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# RADIATION IN POROUS MEDIA: AN UPSCALING METHODOLOGY

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## Abstract

The radiative properties of any porous medium are statistically defined by a cumulated extinction distribution function, a cumulated absorption probability and a bidirectional phase function. These functions are obtained by a Monte Carlo technique, from both the medium morphology data, known or issued from a X or  $\gamma$  ray tomography, and the radiative properties at a scale lower than the tomography spatial resolution. At this local scale, the porous medium phases can be opaque, transparent or semi transparent. The RDFI method leads to the determination, with a precise accuracy criterion, of the extinction and absorption coefficients, as long as the porous medium can be homogenized by one (or two) equivalent semi transparent medium. Under precise conditions discussed here, a radiative conductivity tensor (or two coupled tensors associated with the two phases) can be deduced from these radiative properties for an optically thick REV. Actual cases of porous media involved in new energy technologies are discussed vs the scales involved in the applications.

## Nomenclature

|                            |   |                 |                            |
|----------------------------|---|-----------------|----------------------------|
| $A, A_F$                   | specific area per unit volume, – of the fluid phase                         | $\nu$           | radiation frequency        |
| $G_e, g_e$                 | cumulated extinction distribution function, – for a semi transparent medium | $\Pi$           | porosity                   |
| $k_{ij}$                   | conductivity tensor component   | $\sigma$        | scattering coefficient     |
| $\text{Kn}_\nu^a$          | absorption Knudsen number   |                 |                            |
| $I_\nu$                    | radiation intensity   |                 | <i>indices related to:</i> |
| $P_a, p_a$                 | cumulated absorption probability, -for a semi transparent medium            | $i, j$          | coordinates                |
| $p_\nu$                    | phase function  | $F$             | fluid phase                |
| $\mathbf{q}^R, q_{i\nu}^R$ | radiative flux - <i>i</i> component   | $ot$            | optically thin limit       |
| $T$                        | temperature   | $S$             | solid phase                |
| $\mathbf{u}, u_i$          | unit vector - <i>i</i> component  | $w$             | interfaces                 |
| $\beta$                    | extinction coefficient  | (0)             | zero perturbation order    |
| $\delta$                   | REV size  | (1)             | first perturbation order   |
| $\kappa$                   | absorption coefficient  | $\alpha, \beta$ | phases                     |
|                            |   | *               | equivalent property        |

## 1 Introduction

Radiation in porous media at high temperature plays an important role in many applications, especially in new technologies of energy. It is the case, for instance, of radiation in foams used in catalytic combustion, in porous media used in Solid Oxide Fuel Cells, in solar absorbers, in downgraded nuclear reactor cores. Examples are given in Figs. 1 and 2. In all these applications,

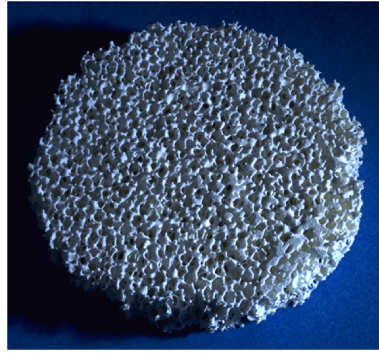


Figure 1: *Mullite foam sample (applied to catalytic combustion).*

it is impossible to directly calculate the radiative transfer at a local scale. An homogenized modeling of the system is required, generally by using an equivalent semi transparent continuous model.

Many studies have been carried out in order to characterize the radiative properties of the homogenized medium, i.e. extinction, scattering and absorption coefficients and phase function. Most of the authors [1-18] determine these radiative properties from experimental sets of data by various parameter identification techniques. Generally, these data are global and related to a finite system, for instance directional or hemispheric, spectral or total transmittances or reflectances of fluxes. On the other hand, a model depending on the parameters to determine allows these authors to calculate the same data in the experimental conditions by solving by different techniques the Radiative Transfer Equation (RTE), itself based on an equivalent semi transparent model. The set of parameters to determine are then optimized by minimization of the discrepancies between the theoretical and experimental data. In this type of approach, the errors introduced by each elementary step are cumulated and lead to generally unknown uncertainties on the identified coefficients and phase function. The errors are due to: i) experimental uncertainties on the global measurements; ii) the validity of the semi transparent model; iii) the accuracy of the RTE solution model; iv) the identification technique. Moreover, the larger is the number of parameters to be determined, the larger is the uncertainty on each parameter and the less physical is the meaning of the determination. The structure of the solution is often forced; it is the case of the phase function, often assumed of the Heyney-Greenstein type for instance.

The development of the X ray and  $\gamma$  ray tomography techniques with excellent spatial resolutions allows us today to use more direct determination of each coefficient and of the phase

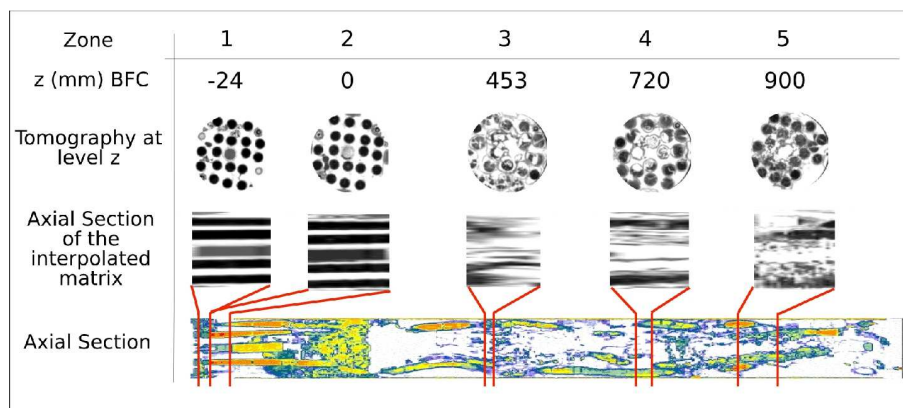


Figure 2: *Cross sections of a degraded nuclear core small scale model issued from a  $\gamma$  ray tomography (FPT1/IRSN/Cadarache).*

function, directly from their physical definitions. In a first step, a whole Representative Element of Volume (REV), or any phase of this REV, of any statistically homogeneous porous medium is characterized by a cumulated extinction distribution function, a cumulated absorption probability and a bidirectional phase function. In a second step, the Radiation Distribution Function Identification method (RDFI, developed in Refs.[19-23] leads, if the precise conditions of validity of the equivalent semi transparent model are verified, to the determination of the extinction and absorption coefficients associated with the whole porous medium or a given phase of this medium.

Section 2 deals with the determination of the radiative properties of such an equivalent semi transparent medium, i.e. extinction, scattering and absorption coefficients and phase function. The last section introduces a rational approach which directly leads to a radiative conductivity tensor for an optically thick radiative Representative Element of Volume or any optically thick phase within this REV. Two transversal goals of this paper are to consider under which conditions: i) a porous medium, or any phase of this medium, can be modeled by an equivalent semi transparent medium for radiative transfer applications; ii) the radiative flux degenerates in a diffusion flux.

## 2 Statistical determination of the extinction and absorption coefficients

### 2.1 Cumulated distribution function identification

The present topic is limited to effects of pores of size larger than the radiation wavelengths involved in the considered applications. Consequently, the diffraction is neglected and the propagation follows the geometrical optics laws. The extinction, absorption and scattering properties of any real porous medium can be completely defined by a statistical approach based on the cumulated distribution function of extinction, the cumulated absorption probability and the phase function, even for media that can't be modeled as semi transparent with a reasonable accuracy. In the following, the porous media will be considered in equilibrium or in non equilibrium.

Let us first consider the simplest case of a radiative Representative Elementary Volume (REV) of a medium of porosity  $\Pi$ , characterized by a uniform temperature  $T$  and a uniform intensity directional field  $I_\nu(\mathbf{u})$ . We focus on the propagation of any ray in a given direction  $\mathbf{u}$ , at the frequency  $\nu$ , from a source point  $M$  of abscissa  $s = 0$ . The source points are randomly chosen anywhere in any non opaque phase of the medium.

The *cumulated distribution function of extinction*  $G_{e\nu}(s, \mathbf{u})$  is the probability for a ray issued from  $M$  to be extinguished in the interval  $[0, s]$  (see Refs[19, 20]). Extinction can occur at a point  $I$ , of abscissa  $s'$ , by reflection or absorption on an opaque wall, by absorption or scattering within any volume element of a non opaque phase, or by reflection or transmission at the interface between non opaque phases, as seen in Fig.3. In all these cases,  $G_{e\nu}(s, \mathbf{u})$  is the cumulated distribution function of the paths  $MI$  of length  $s$ . It is worth of notice that, in the case of a transparent fluid phase and an opaque solid phase,  $G_e(s, \mathbf{u})$  is an intrinsic property of the material, independent of the frequency  $\nu$ .

The *cumulated absorption probability*  $P_{a\nu}(s, \mathbf{u})$  is the probability for a ray issued from  $M$  to be absorbed within the interval  $[0, s]$ . Absorption can occur at a point  $I$ , of abscissa  $s'$ , on an opaque wall or in any volume element of a non opaque phase. The *cumulated scattering probability*  $P_{s\nu}(s, \mathbf{u})$  is the probability for a ray issued from  $M$  to be scattered within  $[0, s]$ . Scattering can occur at a point  $I$ , of abscissa  $s'$ , by reflection on an opaque wall, by scattering in any volume element of a non opaque phase, or by reflection or transmission at the interface

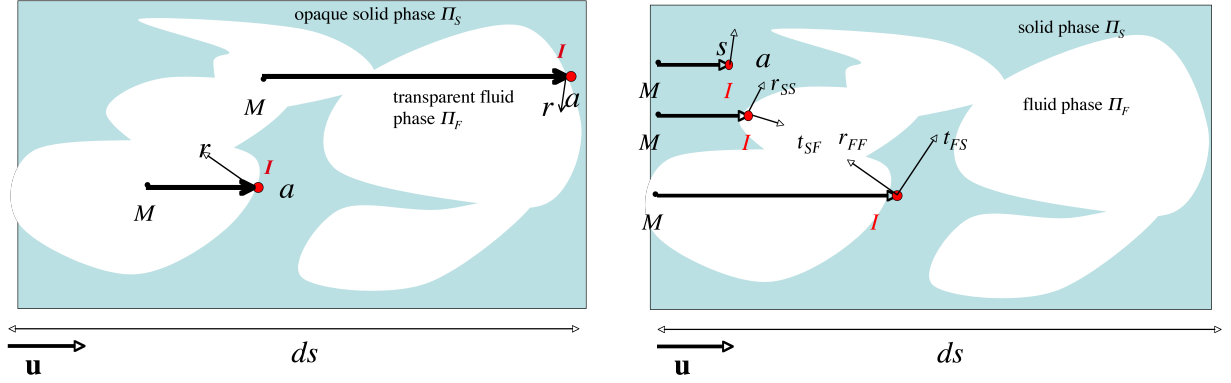


Figure 3: Schematic representation of the events for a given direction  $\mathbf{u}$  within a REV of size  $ds$ : left, opaque solid phase and transparent fluid phase; right, semi transparent solid phase and transparent fluid phase.

between non opaque phases. It is obvious that

$$G_{ev}(s, \mathbf{u}) = P_{av}(s, \mathbf{u}) + P_{sv}(s, \mathbf{u}) \quad (1)$$

and  $G_{ev}(s, \mathbf{u})$  belongs to the range  $[0, 1]$ .  $G_{ev}(s, \mathbf{u})$ ,  $P_{av}(s, \mathbf{u})$ ,  $P_{sv}(s, \mathbf{u})$  and the bidirectional scattering phase function  $p_\nu(\mathbf{u}', \mathbf{u})$  can easily be calculated:

- from morphological data characterizing the porous medium, for instance issued from a X ray or a  $\gamma$  ray tomography;
- from radiative data at a local scale, smaller than the scales accounted for in the morphological data: absorption or scattering laws in a phase that is semi transparent at this local scale, or reflection law for an opaque wall. This point will be discussed in the following from an example.

On the other hand, a semi transparent medium is generally characterized by an extinction coefficient  $\beta_\nu(\mathbf{u})$ , an absorption coefficient  $\kappa_\nu(\mathbf{u})$  and a phase function  $p_\nu(\mathbf{u}', \mathbf{u})$ . But from the previous statistical point of view, it is also characterized by a cumulated distribution function of extinction  $g_{ev}(s, \mathbf{u})$  and a cumulated absorption probability  $p_{av}(s, \mathbf{u})$  given by

$$g_{ev}(s, \mathbf{u}) = 1 - \exp[-\beta_\nu(\mathbf{u})s] \quad (2)$$

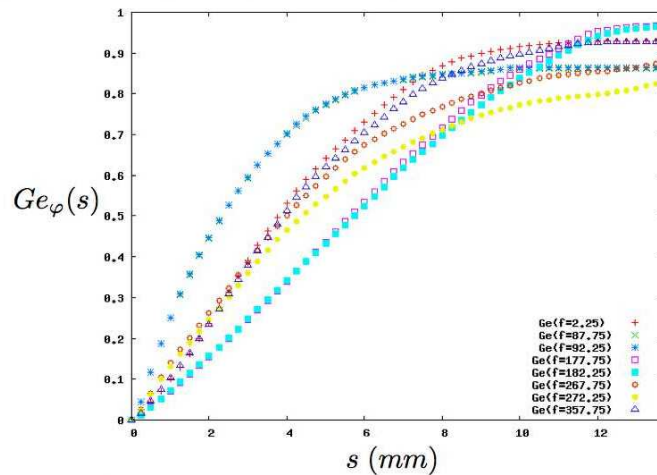


Figure 4: Examples of  $G_e(\theta, \varphi)$  functions related to cross sections of a rod bundle in triangular array ( $\theta = \pi/2$ );  $\varphi = f$  is the azimuth (in degrees); [23].

$$p_{av}(s, \mathbf{u}) = (1 - \exp[-\beta_\nu(\mathbf{u})s]) [\kappa_\nu(\mathbf{u})/\beta_\nu(\mathbf{u})] \quad (3)$$

Consequently, a semi transparent medium is characterized, for any frequency and direction, by an exponential extinction cumulated distribution function and an exponential absorption cumulated probability. This property is not general as seen in Fig.4. A porous medium can be modeled as an homogeneous semi transparent equivalent medium only if the actual  $G_{e\nu}(s, \mathbf{u})$  and  $P_{av}(s, \mathbf{u})$  functions are close to exponential functions. The Radiative Distribution Function Identification method (RDFI) is based on this simple idea. It has been first developed by Tancrez and Taine [19] for virtual porous media with a transparent fluid phase and an opaque solid phase. It has been generalized to real porous media by Zeghondy et al. [20], for a semi transparent solid phase and a transparent fluid phase, and by Petrash [21] and Chalopin et al. [23, 22] for an opaque solid phase and a transparent fluid phase.

In practice,  $G_{e\nu}(s, \mathbf{u})$  and  $P_{av}(s, \mathbf{u})$  are calculated by a stochastic Monte Carlo method, as detailed in the previous references. The global relative standard deviation on  $G_{e\nu}(s, \mathbf{u})$  and  $P_{av}(s, \mathbf{u})$ , obtained by a least square fit method, is small, typically  $10^{-6}$  to  $10^{-4}$ .  $G_{e\nu}(s, \mathbf{u})$  and  $P_{av}(s, \mathbf{u})$  very accurately characterize the porous medium.

The equivalent extinction coefficient  $\beta_\nu^*(\mathbf{u})$  is obtained by identification, also by a least square fit method, of  $G_{e\nu}(s, \mathbf{u})$  with  $g_{e\nu}(s, \mathbf{u})$  and the equivalent absorption coefficient  $\kappa_\nu^*(\mathbf{u})$  by identification of  $P_{av}(s, \mathbf{u})$  with  $p_{av}(s, \mathbf{u})$ . This identification is carried out, with an iterative scheme, for an optical path  $\beta_\nu^*s$  varying in the range  $[0, 3]$ . In this way, the resulting value of  $\beta_\nu^*$ ,  $\kappa_\nu^*$  and  $\sigma_\nu^*$  are valid for all radiative transfer applications, if the identification procedure is accurate.

The accuracy of the identification procedure is measured in the case of  $G_{e\nu}(s, \mathbf{u})$  and  $g_{e\nu}(s, \mathbf{u})$  by the criterion  $\epsilon_e(\beta_\nu^*)$  defined as

$$\epsilon_e(\beta_\nu^*) = \left\{ \sum_{i=0}^N [G_e(s_i, \mathbf{u}) - g_e(s_i, \mathbf{u})]^2 / \sum_{i=0}^N [1 - G_e(s_i, \mathbf{u})]^2 \right\}^{1/2} \quad (4)$$

where the summation is applied to the set of the considered discrete values  $s_i$  of paths  $MI$ , such as  $\beta_\nu^*s_i$  belongs to the range  $[0, 3]$ . A real porous medium can be modeled, after homogenization, by a continuous semi transparent medium  $\kappa_\nu$  only if  $\epsilon_e(\beta_\nu^*)$  is less than some  $10^{-2}$ . A similar criterion is applied for the identification of  $\kappa_\nu$ .

The whole previous approach can easily be generalized to a porous medium in non equilibrium, i.e. with a solid phase ( $S$ ) and a fluid phase ( $F$ ) characterized in a REV centered in  $\mathbf{x}$  by uniform temperatures  $T_S(\mathbf{x})$  and  $T_F(\mathbf{x})$  and uniform intensity directional fields  $I_{S\nu}(\mathbf{u}, \mathbf{x})$  and  $I_{F\nu}(\mathbf{u}, \mathbf{x})$ . The method is separately applied to each non opaque phase, at the local scale, within the medium. Under the conditions of validity of the semi transparent model, it leads to equivalent extinction and absorption coefficients for each phase, i.e  $\beta_{F\nu}^*(\mathbf{u})$ ,  $\beta_{S\nu}^*(\mathbf{u})$ ,  $\kappa_{F\nu}^*(\mathbf{u})$  and  $\kappa_{S\nu}^*(\mathbf{u})$ . If there is no opaque phase in the medium, two directional scattering coefficients and two bidirectional phase functions are defined for each phase, i.e  $\sigma_{FF\nu}^*(\mathbf{u})$ ,  $\sigma_{FS\nu}^*(\mathbf{u})$  and  $\sigma_{SS\nu}^*(\mathbf{u})$ ,  $\sigma_{SF\nu}^*(\mathbf{u})$ ,  $p_{FF\nu}^*(\mathbf{u}', \mathbf{u})$ ,  $p_{FS\nu}^*(\mathbf{u}', \mathbf{u})$  and  $p_{SS\nu}^*(\mathbf{u}', \mathbf{u})$ ,  $p_{SF\nu}^*(\mathbf{u}', \mathbf{u})$ , respectively. The first and third scattering coefficients and phase functions characterize both the scattering phenomena within a phase and the reflection at an interface between the two phases. The second and the last ones characterize the transmission phenomena from a phase to the other one. Consequently, the two RTEs associated with the two sets of coefficients and then the intensity fields  $I_{S\nu}(\mathbf{u}, \mathbf{x})$  and  $I_{F\nu}(\mathbf{u}, \mathbf{x})$  are strongly coupled through the transmission phenomena. The associated RTEs are given in Ref. [20]; each of them depends on the temperature fields of the two phases.

In the case of an opaque phase in non equilibrium with a transparent phase, the homogenized extinction and absorption coefficients and phase function, defined within the real transparent phase, are obtained in the same manner as in the case of a REV in equilibrium.

## 2.2 Some results and validity of the semi transparent model

Let us first consider porous media with an opaque solid phase and a transparent fluid phase. Tancrez and Taine [19] have systematically studied this identification procedure, especially for virtual sets of overlapping transparent spheres characterized by a Gaussian size distribution within a solid opaque phase. These virtual media are statistically homogeneous and isothermal; it is a realistic model for industrial foams. For this application, Fig.5 shows  $\beta^+$  vs the porosity  $\Pi$ ;  $\beta^+$  is the ratio  $\beta^* / \beta_{ot}$ , where  $\beta_{ot}$  is the extinction coefficient for the asymptotic case of an optically thin medium ( $\Pi$  close to 1). For any statistically isotropic and homogeneous porous medium with an opaque solid phase and a transparent fluid phase, it has been shown that  $\beta_{ot}$  is equal to  $A_F/4$  where  $A_F$  is the specific surface per unit volume of the fluid phase. In radiation,  $A_F^{-1}$  is the most pertinent parameter characterizing the pore size, for any type of porous medium. The parameter  $A_F^+$  which also appears in Fig.5 is equal to  $A_F \langle R \rangle$ , where  $\langle R \rangle$  is the mean spheric pore radius in the considered medium. As shown in Fig 5, a medium of this type can be modeled by an equivalent semi-transparent medium with a reasonable accuracy only for a porosity larger than 0.6. It is worth of notice that the equivalent semi transparent medium occupies the volume of the fluid phase, which is transparent at the local scale!

Chalopin et al. have obtained in Refs.[23, 22] respectively, by using the RDFI method, homogenized radiative properties of: i) opaque rod bundles with rectangular or triangular arrays (safe nuclear reactor core bundle); ii) degraded bundles (issued from a severe accident experimental simulation FPT1, see Fig.2). In the first case, extinction and absorption coefficients, strongly dependent on the two Euler angles, have been obtained for both triangular and squared arrays. The treatment of the phase function has to account for the consequences of the dependence on incidence direction of the scattering coefficient. Media of this type are not always accurately modeled by an equivalent semi transparent model as seen in Fig.4.

In a general manner, in the case of an opaque phase and a transparent one, when the validity conditions of a semi transparent model are fulfilled, results are given by expressions of the type

$$\beta^* / \beta_{ot} = K \beta^* / A_F = f(\Pi, \text{geometry}). \quad (5)$$

$$\kappa_\nu^* / \beta^*, \sigma_\nu^* / \beta^* = f(\Pi, \text{geometry}, \text{reflection law}). \quad (6)$$

In Eq.5,  $K$  is a constant depending on the geometry. The medium tortuosity is an important parameter which influences the results.

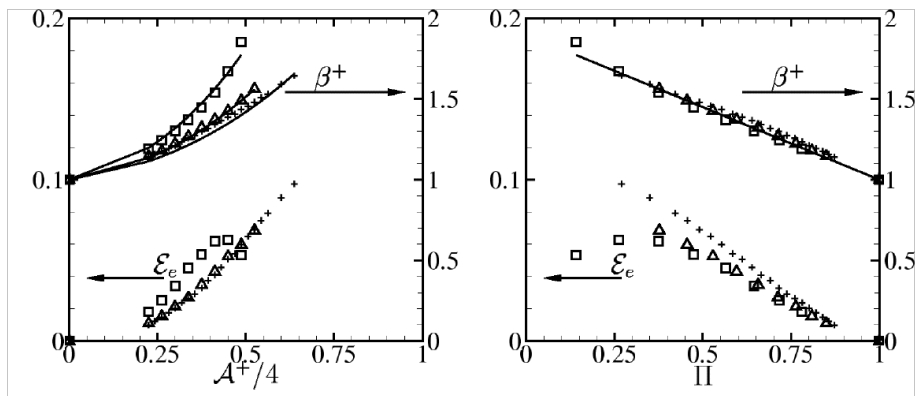


Figure 5: *Non dimensional extinction coefficient vs  $A_F^+$  or  $\Pi$  for sets on overlapping transparent spheres with dispersed radius distributions within a solid phase; associated validity criterion [19]*

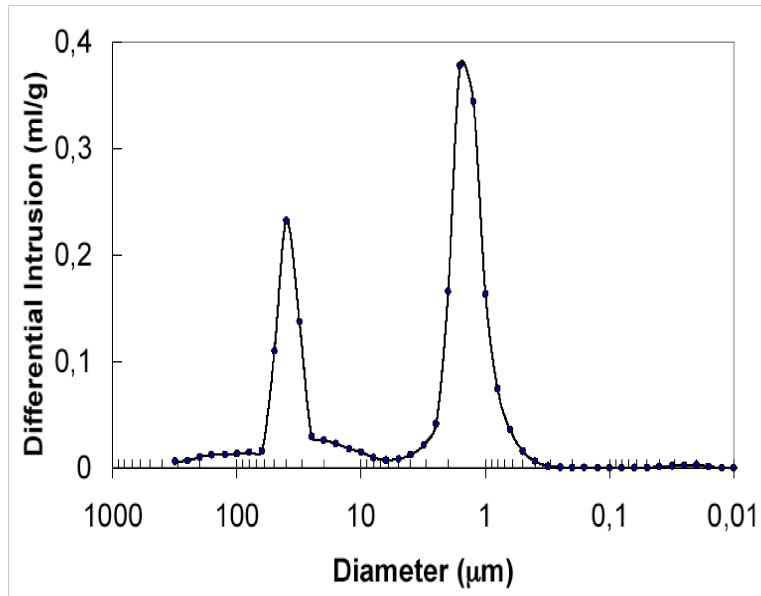


Figure 6: *Mullite foam sample: results of Hg porosimetry measurements; only the two smallest pore classes appear (communication from F. Enguehard, CEA, Le Ripault, France).*

Let us consider now the case of a medium with transparent and semi transparent phases (at least, one of them semi transparent). Zeghondy and al.[20] have studied a foam (mullite) with a semi transparent solid phase at the local scale and a transparent fluid phase. The medium morphology is defined by a 3D voxel matrix, issued from a X ray tomography technique carried out at ESRF with a spatial resolution of  $5 \mu m$ . For a scale less than  $5 \mu m$ , the radiative properties of the mullite have been experimentally determined in Zeghondy and al.[24] (extinction coefficient, albedo close to 0.99, and phase function almost isotropic). It is an extremely scattering material. The RDFI method has been carried out from both the tomography data and the local mullite radiative properties (at a scale less than  $5 \mu m$ ). The whole porous medium has been characterized by an extinction coefficient axisymmetrically depending on the direction, an albedo, and a phase function depending, in practice, only on the scattering angle. These determinations are accurate ( $\epsilon_e(\beta_v^*)$  defined by Eq.4 is less than  $2 \cdot 10^{-2}$ ). In this case, a continuous semi transparent equivalent medium represents accurately the whole porous medium, both the semi transparent solid phase and the transparent fluid phase.

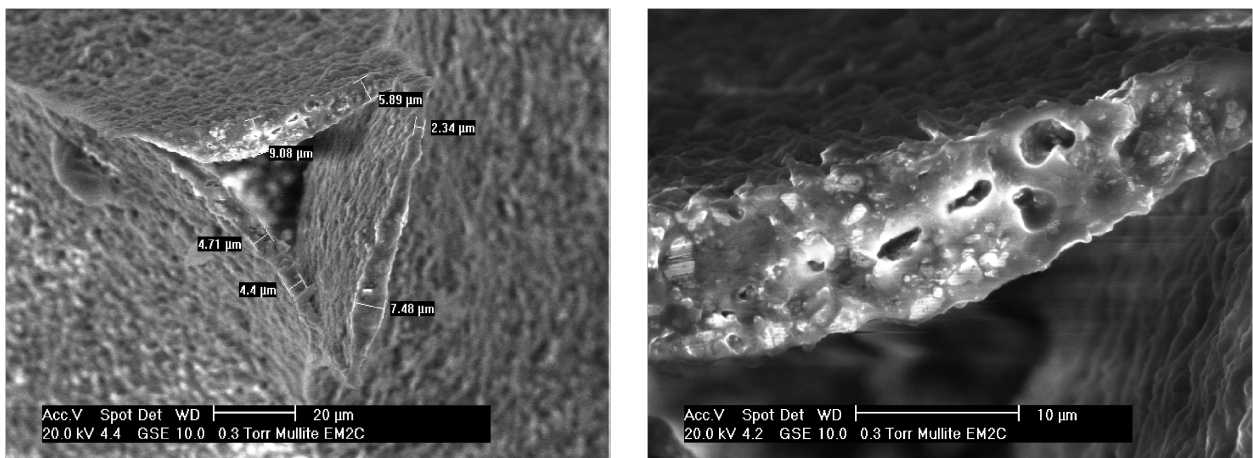


Figure 7: *Scanning Electron Microscopy images of a mullite sample: scales are given in the figures (communication from B. Rousseau and P. Echegut, CEMHTI/CNRS, Orléans, France).*



The considered mullite samples are characterized by three clearly separate and nonoverlapping pore size classes, i.e.  $1 \mu m$ ,  $40 \mu m$  and  $300 \mu m$  with similar probability density functions for each class, as shown for the two smallest classes in Figs.6 and 7. The RDFI method has been applied, on the one hand, with the real spatial resolution of the tomography ( $5 \mu m$ ) and, on the other hand, with a downgraded resolution ( $20 \mu m$ ). In these approaches, the intermediate class is only partially taken into account with the  $20 \mu m$  resolution. It appears that there is no significant difference in the results obtained with the two resolutions. The explanation is due to the fact that the intermediate class partially contributes only to the small  $s$  value distribution, i.e. to optically thin paths. Because the medium is characterized by a unique extinction coefficient in the  $[0, 3]$  optical thickness range, the effect of this class is weak in the identification procedure.

In conclusion, the advantage of the RDFI method is to allow us to validate, on a physical basis, the pertinence of the homogenization of one or all the phases of a real porous medium by a continuous equivalent semi transparent model. In some cases, this model is not valid. New approaches, which generalize the RTE, have now to be developed for the treatment of this type of media (low porosity foams, degraded nuclear reactor core, . . . ), which have important applications.

At this step, the radiative transfer is modeled by a RTE, which has to be coupled with the other transfer modes, through generally hard and tedious calculations. For optically thick media at a scale for which the medium can be considered as isothermal, the RTE degenerates, under some precise conditions, in a tensorial Fourier law. This point is discussed in Sec.3. The next subsection deals with the expression of the radiative flux and power in the general case.

## 2.3 Radiative flux and power

After homogenization of the porous medium by one (or two) equivalent semi transparent medium, with or without the phase equilibrium hypothesis, the radiative flux and the radiative power per unit volume can be calculated in a general manner.

Under the phase equilibrium hypothesis of a porous medium with a semi transparent phase and a transparent phase or two semi transparent ones, each REV centered in  $M(\mathbf{x})$ , of uniform temperature  $T(\mathbf{x})$ , is characterized by a uniform intensity directional field  $I_\nu(\mathbf{u}, \mathbf{x})$ , which strongly depends on the whole temperature field  $T(\mathbf{X})$  of the homogenized medium, i.e. of the temperature of any point  $N(\mathbf{X})$  of the homogenized medium. The radiative flux within the whole REV is

$$\mathbf{q}^R(\mathbf{x}) = \int_0^\infty \int_{4\pi} I_\nu(\mathbf{u}, \mathbf{x}) \mathbf{u} d\Omega d\nu, \quad I_\nu(\mathbf{u}, \mathbf{x}) = f[T(\mathbf{X})] \quad (7)$$

The radiative power per unit volume is then

$$P^R(\mathbf{x}) = -\nabla \cdot \mathbf{q}^R \quad (8)$$

$P^R(\mathbf{x})$  is a source term in the energy balance of an elementary volume  $dV$  of the whole homogenized medium.

On the other hand, in non equilibrium conditions of a porous medium (see Refs. [25, 26]) with a semi transparent phase and a transparent phase ( $\alpha, \beta$ ), or two semi transparent ones, each phase of the REV centered in a point  $M(\mathbf{x})$  is characterized by uniform temperature  $T_\alpha(\mathbf{x})$  and a uniform intensity directional field  $I_{\alpha\nu}(\mathbf{u}, \mathbf{x})$ , which strongly depends on the whole temperature fields  $T_\alpha(\mathbf{X})$  and  $T_\beta(\mathbf{X})$  of the two phases, as pointed out at the end of Sec.2.1. The radiative flux within each phase, conventionally defined for the whole REV section, is

$$\mathbf{q}_\alpha^R(\mathbf{x}) = \Pi_\alpha \int_0^\infty \int_{4\pi} I_{\alpha\nu}(\mathbf{u}, \mathbf{x}) \mathbf{u} d\Omega d\nu, \quad I_{\alpha\nu}(\mathbf{u}, \mathbf{x}) = f[T_\alpha(\mathbf{X}), T_\beta(\mathbf{X})] \quad (9)$$

$\Pi_\alpha$  is equal to  $\Pi$  if  $\alpha$  is the fluid phase, to  $1 - \Pi$  in the other case. The radiative power in each phase, defined by unit volume of the whole medium, is given by an equation similar to Eq.8.

In the case of non equilibrium conditions of a porous medium with, at the local scale, an opaque phase and a transparent phase, the transparent phase of a REV centered in a point  $M(\mathbf{x})$  is characterized, for radiative transfer, by the uniform temperature  $T_w(\mathbf{x})$  of the interface and a uniform intensity directional field  $I_\nu(\mathbf{u}, \mathbf{x})$ , which strongly depends on the whole temperature field of the interface within the homogenized medium  $T_w(\mathbf{X})$

$$\mathbf{q}^R(\mathbf{x}) = \Pi \int_0^\infty \int_{4\pi} I_\nu(\mathbf{u}, \mathbf{x}) \mathbf{u} d\Omega d\nu, \quad I_\nu(\mathbf{u}, \mathbf{x}) = f[T_w(\mathbf{X})]. \quad (10)$$

By convention, the radiative flux is considered in regards to the whole REV, which leads to the factor  $\Pi$  in Eq.10. The radiative power in the transparent phase is here also given by an equation similar to Eq.8. The radiative flux  $d\Phi_w^R$ , applied to the interface within the REV of whole volume  $dV$  assumed in non equilibrium, and the radiative flux per unit area within the whole REV  $\varphi_w^R$  are then

$$d\Phi_w^R = -\nabla \cdot \mathbf{q}^R dV, \quad \varphi_w^R = -\nabla \cdot \mathbf{q}^R / A, \quad (11)$$

where  $A$  is the specific surface by unit volume of the whole medium.

### 3 Radiative conductivity tensor

#### 3.1 Direct modeling

The purpose of this section is to express the radiative flux for a REV, optically thick from the point of view of absorption (or an optically thick phase within the REV).

We consider, in a first step for the sake of simplicity, an opaque solid phase and a transparent fluid phase with isotropic extinction, scattering and absorption coefficients. After homogenization, the radiative flux within the transparent phase is given by Eq.10. The intensity  $I_\nu(\theta, \varphi)$ , associated with the unit direction vector  $u_i(\theta, \varphi)$ , is obtained by solving the RTE, written in tensorial notations

$$\frac{\partial I_\nu}{\partial x_j}(\theta, \varphi) u_j(\theta, \varphi) + \beta^* I_\nu(\theta, \varphi) = \kappa_\nu^* I_\nu^\circ(T_w) + \frac{\sigma_\nu^*}{4\pi} \int_{4\pi} p_\nu^*(\theta', \varphi', \theta, \varphi) I_\nu(\theta', \varphi') d\Omega', \quad (12)$$

where cartesian coordinates  $x_j$  are introduced. As pointed out in the previous section,  $\beta^*$  does not depend on  $\nu$  in the case of a solid opaque phase.  $I_\nu^\circ(T_w)$  is the equilibrium intensity at the *REV interface temperature*  $T_w$ , and  $\sigma_\nu^*$  is the scattering coefficient, equal to  $\beta^* - \kappa_\nu^*$ .

Let the REV be optically thick on a length  $\delta$ , from the point of view of absorption. The RTE can be written under the adimensional form

$$\text{Kn}_\nu^a \frac{\partial I_\nu}{\partial x_j^\dagger}(\theta, \varphi) u_j(\theta, \varphi) + \frac{\beta^*}{\kappa_\nu^*} I_\nu(\theta, \varphi) = I_\nu^\circ(T_w) + \frac{\sigma_\nu^*}{4\pi \kappa_\nu^*} \int_{4\pi} p_\nu^*(\theta', \varphi', \theta, \varphi) I_\nu(\theta', \varphi') d\Omega', \quad (13)$$

where  $x_j^\dagger$  is equal to  $x_j/\delta$  and

$$\text{Kn}_\nu^a = (\kappa_\nu^* \delta)^{-1} \quad (14)$$

is an absorption Knudsen number, small in front of 1. It is the ratio of the photon absorption mean free path  $(\kappa_\nu^*)^{-1}$  to  $\delta$ . Under this form the RTE can be solved, as the Boltzmann equation, by a perturbation method with respect to the parameter  $\text{Kn}_\nu^a$ , small in front of 1 (see [27]). The solution of Eq.13 can be searched under the form

$$I_\nu \simeq I_\nu^{(0)} + I_\nu^{(1)}, \quad (15)$$

where  $I_\nu^{(0)}$  is the zero order perturbation term, independent of  $\text{Kn}_\nu^a$ , and  $I_\nu^{(1)}$  is the first order perturbation term, proportional to  $\text{Kn}_\nu^a$  and consequently small in front of  $I_\nu^{(0)}$ . Indeed, it will be shown in Sec.3.2 under which precise condition on  $\delta$  or  $\text{Kn}_\nu^a$  the terms of higher order,  $I_\nu^{(2)}, I_\nu^{(3)} \dots$ , can be neglected in front of the two first ones.

From Eq.13, we obtain on the one hand the equation verified by the zero order intensity, by neglecting higher order contributions

$$I_\nu^{(0)}(\theta, \varphi) = (1 - \omega_\nu^*) I_\nu^\circ(T_w) + \frac{\omega_\nu^*}{4\pi} \int_{4\pi} p_\nu(\theta', \varphi', \theta, \varphi) I_\nu^{(0)}(\theta', \varphi') d\Omega' \quad (16)$$

where  $\omega_\nu^* = \sigma_\nu^*/\beta^*$  is the albedo. Due to a phase function property (see [22]),  $I_\nu^\circ(T_w)$  is an exact solution of Eq.16, and if we assume the unicity of  $I_\nu^{(0)}$ , we obtain

$$I_\nu^{(0)} = I_\nu^\circ(T_w). \quad (17)$$

This corresponds to the Local Thermodynamical Equilibrium (LTE) radiation. On the other hand, the first order perturbation is the solution of

$$I_\nu^{(1)}(\theta, \varphi) = -\frac{u_j(\theta, \varphi)}{\beta^*} \frac{dI_\nu^\circ}{dT}(T_w) \frac{\partial T_w}{\partial x_j} + \frac{\omega_\nu^*}{4\pi} \int_{4\pi} p_\nu(\theta', \varphi', \theta, \varphi) I_\nu^{(1)}(\theta', \varphi') d\Omega'. \quad (18)$$

This implicit integral equation can be solved by an iterative technique and  $I_\nu^{(1)}$  is then expressed as a linear combination of the temperature gradient components, similarly to the velocity distribution function in the Chapman and Cooling solution method of the Boltzmann equation [28].

As  $I_\nu^{(0)}$ , equal to  $I_\nu^\circ(T_w)$ , is isotropic, its contribution to the radiative flux in Eq.10 is null. The radiative flux, directly obtained by using  $I_\nu^{(1)}$  in Eq.10, can then be expressed as a Fourier law,

$$q_i^R = -k_{ij} \frac{\partial T_w}{\partial x_j} \quad (19)$$

where  $k_{ij}$  are the radiative conductivity tensor components. This is only valid at the limit of a radiative Knudsen number  $\text{Kn}_\nu^a$  small in front of 1 (as shown in Sec.3.2). The radiative conductivity tensor  $k_{ij}$  is generally diagonal, but is reduced to a scalar in the case of a statistically isotropic porous medium. The radiative power per unit volume is then given by

$$P^R = k_{ij} \frac{\partial}{\partial x_i} \frac{\partial T_w}{\partial x_j} \quad (20)$$

The radiative flux  $d\Phi_w^R$  applied to the interface within the REV of whole volume  $dV$ , assumed in non equilibrium, is then  $P^R dV$ . When the whole REV is in equilibrium,  $P^R$  given by Eq.20 is applied to the whole volume (with  $T_w = T$ ).

Chalopin et al.[22] have generalized this approach to extinction, scattering and absorption coefficients strongly dependent on direction and for a phase function strongly dependent separately on both the incidence and the scattering directions, for triangular or squared arrays of opaque rod bundles of axis  $Oz$ . The components  $k_{xx}$  and  $k_{yy}$  of the associated diagonal radiative conductivity tensor are equal. Similar results have been obtained in the case of a downgraded nuclear reactor core small scale model[23].

For a porous medium with an opaque phase and a transparent phase,  $k_{ij}$  is given, in the simple case of gray rod walls for the sake of simplicity, by generally diagonal expressions of the type

$$k_{ij} = \frac{\Pi}{A_F} 4 \sigma T_w^3 k_{aij}^+ (\Pi, \text{geometry, reflection law}). \quad (21)$$

where  $\Pi$  and  $A_F$  are the porosity and the specific area per unit volume of the transparent fluid phase and  $k_{ij}^+$  tabulated non dimensional conductivity tensor components. The proportionality of  $k_{ij}$  to the ratio  $\Pi/A_F$  (or to  $\Pi^2/A$  if  $A$  is the specific area per unit volume of the whole medium) is due to the facts that: i)  $\beta^{*-1}$  which appears in Eq.18 is proportional to  $A_F^{-1}$ , as seen in Eq.5; ii)  $k_{ij}$  defined in regards with the whole REV is proportional to  $\Pi$ . It is worth noticing that, if the whole REV can be considered isothermal at temperature  $T$ , the previous expressions of  $k_{ij}$  have to be simply expressed vs  $T$ .

The previous approach can also be applied when a phase or all the phases of a REV are semi transparent or transparent, as long as the validity criterion introduced in the next subsection is separately fulfilled by the considered phase(s). When the two semi transparent phases of the medium (one of them can be transparent) fulfill the criterion, the intensity  $I_\nu$  is strongly dependent on the temperature fields of the two phases, as pointed out in Eq.9. This fact results from the coupling between the two RTEs associated with the two phases, due to the transmission phenomena through the interfaces and established in Ref.[20]. Consequently, for a given phase  $\alpha$ , the extinction, the internal scattering and the reflection phenomena characterized by  $\beta_{\alpha\nu}^*(\mathbf{u})$ ,  $\sigma_{\alpha\nu}^*(\mathbf{u})$  and  $p_{\alpha\nu}^*(\mathbf{u}', \mathbf{u})$  lead to a conductivity tensor  $k_{\alpha\alpha ij}$ . On the other hand, the interfacial transmission phenomena characterized by  $\sigma_{\beta\alpha\nu}^*(\mathbf{u})$  and  $p_{\beta\alpha\nu}^*(\mathbf{u}', \mathbf{u})$  lead to a second conductivity tensor  $k_{\beta\alpha ij}$ . In these conditions, the radiative flux within the phase  $\alpha$  is given by

$$q_{\alpha i}^R = -k_{\alpha\alpha ij} \frac{\partial T_\alpha}{\partial x_j} - k_{\beta\alpha ij} \frac{\partial T_\beta}{\partial x_j} \quad (22)$$

In this equation, the tensorial convention is only applied to the coordinates  $i$  and  $j$ .  $k_{\alpha\alpha ij}$  and  $k_{\beta\alpha ij}$  are generally diagonal tensors, which degenerates in scalars in the case of a statistically isotropic medium. The radiative power per unit volume in the phase  $\alpha$  is then

$$P_\alpha^R = k_{\alpha\alpha ij} \frac{\partial}{\partial x_i} \frac{\partial T_\alpha}{\partial x_j} + k_{\alpha\beta ij} \frac{\partial}{\partial x_i} \frac{\partial T_\beta}{\partial x_j} \quad (23)$$

If the whole REV can be considered at equilibrium, i.e. the temperatures  $T_\alpha$  and  $T_\beta$  are equal to  $T$ , Eq.22 degenerates in a simple tensorial Fourier law.

### 3.2 Validity range and limitations: radiation and nanoconduction

In this subsection, validity criteria of the radiative conduction model are defined by comparison with the limitations of the classical diffusion model for a material system; limitations of the radiative conduction model are also discussed in some cases of practical interest.

Let us first precise the pertinent scale of a REV, such as the perturbation method can be applied. Equation 18 is identical to Eq.16 after substitution of  $u_j(\theta, \varphi)/\beta^* dI_\nu^\circ/dT(T_w) \partial T_w/\partial x_j$  for  $(1-\omega_\nu^*) I_\nu^\circ(T_w)$ . Consequently  $I_\nu^{(1)}$  is a small in front of  $I_\nu^{(0)}$ , as long as  $u_j(\theta, \varphi)/\beta^* dI_\nu^\circ/dT(T_w) \partial T_w/\partial x_j$ , written  $u_j(\theta, \varphi)/\beta^* \partial I_\nu^\circ(T_w)/\partial x_j$  in a more condensed manner, is small in front of  $(1-\omega_\nu^*) I_\nu^\circ(T_w)$ . A practical criterion is issued from the exponential features of the solution:  $u_j(\theta, \varphi)/\beta^* \partial I_\nu^\circ(T_w)/\partial x_j$  has to be 5 times less than  $(1-\omega_\nu^*) I_\nu^\circ(T_w)$ . A practical formulation in nondimensional quantities is then, for all frequencies involved in the application

$$\kappa_\nu^* \delta > 5, \quad \text{Kn}_\nu^a = \frac{1}{\kappa_\nu^* \delta} < 0.2. \quad (24)$$

where  $\text{Kn}_\nu^a$  is the Knudsen number related to absorption.

The model is valid if:

i) the temperature field and all the medium radiative properties can be assumed uniform in the

REV of size  $\delta$  (or in the considered phase within the REV) for calculations at the zero order of perturbation;

ii) the temperature gradient can be considered uniform in this same REV (or same phase within the REV) for calculations at the first order of perturbation.

The criterion is only based on the absorption coefficient, even if the conductivity mainly depends on the extinction coefficient in this model. Indeed, the diffusive transfer is due to a large number of emission-absorption events in the REV, which play the same role as the collisions in a REV of an assembly of molecules for molecular diffusion. The role of the scattering phenomenon is only to make larger an effective absorption path of the medium.

The paradox is that a scattering phenomenon, which does not contribute to the matter element energy balance, can be preponderant in the expression of the radiative conductivity, due to the factor  $\beta^{*-1}$  in the expression of  $I_\nu^{(1)}$ . For instance, in the case of a medium characterized by a phase function close to the isotropy and an albedo  $\omega_\nu$  close to 1, the conductivity is proportional to  $\sigma^{*-1}$  ! It is a common case, encountered for instance for a mullite foam, used in catalytic combustion applications[20, 24] of albedo higher than 0.99. In this strongly scattering medium,  $\beta^{*-1}$  (or  $\sigma^{*-1}$  ) is approximately equal to the pore size, 0.1 to 0.4 *mm* for instance. Consequently, by accounting for Eq.24, the value of  $\delta$  is as high as 500 times the pore size, typically 5 to 20 *cm*. In practical applications, such foams can only be treated as a semi transparent medium by solving the RTE coupled to other transfer modes for all the considered optical thicknesses. The simple conductive model has in practice no physical basis. Indeed, if the temperature fields can be considered as uniform over three times the mean pore size, i.e.  $\eta = 0,3$  to 1, 2 *mm*, the associated absorption Knudsen number  $\text{Kn}_\nu^a = \kappa_\nu^*/\eta$  is equal to 33. This case is close to the ballistic approximation for conduction in nanosystem; it is, in fact, the most common case in radiation.

In other common cases, for which the albedo is less than 0.5, the REV size  $\delta$  is less than 10 times the pore size, i.e. less than 3 *mm* for a typical pore size of 0.3 *mm*. In these conditions the conduction model can be used if the conditions i) and ii) are satisfied.

An interesting case corresponds to a statistically homogeneous porous medium characterized by different separate and nonoverlapping pore size classes. The question is to define which role plays each size class in a radiative transfer calculation. Let us consider, for instance, a material used in combustion which exhibits three size classes, i.e 1  $\mu\text{m}$ , 40 $\mu\text{m}$  and 350  $\mu\text{m}$  with similar probability density functions for each class. If we consider a value of the albedo close to 0.2, the REV sizes  $\delta$  associated to the different size classes are 6  $\mu\text{m}$ , 240  $\mu\text{m}$  and 2.1 *mm*. If the previous conditions i) and ii) are satisfied along a path larger than 2.1 *mm*, the equivalent medium conductivity is practically the one associated with the largest size pores; indeed, the photon mean free path associated with the second class  $l_2$  is 9 times shorter than for the largest size pores and the effect of this class on the conductivity vanishes as  $l_2^{-1}$ . But in the same application, if we focus on the heat transfer within the flame front of typical extension some 100  $\mu\text{m}$ , the equivalent medium associated with the largest pores is optically thin at this scale and the extinction, absorption and scattering coefficients of the equivalent medium are, in practice, those corresponding to the intermediate pore size class of 40  $\mu\text{m}$ . But, for this particular system the radiative conduction model is not valid and the radiative transfer calculations are now based on the classical RTE.

In conclusion, it appears from the previous discussion that a porous medium which can be modeled by a continuous semi transparent medium can only be represented by a radiative conductivity tensor if the precise conditions i) and ii) are satisfied. But, on the other hand, the semi transparent model can not be applied, for instance in the case of foams, for a porosity less than 0.5. This condition often corresponds, in practice, to an optically thick REV. It is intuitive that a radiative conductive model can then be applied. A continuous radiative model generalizing the RTE has to be developed, in order to determine the associated radiative

conductivity tensor on a rational basis. It is today a major research topic.

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