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Predicting the thermal conductivity of composite materials with imperfect interfaces

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This paper compares the predicted values of the thermal conductivity of a composite made using the equivalent inclusion method (EIM) and the finite element method (FEM) using representative volume elements. The effects of inclusion anisotropy, inclusion orientation distribution, thermal interface conductance, \( h \), and inclusion dimensions have been considered. Both methods predict similar overall behaviour, whereby at high \( h \) values, the effective thermal conductivity of the composite is limited by the inclusion anisotropy, while at lower \( h \) values, the effect of anisotropy is greatly diminished due to the more dominant effect of limited heat flow across the inclusion/matrix interface. The simulation results are then used to understand why in those cases where it has been possible to produce CNF reinforced Cu matrix composites with a large volume fraction of well dispersed CNFs, the measured thermal properties of the composite have failed to meet the expectations in terms of thermal conductivity, with measured conductivities in the range 200-300 W/m·K. The simulation results show that, although degradation of the thermal properties of the CNFs and a poor interfacial
thermal conductance are very likely the reasons behind the low conductivities reported, great care should be taken when measuring the thermal conductivity of this new class of materials, to avoid misleading results due to anisotropic effects.

**Keywords:** C. FEA, B. Interface, A. Short-fibre composites, A. Carbon nanotubes

1. Introduction

With the increase in computing capacity of modern computers, brute force characterization of heterogeneous materials is becoming more and more accessible. What could only be done with simple or highly symmetrical models, especially in 2D, only a couple of decades ago, is now feasible even for home computers. The key concept of this progress is the representative volume element (RVE) [1-3], which defines how large a representative cell must be to give properties of a composite material under a given error. This is the case of the finite element method (FEM), which is based on dividing the geometry under study in a large number of discrete elements. Both, the number of elements and the size of the model, which depends on the number of inclusions and the volume fraction, are the main factors affecting the computational power required to run the simulations. With the currently available computer power, it is now possible to simulate sufficiently large RVEs to study the properties of new metal matrix composites reinforced with carbon nanofibres (CNF). These are a new class of engineering materials that offer opportunities to tailor properties and meet specific requirements, e.g. in heat sinks. For instance, CNF reinforced copper composites constitute an ideal candidate to work as an electrical contact material and/or substrate for semiconductor devices due to their potentially high electrical and thermal
conductivity, small coefficient of thermal expansion, good machinability and low price. Processing these materials is challenging due to the poor wetting between copper and carbon, which makes very difficult the fabrication of dense composites with homogeneously distributed fibres. However, even in those cases where the experimental difficulties have been overcome and a large volume fraction of well dispersed CNFs has been obtained, the measured thermal properties of the composite have failed to meet the expectations in terms of thermal conductivity [4-5]. There are two possible reasons for this: (1) the highly anisotropic properties of CNFs combined with a preferred orientation of the fibres during processing and (2) the interfacial contact resistance of the Cu-C interface limiting the potential benefit of incorporating these inclusions into the matrix.

In this context, the objective of this study has been to use a range of modelling methods to evaluate the effect of fibre anisotropy and interfacial thermal contact resistance into the global thermal properties of a CNF reinforced Cu matrix composite. The modelling method employed has been the FEM of a RVE. Despite the considerable computer power required, the advantage of this approach is that it allows the study of the local fields, which cannot be accomplished by homogenization methods, and that the generated RVE can also be used to study other thermomechanical properties, such as the thermal expansion coefficient or the plastic deformation of the composite. The FEM results, when possible, have been contrasted with results obtained through different homogenization methods such as two different equivalent inclusion methods (EIM), the mean field approach (MFA) and, to some extent, the differential effective medium (DEM) approach. Comparisons have been made for cylindrical and spherical inclusions, while various parameters, such as volume fraction or inclusion conductivity, have been
varied in order to understand the origin of the discrepancies observed between all the methods.

The paper is organized as follows. First, spherical inclusions are analyzed and values of the three methods are compared in two parametric studies, one varying the volume fraction and the other one varying the interfacial conductance. A further comparison is made between FEM and MFA for different inclusion sizes and thermal conductivity differences between phases. This is followed by two other studies for the case of cylindrical inclusions. FEM and MFA are compared for long and short fibres with low and intermediate volume fractions and different values for the interfacial conductance. Finally, FEM is used to carry out a more complete parametric study for short fibre composites as a function of the anisotropy of the inclusion and the interface conductance.

2. Model description

All RVE considered were cubes of periodic geometry. That is, it was assumed that the composite microstructure was given by an infinite translation of this RVE along three axes to eliminate boundary effects and, thus, the fibre positions within the RVE kept this periodicity condition. This means that inclusions cut by a face of the cube reappear in the opposite face with the rest of its volume (see fig. 1). The numerical analyses of the RVE were carried out using the finite element method (Abaqus® [6]). To this end, the prismatic RVE (matrix and inclusions) were meshed using tetrahedral elements. Finally, periodic boundary conditions, which have been demonstrated to improve the accuracy of results [3], were applied to the RVE surfaces to ensure continuity between neighbouring RVEs.
The modelling strategy to calculate the thermal conductivity of the composite in a given direction is depicted in fig. 2. The boundary condition applied is a temperature difference of 100 K in that direction. This is implemented by tying together in temperature the nodes of opposite faces defined by the same coordinates, while node couples of faces parallel to this axis have imposed a temperature difference of 0 K, as heat flux in that direction is to be avoided. Heat going through each face is measured at reference nodes (to which all the nodes of that face are tied), and the thermal conductivity is calculated using Fourier’s law. Thermal resistance at the interface was implemented through a standard interaction tool in Abaqus® [6] that, by defining a value for the thermal conductance, slows the heat transfer at the interface between phases.

As for EIM, in both the MFA and DEM techniques, the thermal interface conductance is implemented by replacing the inclusion with a non-ideal interface by an “effective” inclusion with the thermal conductivity, \( K_i^{\text{eff}} \), given by:

\[
K_i^{\text{eff}} = \frac{K_i}{1 + \frac{K_i}{K_i^{\text{eff}}/h a}}
\]  

(1)

where \( K_i \) is the inclusion thermal conductivity, \( a \) is the inclusion radius and \( h \) the interfacial thermal conductance.

The composite thermal conductivity, \( K_c \), given by MFA for a spherical inclusion is the same as that derived by Hasselman and Johnson [7]:
\[ \frac{K_c}{K_m} = \frac{\phi^{\text{eff}} (1 + 2V_f) + (2 - 2V_f)}{\phi^{\text{eff}} (1 - V_f) + (2 + V_f)} \]

where \( K_m \) is the matrix thermal conductivity, \( V_f \) is the inclusion volume fraction and \( \phi^{\text{eff}} \) is the ratio of the effective inclusion thermal conductivity and the matrix thermal conductivity \( (K_i^{\text{eff}}/K_m) \). The DEM counterpart is given by:

\[ (1-V_f) = \frac{\phi^{\text{eff}} - \frac{K_i}{K_m}}{\phi^{\text{eff}} - 1} \times \left( \frac{K_i}{K_m} \right)^{\frac{1}{2}} \]

The composite thermal conductivity can be obtained by expanding out the equation, which is cubic and is solved analytically.

For the case of spherical inclusions, diameters of 0.1, 1 and 10 μm and volume fractions of 0.2 and 0.4 have been studied. In all the cases, the conductivity of the matrix was considered to be that of copper, 385 W/m-K [8]. In the case of the inclusions, different studies range the thermal conductivity of CNFs between 10 to 3000 W/m-K but these numbers are based on theoretical predictions rather than on actual experimental measurements. To understand the role of the interfacial thermal resistance as a function of inclusion properties, isotropic spherical inclusions with different thermal conductivities have been considered as a function of the thermal conductivity of the Cu matrix. Thus, three different phase contrasts \( (c=K_i/K_m) \) of 0.1, 1 and 10 have been studied, corresponding to \( K_i \) values of 38.5, 385 and 3850 W/ m-K respectively. The length of RVEs considered was ten times the inclusion diameter which is enough to obtain representative results [9].
For the case of fibre reinforced composites, the inclusions were represented by cylinders with an aspect ratio \( l/d \) of 5, where \( l \) stands for the length of the cylinder and \( d \) stands for the diameter. Three different architectures were studied: 3DRandom (fig. 3.a), Planar Random (fig. 3.b) and Uniaxial (UA) (fig. 3.c). The volume fraction studied was 0.28 in all cases, except for the UA architecture in which an extra simulation with a volume fraction of 0.15 was also performed. The fibre length was fixed at \( l = 500 \) nm and the diameter at \( d = 100 \) nm. The RVEs considered were cubes of side \( L = 900 \) nm (1.8 times the inclusion length). Although the size of such a RVE is slightly under the recommendable RVE size according to [2], simulations obtained with different fibre distributions led to very similar results and hence the size of the RVE was considered to be representative. The results presented here are the average value of three independent realizations with the same volume fraction and different fibre distributions.

3. Model Comparison

3.1 Spherical inclusions. Comparison between FEM, MFA and DEM

Figure 4 shows the predicted thermal conductivity of a composite with spherical inclusions \( (K_i=3850 \text{ W/m-K}) \) and a perfect interface \( (h=\infty) \) as a function of inclusion volume fraction using MFA, DEM and FEM techniques. In this case the size of the inclusion is not relevant since the thermal contact at the interface is perfect. The predictions of the different models match perfectly at low volume fractions and begin to deviate at an inclusion volume fraction of 0.3. The DEM consistently predicts higher values than the MFA, where the greatest relative difference between the two (~18%) is observed at a volume fraction of 0.7. Note that the MFA technique assumes that there is
a continuous coating surrounding the inclusion. This condition is unlikely to be met at volume fractions greater than the optimum packing fraction in cubic close packed spheres, which is 0.74. Making predictions using the MFA for composites with volume fractions of 0.5 may be reaching the limit of the model as the inclusion particles begin to form a percolating network. This effect combined with meshing problems when the spacing between neighbouring inclusions is very small effectively limits the volume fraction in FEM simulations to 0.4. Up to this volume fraction it can be observed that FEM predictions are between DEM and MFA values. The values predicted by MFA will also tend to be lower than other prediction schemes as it gives a lower bound estimate [10].

The effect of the interface thermal conductance on the composite thermal conductivity can be observed in figure 5 for volume fractions of 0.2 (fig. 5a) and 0.4 (fig. 5b), again considering a $K_i$ of 3850 W/m·K. It has been discovered that this effect is always the same with the conductivity showing an asymptotic behaviour both for low and high conductances with an inflection point in the middle, that is, behaving as a sigmoid curve. Therefore, three regions can be identified: two plateaus for high and low values of $h$ and a transition zone. For low $h$ the composite behaves as a porous material whereas for high $h$ the composite conductivity corresponds to a material with a perfect interface and hence it depends on the inclusion properties. In all the figures from now on, figure 5 included, values of the thermal conductance have been chosen to show these three regions. The model studied includes spheres with a diameter of 10 μm and interface thermal conductances ranging from 1 to $10^{15}$ W/m²·K. In the transition zone with the three methods the behaviour of the composite is dominated by the interface thermal conductance. In both instances, the DEM, MFA and FEM schemes exhibits
discrepancies for the extreme values whereas very close predictions are obtained in the transition zone. For low $h$ the upper bound is predicted by FEM and the lower bound by DEM whilst for a perfect interface the upper bound is given by DEM and the lower bound by MFA, as discussed previously. The greatest difference occurs for high conductances and is more significant in the case of an inclusion volume fraction of 0.4 with a difference of 11%. Note that the transition occurs at the same range of $h$ for both volume fractions.

A more thorough comparison is made between the FEM and MFA techniques in Figure 6, where variations in the phase contrast and particle size are made. The differences in predicted values between these techniques for all cases are not significant, always below 6%. The overall trends are very similar and consistent, where an inclusion with higher thermal conductivity increases the composite thermal conductivity and the increase in the inclusion size has improved composite thermal conductivity at lower interface thermal conductance values. As previously stated, the predictions of the different schemes are almost equal in the transition zone, however, it is observed that as the phase contrast increases the maximum composite thermal conductivity is reached for higher values of $h$, that is, the extension of the transition zone increases with the phase contrast. Nevertheless, FEM and MFA predict the same extension of the transition zone for different values of the inclusion size. It is important to note that the method used to introduce the thermal interface conductance in both techniques is valid since no differences are observed in the transition zone and the discrepancies in the extreme values do not depend on the size of the reinforcement, *i.e.*, on the surface area of interface.
3.2 Cylindrical inclusions. Comparison between FEM and MFA

Table 1 compares the predicted thermal conductivities by FEM and MFA for the three possible architectures (3DRandom, Planar Random and Uniaxial) of a fibre reinforced composite for a fibre volume fraction of 0.3. It should be pointed out that while the fibres simulated by FEM had an aspect ratio $l/d=5$, the MFA simulation results correspond to a long fibre reinforced composite. Contrary to the case of spherical inclusions that was used to compare the different simulation strategies, in this case the anisotropy of the fibres was taken into account, assuming a longitudinal thermal conductivity of 3000 W/m-K and a transversal thermal conductivity of 6 W/ m-K, which are typical values for carbon nanotubes found in the literature [11]. The matrix thermal conductivity was chosen to be that of Cu, 385 W/m-K, as before. In all cases, the interfacial thermal conductance was set at a relatively high value of $10^{12}$ W/m²·K, to ensure that the behaviour lays on the inclusion dominated plateau and not on the interface dominated transition zone. The predictions of MFA and FEM match within 1-3%, except in the directions where there is a substantial fraction of fibres in the direction of the heat flow, in which case the FEM predicts lower values (12% lower in the planar case and 14% lower in the uniaxial case) than MFA. This seems to be in contradiction with the spherical inclusion results in which MFA represented a lower bound. However, this trend is related more to the length of the fibres, that were long in the case of the MFA computations and short, with an aspect ratio of $l/d=5$, for FEM and the size of the RVE simulated by FEM.

To confirm this, some MFA results were computed again for the case of a uniaxial orientation of fibres with a volume fraction of 0.15 (within the range of low volume
fractions where MFA and FEM gave identical results for spherical inclusions) and considering spheroidal inclusions, instead of long fibres. In this case, the interfacial conductance was varied from 500 W/m²-K to $10^{13}$ W/m²-K and the results are compared in figure 7. The differences between both methods are kept always below an acceptable 6%, indicating that the size of the RVE considered in FEM is large enough to capture the thermal behaviour of the composite with good precision.

4. Thermal conductivity of the composite

The results presented above show that, given enough precautions are taken to define the RVE, FEM is a reliable method to compute the thermal conductivity of metal matrix composites and is specially well suited for intermediate reinforcement volume fractions (~0.3), with the advantage on top of homogenization methods that the local fields can be studied in detail at the scale of the reinforcement. As explained in the introduction, the main objective of this work has been to use simulation tools to understand why in those cases where it has been possible to produce CNF reinforced Cu matrix composites with a large volume fraction of well dispersed CNFs, the measured thermal properties of the composite have failed to meet the expectations in terms of thermal conductivity, with measured conductivities in the range 100-300 W/m-K [4-5]. To do this, the FEM tools described above were used to carry out a complete parametric study as a function of the thermal anisotropy of the inclusion and the interface conductance to identify the most important factors limiting the real thermal conductivity of this new class of materials.

Fig. 8 represents the simulated thermal conductivity of a CNF reinforced Cu composite as a function of interface thermal conductance, assuming three different preferred
orientations of the CNFs: 3DRandom (8.a), Planar Random (8.b) and UA (8.c). The conductivities of matrix and fibres are the same as those assumed in section 3.2. The results show that in those cases where a preferred orientation of the CNFs is assumed (Planar Random and Uniaxial), the thermal conductivity can boost in these directions even at values higher than that of the Cu matrix, provided the thermal conductivity of the interface is above a threshold value of around $10^9-10^{10}$ W/m$^2$K. However, in these cases, the directions perpendicular to the preferred directions of the fibres show a reduced thermal conductivity due to the thermal anisotropy of the CNFs, as the transversal conductivity of the fibres (6 W/m-K) is much lower than that of the matrix. The results also show that if the fibres are randomly oriented in all directions, it is only possible to obtain an enhancement in thermal conductivity with respect to Cu if the interface thermal conductance reaches values as high as $10^{10}$ W/m$^2$K, and even in those cases the predicted values are very close to the thermal conductivity of Cu.

The experimentally measured thermal conductivities found elsewhere [4-5] correspond to CNF-Cu composite plates processed by hot-press. During processing, the CNFs tend to align perpendicular to the applied load, and hence, the CNFs display a random planar preferred orientation. Hence, there are three possible explanations for the low thermal conductivities obtained: (1) degradation of the thermal properties of the CNFs, as has been shown to occur in some cases [12], (2) a poor interfacial thermal conductance, very likely in Cu-CNF composites and (3) that the measurements are taken in transversal directions in which the CNFs display a very poor thermal conductivity. Although the most plausible explanations are (1) and (2), (3) cannot be discarded, as most of the time the composites display a random planar configuration and the measurements are taken in the transversal direction by a laser flash method. Therefore,
it should be emphasized that, when measuring the thermal conductivity of these composites, special care should be taken in order to evaluate their thermal anisotropy and avoid misleading results due to anisotropic effects. Techniques such as modulated photothermal radiometry constitute a good choice to carry out such studies [13].

5. Conclusions

A thorough comparison of different simulation strategies to model the thermal conductivity of CNF reinforced Cu matrix composites has been carried out. The different strategies were based on the FEM of a RVE and on homogenization methods, such as MFA and DEM. In order to separate the influence of the different parameters involved (size of reinforcement, volume fraction, phase contrast, interface thermal conductance, anisotropy of the reinforcements), the comparison has been made first with spherical inclusions and then with short fibres represented by cylinders. The predictions show three regions: two plateaus at high and low thermal conductances and a transition zone between them.

First, in order to study the influence of the volume fraction, size of the reinforcement, phase contrast (difference in thermal conductivities between matrix and reinforcement) and interface thermal conductance, a composite with spherical isotropic inclusions was considered. The position of the transition zone depends strongly on the inclusion size, with smaller inclusions showing a more interface dominated behaviour, as expected. The results show a perfect match between MFA, DEM and FEM in the interface dominated transition zone, while some difference in the predictions were observed in the plateau. The differences are larger the larger the volume fraction and the phase
contrast between matrix and inclusions. It has to be noted too, that larger inclusion
radius and, therefore, less interface area per unit volume, results into the transition zone
starting at lower thermal conductances, i.e., the interface thermal conductance is less
critical.

Secondly, the case of a short fibre reinforced composite was considered. In this case, the
most important parameters controlling the thermal conductivity are the interface thermal
conductance and the preferred orientation of the fibres. For largely anisotropic
inclusions, the thermal conductivity is limited by the transversal thermal conductivity if
the fibres are randomly oriented. In this case, a substantial enhancement in thermal
conductivity is only possible if the fibres are randomly oriented in the plane or if they
are all aligned in one direction, but at the expense of displaying very poor thermal
conductivities in the transversal directions. To benefit from these effects, a minimum
interfacial thermal conductance of $10^9 - 10^{10}$ W/m$^2$K would be needed. Finally, the
simulation results show that, although degradation of the thermal properties of the CNFs
and a poor interfacial thermal conductance are very likely the reasons behind the low
conductivities reported, great care should be taken when measuring the thermal
conductivity of this new class of materials, to avoid misleading results due to
anisotropic effects.

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8. 

http://www.matweb.com/search/DataSheet.aspx?MatGUID=9aebe83845c04c1db5126fada6f76f7e&ckck=1


**Fig. 1:** A simple 2D model of a periodic short-fibre unit cell. When a fibre is cut, the rest of its volume reappears periodically through the opposite face.

**Fig. 2:** Example of periodic boundary conditions. A difference of 100 K is imposed in X axis. Each node in surface X = L is 100 K hotter than the node couple in the same position in X = 0. Node couples in surfaces Y = 0, L have the same temperature.

**Fig. 3:** Reinforcement architectures studied. a) 3DRandom: All the fibres are randomly positioned and oriented. b) Planar: Same as 3DRandom, but all the fibres are
perpendicular to axis Z. c) Uniaxial: All the fibres are randomly positioned and are parallel to axis Z

**Fig. 4:** Thermal conductivity of a composite with spherical inclusions and a perfect interface as predicted by FEM, MFA and DEM schemes for different volume fractions. FEM is limited to a maximum volume fraction of 0.4

**Fig. 5:** Variation in composite thermal conductivity with interface thermal conductance for a composite containing spherical inclusions with 10 μm in diameter and with a phase contrast of 10 (K_i=3850 Wm⁻¹ K⁻¹), using the MFA, FEM and DEM schemes. Volume fractions are a) 0.2 and b) 0.4 (0.387 for FEM predictions).

**Fig. 6:** Variation in composite conductivity with interface thermal conductance for a composite with V_f=0.4, three different inclusion diameters (0.1, 1 and 10 μm) and three different phase contrasts (K_i=38.5, 385, 3850 Wm⁻¹ K⁻¹).

**Fig. 7:** Comparison of MFA and FEM results for Uniaxial architecture and V_f = 0.15

**Fig. 8:** Effect of the interfacial conductance on global conductivity for short fibre composites. a) 3D Random, b) Planar and c) Uniaxial architectures with V_f = 0.28. Averaged over three different realizations.

**Table 1:** MFA and FEM results with %95 CI for the three architectures with averaged values over equivalent directions. In the case of FEM, values are averaged over three realizations. MFA simulates long fibre and V_f = 0.3, while FEM simulates short fibre with l/d = 5 and V_f = 0.28.
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