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A composite sphere assemblage model for porous oolitic rocks: Application to thermal conductivity

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ABSTRACT

The present work is devoted to the determination of linear effective thermal conductivity of porous rocks characterized by an assemblage of grains (oolites) coated by a matrix. Two distinct classes of pores, i.e. micropores or intra oolitic pores (oolite porosity) and mesopores or inter oolitic pores (inter oolite porosity), are taken into account. The overall porosity is supposed to be connected and decomposed into oolite porosity and matrix porosity. Within the framework of Hashin composite sphere assemblage (CSA) models, a two-step homogenization method is developed. At the first homogenization step, pores are assembled into two layers by using self-consistent scheme (SCS). At the second step, the two porous layers constituting the oolites and the matrix are assembled by using generalized self-consistent scheme (GSCS) and referred to as three-phase model. Numerical results are presented for data representative of a porous oolitic limestone. It is shown that the influence of porosity on the overall thermal conductivity of such materials may be significant.

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1. Introduction

The present work is devoted to the determination of overall thermal conductivity of heterogeneous isotropic porous rocks composed of an assemblage of quasi-spherical porous grains (oolites (a) and mesopores (b)) and solid grains (sparitic cement (c)). Oolitic limestones, recently studied by Ghabezloo et al. (2009), Grgic (2011), Nguyen et al. (2011) and Giraud et al. (2012) within the framework of the theory of random heterogeneous materials, are characterized by such type of microstructure. It can also refer to Regnet et al. (2015) for a recent synthesis of mechanical properties, permeability, and microstructural observations of oolitic carbonate rocks. In the limiting case of low or moderate volume fractions of grains, a matrix-inclusion homogenization method based on Eshelby approach with appropriate scheme may be used to estimate the effective poroelastic properties (see Dormieux et al., 2006; Levin and Alvarez-Tostado, 2006). For microstructures such as intermediate and large volume fractions of grains, an alternative approach is given by the composite sphere assemblage (CSA) models introduced by Hashin (1962). This homogenization method has been extensively developed by many authors for two-phase, three-phase and N-phase composite spheres (referred to as multi-coated spheres) (Schapery, 1968; Rosen and Hashin, 1970; Christensen, 1979; Christensen and Lo, 1979; Siboni and Benveniste, 1991; Hervé and Zaoui, 1993; Hervé, 2002; Schmitt et al., 2002; Torquato, 2002; He and Benveniste, 2004; Quang and He, 2007; Benveniste, 2008). In the context of geomaterials, micromechanical models for porous rocks may be found in Zimmerman (1991, 2000), Berryman and Berge (1993), Berryman and Pride (1998), and Jaeger et al. (2007). Many applications of CSA models to cement-based materials and concrete have been presented in Li et al. (1999a, b), Hashin and Monteiro (2002), Heukamp et al. (2005), and Bary and Béjaoui (2006). CSA model and its basic version, the three-phase model, will be adopted in this paper. It may be noticed that different approaches have been recently presented on the basis of the reformulation of the Maxwell's homogenization method for elastic composites presented in Sevostianov and Giraud (2013), which was applied to the estimate of effective elastic coefficients of oolitic rocks in Giraud and Sevostianov (2013). A micromechanical model for effective thermal conductivity of oolitic rocks based on Maxwell's homogenization method has been recently presented in Giraud et al. (2015). Details on recent works related to Maxwell's homogenization method can be found in Levin et al. (2012) and Sevostianov et al. (2015), and those on works related to conductivity can be found.
in Kushch et al. (2014) and Sevostianov and Mishuris (2014). This approach allows taking into account a microstructure description more realistic than CSA and further comparisons between these methods may be interesting. The model presented in this paper, based on CSA, has the advantage of simplicity, but the weakness is that it does not respect, at the mesoscale, the geometry of the microstructure, as it is based on a fractal representation of the heterogeneous medium. The strength of the CSA-based models is that it allows easily studying the effect of interfaces, like interfacial transition zone (ITZ) coating the grains (see Lutz and Zimmerman, 2005). It may be noticed that the effect of ITZ on conductivity has been recently discussed in Lutz and Zimmerman (2016) within the framework of Maxwell’s homogenization scheme. Maxwell’s method coincides with the one of the Hashin-Shtrikman bounds, and with Hashin’s CSA model in the particular case of homogeneous spherical inclusions.

In Section 4, numerical results are presented for representative oolitic limestone porous rocks. Two limiting cases are considered for numerical applications, i.e. the fully dry case in which all pores are saturated by air, and the fully water-saturated case. The objective is to evaluate the potential impact of the porosity on the overall thermal conductivity.

2. Background

Hereafter we define some notations and recall some results which are needed later. Bold letters refer to the second-order tensors; underlined letters \( \underline{e} \) and \( \underline{g} \) refer to the first-order tensors; \( \underline{i} \) represents the second-order identity tensor; \( \delta_{ij} \) denotes the Kronecker delta symbol, \( \delta_{ij} = 1 \) if \( i = j \), otherwise \( \delta_{ij} = 0 \). Oblate spheroid (\( 0 < \gamma < 1 \), where \( \gamma \) denotes the aspect ratio) and prolate spheroid (\( \gamma > 1 \)) with symmetry axis \( x_3 \) are described by the following equations:

\[
\begin{aligned}
\underline{e} &= \frac{\gamma^2 - 1}{a^2} \underline{e}_3 + \frac{\gamma^2}{a^2} \underline{e}_1 \\
a_1 &= a_2 = a \\
a_3 &= a \gamma
\end{aligned}
\]

(1)

The general case of the three-dimensional (3D) ellipsoid could be also considered but, for the sake of simplicity, only spheroids will be considered in this paper.

2.1. Polarization tensor for a spheroidal inhomogeneity

Hill polarization tensor of a spheroidal inhomogeneity in an infinite isotropic medium with conductivity \( \lambda \) can be written as (Torquato, 2002):

\[
\underline{P} = \frac{Q(\gamma)}{\lambda} \underline{i}_T + \frac{1 - 2Q(\gamma)}{\lambda} \underline{i}_N
\]

(2)

\[
\begin{aligned}
\underline{i}_N &= \underline{e}_3 \otimes \underline{e}_3 \\
\underline{i}_T &= \underline{e}_1 \otimes \underline{e}_1 + \underline{e}_2 \otimes \underline{e}_2 \\
\underline{i} &= \underline{i}_N + \underline{i}_T
\end{aligned}
\]

(3)

\[
\begin{aligned}
Q(\gamma) &= \left\{ \begin{array}{ll}
\frac{1}{2} \left[ 1 + \frac{1}{u} \left[ 1 - \frac{\gamma}{\sqrt{u}} \arctan \left( \frac{\sqrt{u}}{\gamma} \right) \right] \right] & (\gamma < 1) \\
1/3 & (\gamma = 1) \\
\frac{1}{2} \left[ 1 + \frac{1}{u} \left[ 1 - \frac{\gamma}{2\sqrt{u}} \ln \left( \frac{\gamma + \sqrt{u}}{\gamma - \sqrt{u}} \right) \right] \right] & (\gamma > 1)
\end{array} \right.
\]

(4)

where \( v = 1 - \gamma^2, u = \gamma^2 - 1 \).

Hill polarization tensor of a spherical inhomogeneity (\( \lambda = 1 \) and \( Q = 1/3 \)) is

\[
\underline{P} = \frac{1}{3\lambda} \underline{i}
\]

(5)

A recent work was presented in Pabst and Gregorová (2014), which focused on the thermal conductivity of porous media with spheroidal pores.

2.2. Dilute scheme

Dilute concentration tensor related to an ellipsoidal inhomogeneity of thermal conductivity \( \lambda_i \) embedded in an infinite matrix of thermal conductivity \( \lambda \) can be written as

\[
\underline{A}_i = \left[ \underline{i} + \underline{P}(\lambda_i - \lambda) \right]^{-1}
\]

(6)

For a spherical isotropic inhomogeneity \( i \) embedded in an infinite isotropic matrix, this tensor is isotropic (zero deviatoric part):

\[
\underline{A}_i = \underline{A}_i \underline{i}
\]

(7)

In the case of an isotropic ellipsoidal inhomogeneity with conductivity tensor \( \lambda_i = \lambda_i \) surrounded by an infinite isotropic matrix with conductivity tensor \( \lambda = \lambda_0 \), combining Eq. (1) with Eq. (5) gives

\[
\underline{A}_i = \lambda \left\{ \frac{1}{\lambda[1 - Q(\gamma)] + \lambda_0 Q(\gamma)} \underline{i}_T + \frac{1}{2\lambda_0 Q(\gamma) + \lambda_0[1 - 2Q(\gamma)]} \underline{i}_N \right\}
\]

(8)

Orientational average (case of a random orientation distribution) writes

\[
\underline{A} = \frac{1}{3} \left( \text{tr} \underline{a} \right) \underline{i}
\]

(9)

One has

\[
\underline{A}_i = \underline{A} \underline{i}
\]

(10)

with

\[
g(\lambda, \lambda_i, \gamma) = \frac{\lambda}{3} \left\{ \frac{2}{\lambda[1 - Q(\gamma)] + \lambda_0 Q(\gamma)} + \frac{1}{2\lambda_0 Q(\gamma) + \lambda_0[1 - 2Q(\gamma)]} \right\}
\]

(11)

2.3. Self-consistent scheme (SCS)

Self-consistent approximation for a two-phase material with spherical particles is the positive root of the quadratic equation (Torquato, 2002):

\[
\frac{f_1 \lambda_1}{\lambda_1 + 2 \lambda_2} + \frac{(1 - f_1) \lambda_2}{\lambda_2 + 2 \lambda_2} = \frac{1}{3} = 0
\]

(13)

It was originally developed by Bruggeman (1935). The solution is (Torquato, 2002):

\[
h(\lambda_1, \lambda_2, f_1) = \left( \alpha_{12} + \sqrt{\alpha_{12}^2 + 8 \lambda_1 \lambda_2} \right) / 4
\]

(14)

where
\[ \alpha_{12} = \lambda_1 (3f_1 - 1) + \lambda_2 (2 - 3f_1) \] (15)

Then the self-consistent approximation is written as

\[ J_{\text{SCS}}^{\text{GSCS}} = h(\lambda_1, \lambda_2, f_1) \] (16)

The self-consistent approximation with randomly oriented ellipsoidal particles is the solution of the following equation (Torquato, 2002):

\[ f_1 \left( \lambda_1 - \lambda^{\text{SCS}} \right) g(\lambda^{\text{SCS}}, \lambda_1, \gamma_1) + (1 - f_1) \cdot \left( \lambda_2 - \lambda^{\text{SCS}} \right) g(\lambda^{\text{SCS}}, \lambda_2, \gamma_2) = 0 \] (17)

It conduces to a polynomial equation of the fifth degree which may be easily solved numerically. One may notice that this equation could be also generalized by taking into account supplementary phases.

2.4. Generalized self-consistent scheme (GSCS)

The GSCS was introduced by Christensen (1979) and Christensen and Lo (1979), and has been extensively used in the context of granular and cement-based materials (Torquato, 2002). For a two-phase heterogeneous medium with microstructure, it is described by a CSA of two-phase composite spheres (or coated spheres). The third phase, surrounding the composite sphere, corresponds to the homogeneous equivalent medium with conductivity \( \lambda^{\text{SCS}} \) which is defined by a self-consistent type equation on the outer radius of the composite sphere. By denoting \( \lambda_1 \) and \( f_1 \) the thermal conductivity and the volume fraction of the core phase 1 surrounded by a matrix phase with the conductivity \( \lambda_2 \) and volume fraction \( f_2 = 1 - f_1 \), the effective thermal conductivity of the two-phase porous composite is

\[ \lambda^{\text{GSCS}} = \lambda_2 \left( \frac{f_1}{(1 - f_1)/3 + \lambda_2/(\lambda_1 - \lambda_2)} \right) \] (18)

The well-known relation (Eq. (18)) corresponds to the one in Christensen (2005). In the case of \( \lambda_2 < \lambda_1 \), it corresponds to Hashin-Shtrikman lower bound for an isotropic two-phase material. Considering the notations used for the dilute scheme, the core phase 1 corresponds to the inhomogeneity phase i (with the conductivity \( \lambda_i \)), phase 2 corresponds to the matrix phase (with the conductivity \( \lambda \)) of the dilute scheme, Eq. (18) can thus be rewritten as

\[ \lambda^{\text{GSCS}} = \lambda \left( 1 + \frac{f_1}{(1 - f_1)/3 + \lambda/(\lambda_1 - \lambda)} \right) \] (19)

2.5. Maxwell’s scheme

We only recall the homogenization formula of Maxwell’s scheme for a three-phase isotropic medium composed of a matrix (phase 3, with the conductivity \( \lambda_3 \)), spherical inclusions (phase 1, with the conductivity \( \lambda_1 \)) and randomly oriented oblate spheroidal inclusions (phase 2, with the conductivity \( \lambda_2 \) and aspect ratio \( \gamma_2 \)). Details on Maxwell’s homogenization scheme may be found in Torquato (2002) and Giraud et al. (2015):

\[ \lambda^{\text{MX}} = \lambda_3 \left( \frac{3 - 2\lambda_3 (f_3\Pi_2 + f_1H_1)}{3 + \lambda_3 (f_2H_2 + f_1H_1)} \right) \] (20)

3. Two-phase composite spheres and three-phase model

This section is devoted to presenting a two-step homogenization scheme for linear poroelastic properties of porous rocks. The microstructure of the investigated material is characterized by grains (referred to as oolites) surrounded by a matrix. Both oolites and matrix are porous materials and the respective parts of the porosity (matrix and oolite porosities) are supposed to be connected. This model is presented in Fig. 1. Microstructure of the reference Lavaux limestone is recalled in Fig. 2.

\[ H_1 = \frac{3(\lambda_3 - \lambda_1)}{\lambda_3 (2\lambda_3 + \lambda_1)} \] (21)

3.1. Volume fractions and constituents

At the macroscale, a two-phase composite material, composed of two isotropic poroelastic media: oolites (\( o \)) and matrix (\( m \)) surrounding the oolites, is considered. Thus we have

\[ \Omega = \Omega_o + \Omega_m, f_o = \frac{\Omega_o}{\Omega} f_m = \frac{\Omega_m}{\Omega} f_o + f_m = 1 \] (22)

At the mesoscale, the porosity is homogenized and the total porosity is decomposed into two parts, i.e. oolite porosity \( \Omega_{op} \) and matrix porosity \( \Omega_{mp} \), which are supposed to be connected with each other.

---

Fig. 1. A two-step model for porous oolitic rocks: Three-phase sphere assemblage model (two phases, or coated composite sphere, oolite \( o \), matrix \( m \)) (two-dimensional (2D) representation of a 3D structure).
At the first step, the oolite and matrix porosities are defined as

\[
f_{\text{op}} = f_{\text{op}} + f_{\text{mp}} = \frac{f_{\text{op}}}{f_{\text{op}}} = \frac{f_{\text{mp}}}{f_{\text{mp}}} (24)
\]

The solid particles constituting oolites and surrounding matrix are both supposed to be isotropic, with thermal conductivities \(\lambda_o\) and \(\lambda_m\), respectively. As the solid grains are composed of calcite, it is considered that \(\lambda_o = \lambda_m = \lambda_c\) for numerical applications, where \(\lambda_c\) denotes the isotropized conductivity of the calcite mineral.

### 3.2. First transition from the microscale to the mesoscale (step I)

The first step represents the transition from the microscale to the mesoscale. Oolite pores and matrix pores are homogenized and the two porous materials, i.e. porous oolite and porous matrix, are produced at the first step. Oolite solid and matrix solid are both supposed to be isotropic. As previously indicated, the granular and random microstructure of oolites conduce to choice of the self-consistent approximation (Bruggeman, 1935) for the step I. By respectively denoting \(\lambda_{\text{Iop}}\), \(\lambda_p\) and \(\lambda_o\) the conductivities of the oolite at the mesoscale, the intra oolitic porous phase, and the micritic solid grains (\(\lambda_o = \lambda_c\)), the well-known self-consistent approximation for a two-phase material with spherical particles (Eq. (13)) writes

\[
\frac{f_{\text{op}}^I}{\lambda_p} + \frac{1 - f_{\text{op}}^I}{\lambda_o} \frac{\lambda_o}{\lambda_p + 2\lambda_o^I} - \frac{1}{3} = 0 (25)
\]

Combining Eqs. (14)–(16), the solution to Eq. (25) is obtained:

\[
\lambda_{\text{Iop}}^I = h\left(\lambda_p, \lambda_o, f_{\text{op}}^I\right) (26)
\]

The SCS is also adopted for the matrix. This matrix corresponds to the filling material between oolites. By denoting \(\lambda_m\) the conductivity of the solid grains in the matrix, we have

\[
\lambda_{\text{Im}}^I = h\left(\lambda_p, \lambda_m, f_{\text{mp}}^I\right) (27)
\]

Based on the work of Giraud et al. (2015), another representation of the matrix porosity can be proposed. It corresponds to the inter oolitic porosity which may be represented by randomly oriented oblate spheroids and concave pores. In this paper, concave pores are neglected and replaced by oblate spheroids. Solid particles are represented by ellipsoids with similar aspect ratio. The self-consistent approximation with randomly oriented ellipsoidal particles is given by the solution to Eq. (17) which can be written as

\[
\begin{align*}
&f_{\text{mp}}^I (\lambda_p - \lambda_{\text{mp}}) + \left(1 - f_{\text{mp}}^I\right) \left(\lambda_{\text{mp}}^I - \lambda_m\right) \left(\lambda_{\text{mp}}^I - \lambda_m\right) = 0
\end{align*}
\]

### 3.3. Second transition from the mesoscale to the macroscale (step II)

In the second transition from the mesoscale to the macroscale, the microstructure of the medium is described by a CSA of two-phase composite spheres (or coated spheres), i.e. GSCS mentioned in Section 2. It corresponds to the three-phase model introduced by Christensen (1979) and has been widely used (Torquato, 2002). The
effective thermal conductivity of the two-phase porous composites can be written as (see Eq. (19)):

\[ \lambda_{\text{eff}}^{3\text{Phases}} = \frac{1}{\lambda_m} \left[ 1 + \frac{f_o \lambda_i (R_i^3 - 1) / R_i^3}{\lambda_i (R_i^3 - 1) + (R_i^3 - R_j^3) / (3R_j^3)} \right] \]  

(29)

3.4. Extension to four-phase model

It must be emphasized that the four-phase model could be easily developed as all the theoretical background is well-known. In the second transition from the mesoscale to the macroscale, the microstructure of the medium is described by a CSA of three-phase composite spheres (doubly coated, see Fig. 5). As previously mentioned, it corresponds to the four-phase model introduced by Christensen (1979), Ramesh et al. (1996), and Hashin and Monotero (2002) for the elastic problem and Hervé (2002) for the conductivity problem. The following procedure described in Hervé (2002) could be easily used by considering the doubly coated sphere:

\[ \lambda_{\text{eff}}^j = \lambda_j + \frac{\lambda_j (R_j^3 - 1) / R_j^3}{\lambda_j (R_j^3 - 1) + (R_j^3 - R_i^3) / (3R_i^3)} \]  

(30)

\[ R_1 = A, R_2 = B, R_3 = C \]  

(31)

For oolite core, we have

\[ \lambda_1 = \lambda_i^o \quad (0 \leq r \leq R_1) \]  

(32)

For ITZ, we have

\[ \lambda_2 = \lambda_i^i \quad (R_1 \leq r \leq R_2) \]  

(33)

For matrix, we have

\[ \lambda_3 = \lambda_i^m \quad (R_2 \leq r \leq R_3) \]  

(34)

Volume fractions of the doubly coated composite are

\[ f_0 = \frac{A^3}{C^3}, f_i = \frac{B^3}{C^3} - \frac{A^3}{C^3}, f_m = 1 - \frac{B^3}{C^3} \]  

(35)

Iterative procedure in this case becomes:

\[ \lambda_{\text{eff}}^j = \lambda_j + \frac{\lambda_j (R_j^3 - 1) / R_j^3}{\lambda_j (R_j^3 - 1) + (R_j^3 - R_i^3) / (3R_i^3)} \]  

(36)

\[ \lambda_{\text{eff}}^3 = \lambda_3 + \frac{\lambda_3 R_3^3 / R_4^3}{(\lambda_{\text{eff}}^2 - \lambda_3 + (R_2^3 - R_1^3) / (3R_1^3)} \]  

(37)

where

\[ \lambda_{\text{eff}}^2 = \lambda_i \left[ 1 + \frac{f_o / (f_o + f_i)}{\lambda_i (\lambda_i^o - \lambda_i) + (1 - f_o / (f_o + f_i)) / 3} \right] \]  

(38)

Effective conductivity can be obtained as follows:

\[ \lambda_{\text{eff}}^{4\text{Phases}} = \frac{1}{\lambda_m} \left[ 1 + \frac{f_o + f_i}{\lambda_i (\lambda_{\text{eff}}^2 - \lambda_m) + (1 - f_o - f_i) / 3} \right] \]  

(39)

Due to the lack of experimental data for thermal conductivity of the reference oolitic rock studied in this paper (Lavoux limestone), this study focuses on the simplest three-phase model. Nguyen et al. (2011) showed that the ITZ plays a crucial role in poroelasticity of porous oolitic rocks. Extension to imperfect interfaces has been presented and discussed in detail in Quang (2016a, b) for the conductivity problem. It may be noticed that the influence of a highly porous ITZ coating the oolites on the overall thermal conductivity may be significant in porous oolitic limestones with an important volume fraction of oolites. It would be particularly significant in the case of air-saturated pores, due to the high contrast of thermal conductivity between air and solid grains; a highly porous ITZ could be seen as an insulating layer coating oolites core in the dry case.

4. Numerical results

The sensitive study is similar to the one presented in recent studies (Giraud and Sevostianov, 2013; Giraud et al., 2015). Two cases are considered: porosity fully saturated with liquid water (wet case) and porosity fully saturated with air (dry case). The thermal conductivity of the pure calcite mineral is assumed as \( \lambda_C = 3.3 \) W/(m K) (Vasseur et al., 1995; Guéguen et al., 1997). The numerical values of thermal conductivities for liquid and air are \( \lambda_l = 0.5984 \) W/(m K) and \( \lambda_g = 0.0255 \) W/(m K), respectively (Clauser and Huenges, 1995). The reference data set corresponds to Lavoux limestone (Giraud and Sevostianov, 2013; Giraud et al., 2015). Volume fractions of oolite phase (o), matrix phase (m) and total poroelastic phase (p) are, for the Lavoux oolitic limestone, \( f_0 = 0.74 \), \( f_m = 0.26 \), \( f_0 = 0.26 \). Oolite porosity \( f_{op} = 0.14 \) and matrix porosity \( f_{mp} = 0.12 \). Relative porosities at the mesoscopic level are \( f_{op}^{me} = 0.19 \) and \( f_{mp}^{me} = 0.46 \). For the numerical sensitive study, the following assumption has been done (Giraud and Sevostianov, 2013):

\[ f_{op} = f_{mp} = f_0 / 2 \]  

(40)

Figs. 6–10 illustrate the dependence of effective thermal conductivity on the porosity (assuming that oolite pore and matrix pore families have the same volume fractions) at different volume fractions of oolites \( f_0 = 0.4, 0.5, 0.6 \) and \( 0.7 \).

The shape of matrix pores, which correspond to the inter oolitic pores, has a significant influence on the overall conductivity (see Figs. 6–9). As expected (see also Zimmerman, 1989), the higher impact is obtained for randomly oriented pores (\( \gamma = 0.01 \)) similar to...
penny-shaped cracks, and the lower for spherical pores ($\gamma = 1$). A comparison between three-phase model and Maxwell’s model (Giraud et al., 2015) is presented in Fig. 11.

Using Eqs. (20) and (21), we have ($\lambda_b = \lambda_g, f_b = f_g/2, \gamma_b = 0.05$):

$$\lambda_{\text{MX}}^{\text{M}} = \lambda_c \frac{3 - 2\lambda_c f_b H_b + f_o H_o}{3 + \lambda_c (f_b H_b + f_o H_o)}$$  (41)

where

$$H_b = \frac{\lambda_c - \lambda_b}{\lambda_c} g(\lambda_c, \lambda_b, \gamma_b), H_o = \frac{3 \left( \lambda_c - \lambda_o \right)}{2 \lambda_c + \lambda_o}$$  (42)

$$\lambda_o^l = h(\lambda_p, \lambda_o, f_p)$$  (43)

The impact of porosity, in the dry case, on the overall thermal conductivity is not significantly different between the models, which is slightly higher for the Maxwell’s scheme. These results need to be experimentally confirmed by characterization of thermal conductivity of such rocks in partially saturated range. The potential impact of ITZ is presented in Fig. 12. One has considered the extreme case of a highly porous ITZ having the thermal conductivity of air, i.e. $\lambda_{\text{ITZ}} = \lambda_g$, and two volume fractions ($f_{\text{ITZ}} = f_p/5$ and $f_{\text{ITZ}} = f_p/10$). Even if the considered values of volume fractions of ITZ combined with the thermal conductivity of air are not realistic, it confirms the potential role of ITZ which may be of crucial importance, but it also needs an experimental validation. For the two cases investigated, i.e. random distribution of flat or elongated pores, potential influence of porosity on thermal conductivity seems potentially significant, especially in the case of air saturated pores. As expected, due to the higher conductivity contrast between solid calcite mineral and air ($\lambda_c/\lambda_g \approx 129.4$), the maximal influence of ITZ is obtained in the case of air-saturated pores.

Fig. 6. Pores saturated with air. Oblate spheroidal pores $\gamma = 0.01$. Effective thermal conductivity as a function of porosity for different volume fractions of oolites.

Fig. 7. Pores saturated with air. Oblate spheroidal pores $\gamma = 0.05$. Effective thermal conductivity as a function of porosity for different volume fractions of oolites.

Fig. 8. Pores saturated with air. Oblate spheroidal pores $\gamma = 0.2$. Effective thermal conductivity as a function of porosity for different volume fractions of oolites.

Fig. 9. Pores saturated with air. Spherical pores $\gamma = 0.2$. Effective thermal conductivity as a function of porosity for different volume fractions of oolites.

Fig. 10. Pores saturated with water liquid. Spherical pores $\gamma = 1$. Effective thermal conductivity as a function of porosity for different volume fractions of oolites.
5. Conclusions

The presented simplified models based upon CSA may be used as a first approach for estimating thermal conductivity of isotropic materials constituted by grains coated by a matrix phase. Numerical results confirm the importance of the shape of mesopores for the conductivity problem (see Giraud and Sevostianov (2013) for the elastic problem). An assumption of spherical shape for all constituents (Giraud et al., 2012) at the mesoscale is irrelevant for this type of oolitic rocks in the range of high volume fraction of oolites.

Inter oolitic pores, which correspond in the three-phase model to the matrix pores, have shapes which can be approximated by oblate spheroids, or superspheres (concave shape), and the assumption of spherical shape is irrelevant (Giraud and Sevostianov, 2013; Giraud et al., 2015).

The advantages of CSA models are well-known and may be resumed, for the studied materials, as follows:

1. They are very simple and easy to use relations for effective thermal conductivity which allows taking into account volume fractions and conductivities of the constituent.
2. They are useful in the case of high contrast of properties between grains and surrounding matrix: the composite sphere may be easily generalized by adding supplementary phase, such as ITZ (it is very important and widely used for cement-based materials) (Hashin and Monteiro, 2002; Bary and Béjaoui, 2006).
3. Imperfect interfaces could be easily introduced.

Also, the weakness of CSA models are described as follows:

1. By their nature, due to their fractal construction, CSA models do not respect the geometry of the microstructure. For example, scale separation between micropores and mesopores is not respected, and so is the granulometry.
2. Compared to the porous oolitic rocks studied in this paper, the description of the mesoscale structure is far from reality as the shape of the material filling the space between grains is not correctly approximated.
3. For the oolitic rocks investigated in this paper, CSA models could be used as a first approach. Maxwell’s models seem to be more accurate and relevant as they allow more closely respecting the geometry of the microstructure at the mesoscale (Giraud et al., 2015). On-going work concerns experimental characterization of the thermal conductivity of such rocks at different saturation ratios and comparison with numerical results needs to be performed.

Conflict of interest

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