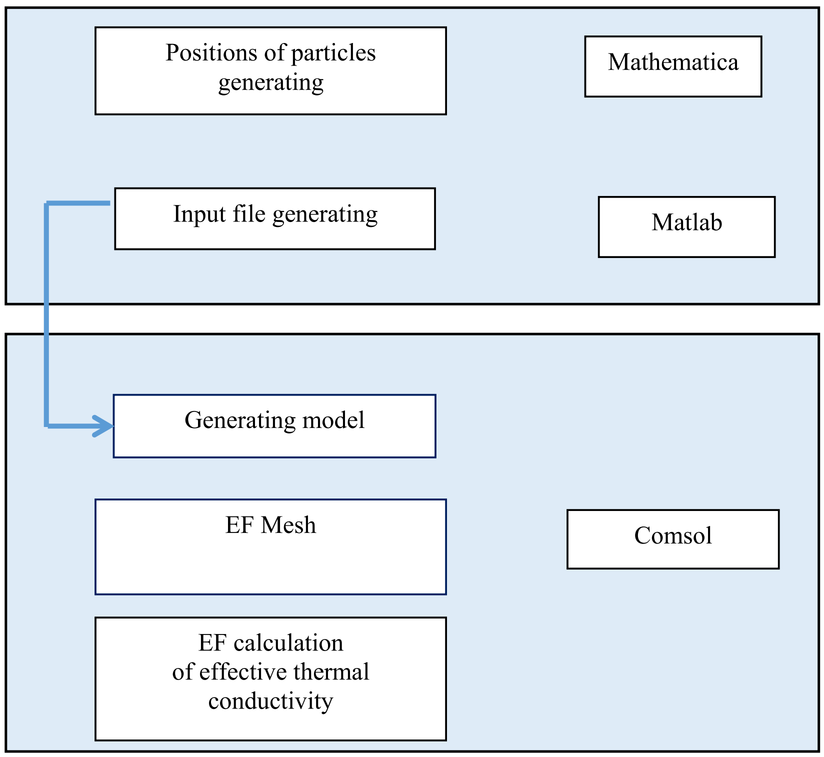


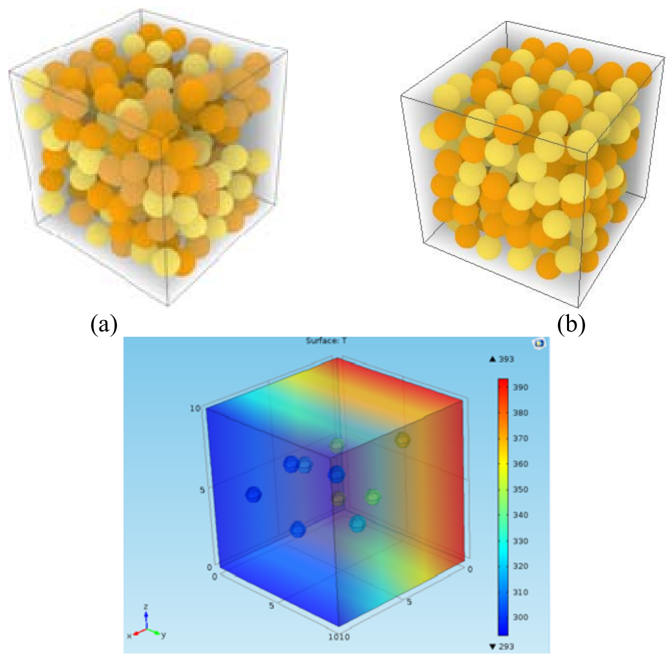
Fig 1: Process of generating model and meshing technic.

Table1.
Cases of composites used for numerical computations.

Material	Case	Properties
3-Phase composite	Case 1	$\phi_1 = \phi_2 = 5\%, R_1 = R_2$
	Case 2	$\phi_1 = \phi_2 = 15\%, R_1 = R_2$
	Case 3	$\phi_1 = \phi_2 = 25\%, R_1 = R_2$
	Case 4	$\phi_1 = \phi_2 = 15\%, R_1 = 2R_2$
4-Phase composite	Case 5	$\phi_1 = \phi_2 = \phi_3 = 10\%, R_1 = R_2 = R_3$

Table2.
Thermal properties of each phase used for numerical computations.

Phase	Matrix	Inclusion 1	Inclusion 2	Inclusion 3
λ Thermal conductivity [W/m.K]	30	10	20	1



(a) (b)

(c)

Fig. 2. Example of material used in this investigation, (a) 3-phase material, (b) 4-phase material, (c) temperature map for the 3-phase material with 10 inclusions.

2.2. Mesh generation and mesh density

A regular 3D finite element mesh is superimposed on the image of the microstructure using the so-called multi-phase element technique. This technique was developed by [17] and used for the homogenization of virtual or real images by several authors as [16, 18, 19, 20]. As a result of convergence, a good mesh density of quadrangular finite elements was adopted in all this investigation for all simulations.

2.3. Boundary conditions

For the thermal problem, the temperature, its gradient and the heat flux vector are denoted by T, ∇T and $\underline{q}$ respectively. The heat flux vector and the temperature gradient are related by Fourier’s law, that reads:

$$\underline{q} = -\lambda \nabla T \tag{1}$$

In the isotropic case, the scalar λ is the thermal conductivity coefficient of the considered phase. Two types of boundary conditions are used in this paper. For mathematical expressions of these boundary conditions, see [16].

- Uniform gradient of temperature at the boundary (UGT) :

$T = \underline{G} \cdot \underline{x}, \forall \underline{x} \in \partial V$ .Where $\underline{G}$ is a constant vector independent of $\underline{x}$ and ∂V the outside of the volume.

- Periodic boundary conditions (PBC) : The temperature field takes the form : $T = \underline{G} \cdot \underline{x} + t, \forall \underline{x} \in \partial V$. The fluctuation temperature t is periodic.

We note here that the periodic boundary conditions PBC converge faster in terms of RVE size, to the mean overall effective thermal conductivity by [16].

3. Theoretical models

A large number of theoretical models for prediction of effective properties for multiphase materials have been developed, mainly because of the significance and interest in the effective thermal conductivity.

3.1 model of Woodside and Messmer

For three-phase media, Woodside and Messmer [6] proposed the 'quadratic parallel' (QP) model for the effective conductivity:

$$\lambda_e = \left(\lambda_1^{1/2}\phi_1 + \lambda_2^{1/2}\phi_2 + \lambda_3^{1/2}\phi_3\right)^2 \tag{2}$$

where λ_i with $i = \{1, 2, 3\}$ is the conductivity of each phase and ϕ_i the corresponding volume fraction. This model appears to be applicable when $i > 3$. Various weighted average models have also been proposed for such multiphase mixtures.

3.2 model of Dobson

The Maxwell models are the most suitable to developing such mixture models in general, and the Maxwell–De Loor model in particular is widely used, for it requires no geometrical parameters. Dobson et al. [7] rewrote this model for a four-phase system into:

$$\lambda_e = \frac{3\lambda_1 + 2(\phi_2 - \phi_3)(\lambda_2 - \lambda_1) + 2\phi_3(\lambda_3 - \lambda_1) + 2(\phi_4 - \phi_2)(\lambda_4 - \lambda_1)}{3 + (\phi_2 - \phi_3)\left(\frac{\lambda_1}{\lambda_2} - 1\right) + \phi_3\left(\frac{\lambda_1}{\lambda_3} - 1\right) + (\phi_4 - \phi_2)\left(\frac{\lambda_1}{\lambda_4} - 1\right)} \quad (3)$$

4. Results and discussion

In this part, for the determination of the thermal conductivity of heterogeneous materials, numerical technique with RVE notion developed by [16] is used. It consists in considering different realizations of random microstructures in order to obtain the effective properties. The RVE is the volume that allows the estimation of the effective property with one realization.

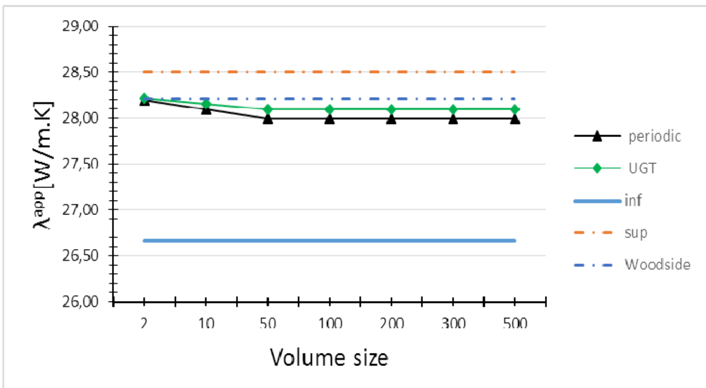


Fig 3. Variation of the thermal conductivity λ^{app} for case 1 in terms of volume size for 3-phases material

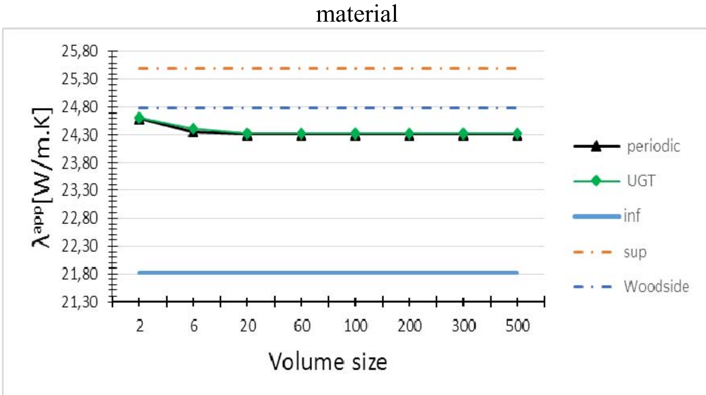


Fig 4. Variation of the thermal conductivity λ^{app} for case 2 in terms of volume size for 3-phases material

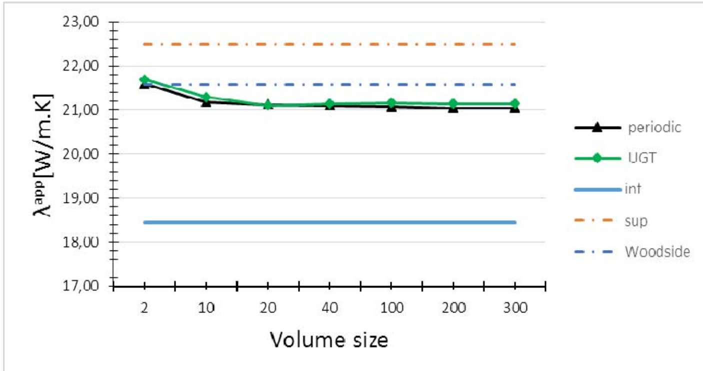


Fig 5. Variation of the thermal conductivity λ^{app} for case 3 in terms of volume size for 3-phases material

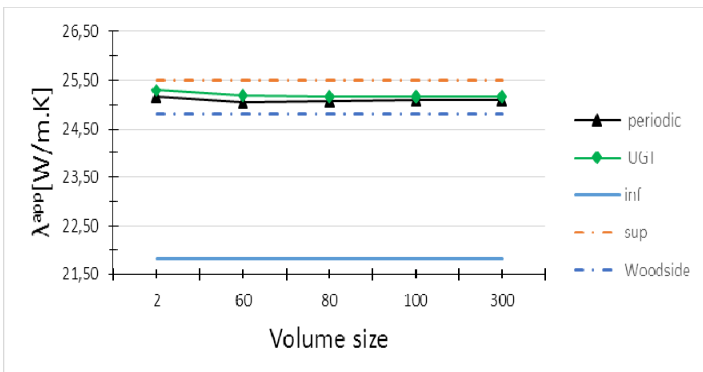


Fig 6. Variation of the thermal conductivity λ^{app} for case 4 in terms of volume size for 3-phases material

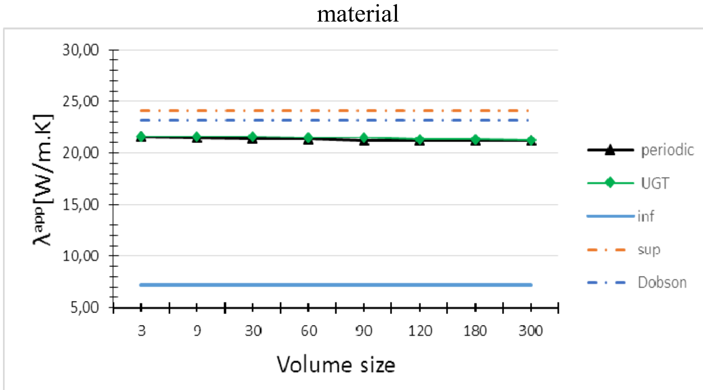


Fig 7. Variation of the thermal conductivity λ^{app} for case 5 in terms of volume size for 4-phases material

4.1 Determination of the RVE

For numerical simulations, two boundary conditions (UGT and PBC) are applied on each volume containing N inclusions. The found results are presented on Figures. 3–7.It appears that the dispersion of the results decreases when the size of the volume increases. The RVE size is obtained when the mean values of two boundary conditions used coincided and give the same value of the thermal conductivity, see table 3.

Table3.
RVE size from numerical computations.

Material	Ase	Properties	Rve
3-Phase composite	Case 1	$\phi_1 = \phi_2 = 5\%, R_1 = R_2$	50
	Case 2	$\phi_1 = \phi_2 = 15\%, R_1 = R_2$	20
	Case 3	$\phi_1 = \phi_2 = 25\%, R_1 = R_2$	30
	Case 4	$\phi_1 = \phi_2 = 15\%, R_1 = 2R_2$	50
4-Phase composite	Case 5	$\phi_1 = \phi_2 = \phi_3 = 10\%, R_1 = R_2 = R_3$	60

4.2 Effect of boundary conditions on thermal conductivity

It can be observed from figures 3-7 that the results obtained given by PBC and UGT fluctuates slightly as a function of the microstructure size. According to [20], PBC leads to rapid convergence, in terms of effective physical properties, than by applying other boundary conditions. An important bias is found in the results provided with small volume sizes, the value being different from the effective one obtained for large volume. El Moumen in [21], obtains the same results for mechanical properties. Kaddouri in [22], obtain the same results for physical properties. For small volumes, the average moduli obtained by simulations depend on the boundary conditions: UGT produces results over numerical prediction of PBC. It appears also that for sufficiently large sizes, the results obtained with PBC do not depend on the size of microstructures.

4.3 Effect of volume fraction of particle on thermal conductivity

Many authors have studied the effect of volume fraction on thermal conductivity of composite materials. In order to investigate the effect of volume fraction of inclusion on the effective thermal conductivity, different volumes fractions of inclusions were investigated, the effective thermal conductivity increase with the increase of the volume fraction of inclusions, see figure 8.

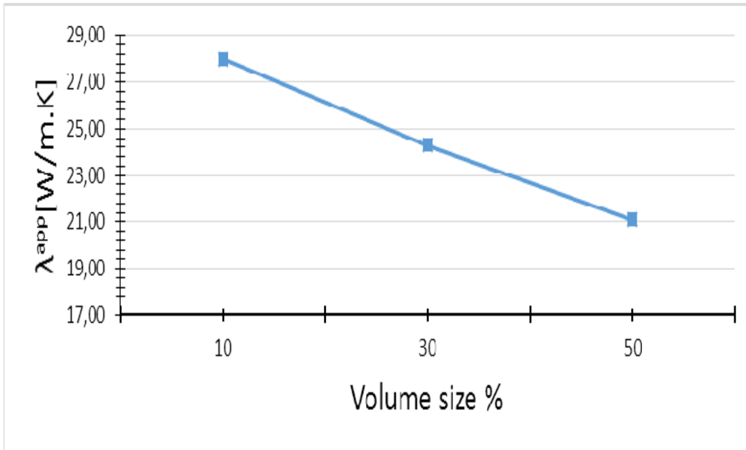


Fig 8. Variation of the thermal conductivity λ^{app} in terms of volume fraction for 3-phases material.

5. Conclusions

To use the composites, their physical properties and performances should be well identified. One of the important properties of the composites is *ETC*. Since the thermal conductivity of the composites depends on several parameters, it is not very easy to predict thermal properties exactly. In this paper, finite element method coupled with a script for creating RVE was very beneficial to predict effective thermal conductivity of composites reinforced with randomly distributed inclusions. The computational thermal homogenization is based on a *3D* random material with spherical inclusions. The Poisson process was used to generate the random particle dispersions. Two types of microstructures are considered: microstructure 1 with 3-phases identical non-overlapping inclusions, and microstructure 2 with 4-phases identical non-overlapping inclusions. Two types of boundary conditions are tested here. Several volumes, named realizations, are considered. These realizations are used to determine the effect of some element on the *ETC* of the microstructure. The effective properties and the size of the representative volume element (RVE) are related with all microstructure parameters. The variation of the RVE as a function of the volume size was studied.

R E F E R E N C E S

[1]. *D. M. Bigg*, Thermally conductive polymer compositions. Polym Compos; 7: 125, 1986.

[2]. *P. Koudelka, T. Doktor, J. Valach, D. Kytyr and O. Jirousek*, Effective elastic moduli of closed-cell aluminium foams - homogenization method, U.P.B. Sci. Bull., Series D,75: Iss. 1, 2013.

[3]. *I. Petrescu, C. Mohora, C. Ispas*, the determination of young modulus for cfrp using three point bending tests at different span lengths. U.P.B. Sci. Bull., Series D, 75: Iss. 1, 2013.

[4]. Y. *Agari, A. Ueda and S. Nagai*, Thermal conductivities of composites in several types of dispersion systems. J of Appl Polym Sci; 42: 1655–1669, 1991.
[5]. *R.C Progelhof, JL Throne and RR. Reutsch*, Methods for predicting the thermal conductivity of composite systems: a review. Polym Eng Sci 1976; 16: 615–625.
[6]. *W. Woodside, J.H. Messmer*, Journal of Applied Physics 32 (1961) 1688.
[7]. *M.C. Dobson, F.T. Ulab y, M.T. Hallikainen, M.A. Elrayes*, IEEE Transactions onGeoscience and Remote Sensing 23 35–46, 1985.
[8]. *M. Karkri A. Boudenne and L Ibos*, Numerical and experimental study of thermophysical properties of spheres composite materials. Int J High Temp High Pressures; 40: 61–81, 2011.
[9]. Y. *Agari and T. Uno*, Estimation on thermal conductivities of filled polymers. J Appl Polym Sci;2:5705–5712, 1986.
[10]. J. *Floury, J. Carson and Q Tuan Pham*, Modelling thermal conductivity in heterogeneous media with the finite element method. Food Bioprocess Technol; 1: 161–170, 2008.
[11]. B. *Mortazavi M. Baniassadi J. Bardon*. Modeling of two-phase random composite materials by finite element, Mori–Tanaka and strong contrast methods. Composites Part B 2013; 45: 1117–1125.
[12]. *X. Lu and G. Xu* . Thermally conductive polymer composites for electronic packaging. J Appl Polym Sci; 65: 2733, 1997.
[13]. A. *Moussa, K. Mustapha, I. Laurent and EM Mimoun*, Effective thermal conductivity of random two-phase composites, Journal of Reinforced Plastics and Composites, Vol 33(1) 69–80, 2014.
[14]. *C. Mourad A. Boudjemaa,H. Manel and B. Abderrahim*, Numerical modelling of the effective thermal conductivity of heterogeneous materials, Journal of Thermoplastic Composite Materials 26(3) 336–345, 2011.
[15]. *W. Moran and P. Ning*, Predictions of effective physical properties of complex multiphase materials, Materials Science and Engineering R 63 1–30, 2008.
[16]. *T. Kanit, S. Forest, I. Galliet, V. Mounoury, D. Jeulin*, Determination of the size of the representative volume element for random composites, statistical and numerical approach .Int. J. Solids Struct.40, 3647–3679(2003).
[17]. *N. Lippmann,* Comput. Mater. Sci. 9 (1997) 28–35.
[18]. *A. El Moumen, T., Kanit, A. Imad,, H. ElMinor*. Computational thermal conductivity in porous materials using homogenization techniques: numerical and statistical approaches. Comput. Mater. Sci. 97, 148–158, 2015.
[19]. *A. El Moumen, T. Kanit , A. Imad, H. El Minor,* Effect of reinforcement shape on physical properties and representative volume element of particles reinforced composites: statistical and numerical approaches, Mech Mater.83,1–16, 2015.
[20]. *S. Forest,* Milieux continus généralisés et matériaux hétérogènes. Mines, Paris. 2006.
[21]. *A. El Moumen, T. Kanit , A. Imad, E. Hilali, and H. El Minor,* A multiscale approach and microstructure design of the elastic composite behavior reinforced with natural particles .Compos. Part B: Eng. 66, 247–254, 2014.
[22]. *W. Kaddouri, A. El Moumen, T. Kanita, S. Madani and A.Imad*, On the effect of inclusion shape on effective thermal conductivity of heterogeneous materials, Mechanics of Materials 92 28–41(2016).