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Thermal conductive and radiative properties of solid foams: Traditional and recent advanced modelling approaches

Propriétés thermiques conductives et radiatives des mousses solides : Approches de modélisation traditionnelles et avancées

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ABSTRACT

The current paper presents an overview of traditional and recent models for predicting the thermal properties of solid foams with open- and closed-cells. Their effective thermal conductivity has been determined analytically by empirical or thermal-resistance-network-based models. Radiative properties crucial to obtain the radiative conductivity have been determined analytically by models based on the independent scattering theory. Powerful models combine three-dimensional (3D) foam modelling (by X-ray tomography, Voronoi tessellation method, etc.) and numerical solution of transport equations. The finite-element method (FEM) has been used to compute thermal conductivity due to solid network for which the computation cost remains reasonable. The effective conductivity can be determined from FEM results combined with the conductivity due to the fluid, which can be accurately evaluated by a simple formula for air or weakly conducting gas. The finite volume method seems well appropriate for solving the thermal problem in both the solid and fluid phases. The ray-tracing Monte Carlo method constitutes the powerful model for radiative properties. Finally, 3D image analysis of foams is useful to determine topological information needed to feed analytical thermal and radiative properties models.

RÉSUMÉ

Cet article présente une vue globale des modèles traditionnels et récents de prédiction des propriétés thermiques et radiatives des mousses solides ayant des cellules ouvertes ou fermées. Leur conductivité thermique effective est déterminée par des modèles empiriques ou analytiques basés sur le réseau de résistances. Les propriétés radiatives nécessaires pour remonter à la conductivité radiative sont déterminées par des modèles analytiques basés sur la théorie de diffusion indépendante. Les approches robustes couplent la modélisation tridimensionnelle (3D) de mousses (par exemple, par la tomographie à rayons X, la mosaïque de Voronoï, etc.) et la résolution numérique des équations de transport. La conductivité thermique due à la phase solide est directement calculée par la méthode des éléments finis (EF), avec un coût de calcul raisonnable. La conductivité

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thermique effective, quant à elle, peut être déterminée à partir des calculs EF combinés avec la conductivité thermique due à la phase fluide. Cette dernière peut être évaluée de façon précise par des formules simples dans le cas de l'air ou d'un gaz faiblement conducteur thermique. Cependant, la méthode des volumes finis apparaît la mieux appropriée pour résoudre le problème thermique, à la fois dans la phase solide et la phase fluide. La méthode de Monte Carlo et de tracé de rayons constitue une approche solide pour calculer les propriétés radiatives. Enfin, la reconstruction d'image 3D des mousses est essentielle pour déterminer les informations topologiques nécessaires pour alimenter les modèles analytiques de conductivité thermique et de propriétés radiatives.

1. Introduction

Cellular foams are a key material for many engineering applications. Their high porosity (or low relative density) and their large specific area play an important role from the thermal point of view [1]. For example, high porosity polymer foams allow realizing efficient insulating materials [2–5]. High-porosity open-cell metallic foams attracted much attention for designing compact heat exchangers and heat sinks [6–8]. Metallic foams filled with phase change materials offer a possibility for heat energy storage applications [9,10]. Thanks to the good resistance to high temperatures and strong chemical corrosion resistance of open-cell ceramic foams, they have been used as combustion support for porous burners [11–15], as catalyst support [16,17], and as volumetric absorbers in solar thermal and thermochemical reactors [18–20]. In all of these examples, the better knowledge of the foam thermal properties is of primary importance. They include thermal conductivity, radiative properties and convection exchange coefficient.

Heat transfer within highly porous cellular materials in the absence of forced fluid flow is essentially due to conductive and radiative heat transfers. Indeed, convective heat transfer within the fluid phase can be neglected since the usual size of the pores is sufficiently small, avoiding the natural convective flow to be significant. The conductive heat transfer may occur through the solid network, by lattice vibration and by charged particles, and through the fluid, by molecular interactions. The radiative heat transfer is by electromagnetic radiation exchanged between solid walls or struts. The combined conductive and radiative heat transfers in cellular materials for most of thermal applications can be characterized by an overall thermal conductivity, expressed as the sum of conduction and radiation contributions,

$$k^*_{\text{tot}} = k^*_{\text{rad}} + k^*_{\text{cond}} \tag{1}$$

where k^*_{rad} and k^*_{cond} refer respectively to radiative conductivity and effective thermal conductivity. The later comes from heat conduction in solid and fluid phases of the foams.

In this paper, we present a survey of the literature describing the state-of-the-art on the modelling of thermal conductive and radiative properties of solid foam materials. Discussion about the thermal convection properties is out of the scope of this work and can be found elsewhere (e.g., in [21,22]). A special attention is paid to recent studies attempting to understand the impact of microstructure on thermal properties of foams.

The paper is divided into four parts. The Section 2 talks about the effective thermal conductivity. The Section 3 focuses on thermal radiative conductivity and especially on radiative properties. The Section 4 discusses the existing models and ongoing challenges. And the last Section 5 is devoted to the conclusion.

2. Effective thermal conductivity

Several approaches have assumed the conductive heat transfers within two-phase systems as a superposition of non-interacting heat flows within the two phases. In this manner, the effective thermal conductivity of a foam material, k^*_{eff}, has been expressed as [23]:

$$k^*_{\text{eff}} = k^*_{\text{s}} + k^*_{\text{f}} \tag{2}$$

The subscripts "s" and "f" refer respectively to the solid- and fluid-phase thermal conductivities. For cellular materials for which the pore size is about a hundred of micrometres or greater, and for weakly thermally conducting gas as fluid, the confined fluid thermal conductivity, k^*_{f}, can be estimated as:

$$k^*_{\text{f}} = (1 - \rho^*)k_{\text{f}} \tag{3}$$

In Eq. (3), k_{f} refers to the bulk conductivity of the gas, i.e. not influenced by the presence of the solid walls. At given pressure and temperature, k_{f} can be calculated using the kinetic theory for gases or deduced from tabulated data (e.g., those given in [24]).

Application of Eq. (3) to foams was initially suggested by Glicksman and Schuetz [23]. As shown in [25], measurements and computation based on finite-volume method (FVM) applied in the solid and fluid phases confirm the suitability of Eq. (3) for highly porous closed- and open-cell foams (porosity greater than 80%) with gas such as air as the fluid substance.

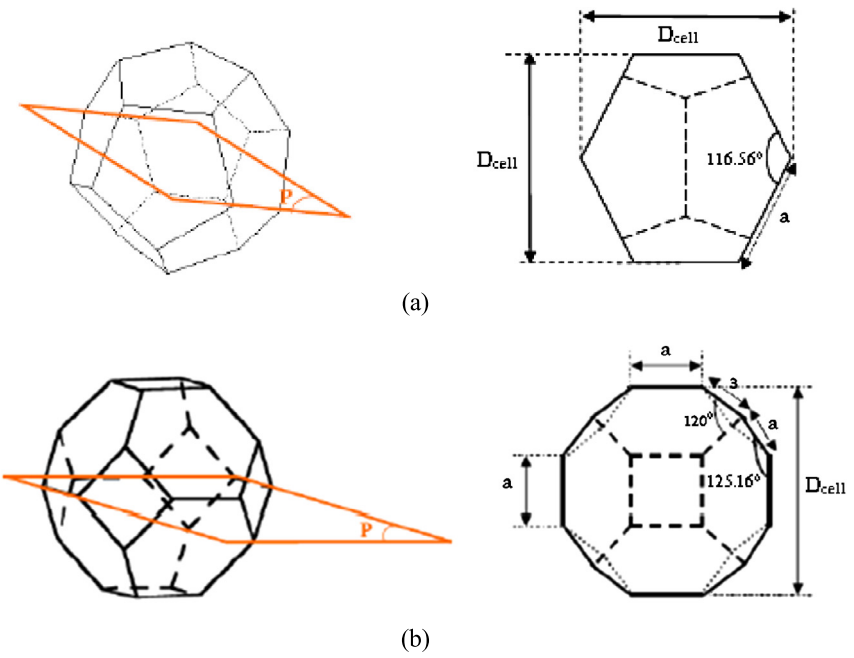

Fig. 1. (Colour online.) Three-dimensional (left) and plane views (right) of regular cells used for modelling foam structures: (a) dodecahedron; and (b) tetrakaidecahedron or Kelvin's cell.

The thermal conductivity of the solid network, k_s^*, in Eq. (2) can be determined analytically by using a drastic simplification of the foam geometry (see the model derivation in [23]); otherwise, it needs to be computed from numerical methods due to the complex structure of cellular materials. It is important to note that in the presence of a highly conducting fluid such as water, the validity of Eq. (2) with Eq. (3) may be questionable [26,27] and there is no simple common relation for computing only the confined fluid thermal conductivity k_f^*. Therefore, common approaches have consisted in determining the effective thermal conductivity k_{eff}^* from numerical simulation.

2.1. Classical models of effective thermal conductivity

Earlier modelling approaches of thermal conductivity of porous materials were conducted by Russell [28]. He derived an analytical relation of effective conductivity from the analysis of the conduction through a solid matrix containing cubic cells (of uniform wall thickness and without struts) arranged inline. As reported in [29], other authors such as Doherty, Maxwell, Eucken, Bruggemann, and Misnar developed other formulas, in which the porosity is the structural parameter, while an empirical parameter has to be determined. Bauer [30] provided a numerical value of Bruggemann's empirical parameter by fitting with measurements. As consequence, all of these models do not permit a clear understanding of foam microstructure effects.

Nowadays, it is known that microstructure may influence heat transfer in solid foams. Therefore, more extensive studies were conducted. The earlier approaches considered that the porous structure of foams is made of cell struts and/or cell windows if the foams have opened cells or not. Several simplifications on the cell morphology such as dodecahedron shape (Fig. 1a), walls with uniform thicknesses, and struts with square cross-sectional areas were applied. By applying a thermal resistance network model, Glicksman and Schuetz [23] obtained an analytical expression of the effective thermal conductivity of high-porosity polymer foams. More recently, Ahern et al. [26] have pointed out that the simplifications used by Glicksman and Schuetz may lead to some errors in the prediction of the effective conductivity. As a consequence, they proposed another model, based on the Maxwell relation for mixtures of materials with different conductivities. Through a numerical study of the effective thermal conductivity of open-cell metallic and closed-cell polymer foams, Coquard and Baillis [27] pointed out that the Ahren et al.' model is more largely applicable to foams than that of Glicksman and Schuetz. However, neither the Glicksman and Schuetz's nor the Ahren et al.' model is able to take into account the concentration of the solid phase at the vertices of struts and the irregularity of cell shape coming from the foaming processes.

It is worth noting that a large number of analytical models aiming to better understand the effect of morphology features of real foams on effective thermal conductivity were developed during the ten last years. The models proposed by Boomsma and Poulikakos [31], Bhattacharya et al. [32] are typical examples. In [31], a more realistic representation of foam morphology was introduced. More particularly, the unit cell was modelled as a 3D tetrakaidecahedron, referred to also as Kelvin's cell (Fig. 1b). To account for the repartition of the solid phase in the struts, cubical (or spherical) nodes of finite size, called lumps, were placed on the vertices of the cells, and cylindrical ligaments were used to join the nodes (Fig. 2a)

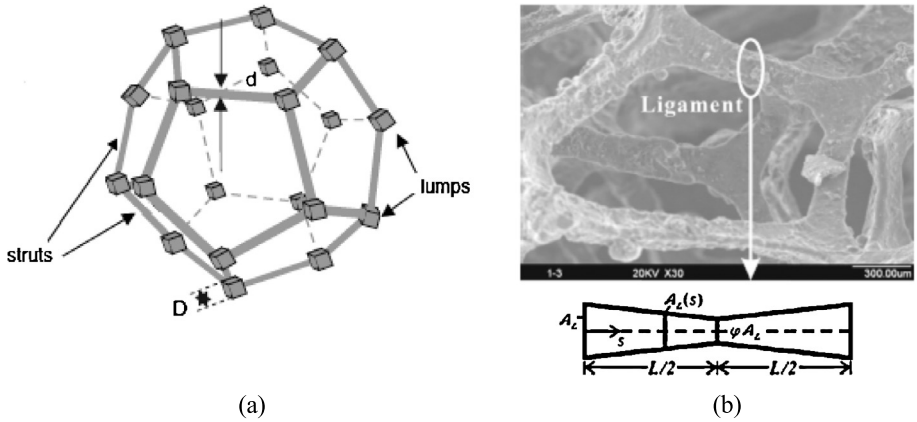

Fig. 2. Repartition of solid phase in open-cell structures. (a) Lumps and struts suggested by Boomsma and Poulikakos [31]; and (b) SEM image and the idealized linear strut cross-section used in Yang et al.' model (taken from Yang et al. [35]).

[31,32]. Alternatively, Singh and Kasana [33] proposed an empirical approach for estimating the effective conductivity of metal foams. Their model was a product of parallel and serial models weighted by power factors, which have been introduced to take into account foam topology. Note that a rich state-of-art of theoretical studies on thermal properties of foams until 2008 was reported by Coquard and Baillis [34]. Therefore, we present only the main recent studies in the next part of this section.

Using a similar theoretical development to that of Glicksman and Schuetz [23], Yang et al. [35] obtained an analytical effective thermal conductivity model for high-porosity open-cell metal foams saturated with air or water. The model assumes a 3D tetrakaidecahedron unit cell as in the work of Boomsma and Poulikakos [31], but introduced struts with cross-sectional areas varying linearly along the strut axis, as depicted in Fig. 2b. It can be noted that such strut representation appears more realistic than cylindrical ligaments with cubic or spherical nodes at ends used in [31,32].

Kumar et al. [36] extended the model of Singh and Kasana [33], based on product of serial and parallel models, to air or water saturated open-cell metal foams. The power factor parameter of the Singh and Kasana' model was adjusted from numerical simulation of conduction heat transfer within a 3D tetrakaidecahedron saturated with a fluid having a wide range of thermal conductivity.

Li and Wang [37] modelled the structure of open-cell ceramic foams using the so-called "body-centred cubic (bcc) unit cell". The unit cell was obtained by subtracting the unit-cell cube from nine equal spheres at the various bcc lattice points. The bcc unit cell allows modelling a non-linear cross-sectional area of struts. These authors adopted the analytical thermal conductivity model proposed initially by Boomsma and Poulikakos [31] but adjusted the strut topological parameter, namely the ratio of the cubic node size (D in Fig. 2a) and the ligament's length from thermal conductivity measurements.

Comparative studies with available numerical (e.g., in [27]) and/or experimental data of thermal conductivities have shown that: (i) there is huge disparity between the results from the above-mentioned analytical models when applied to various available foams; and (ii) each theoretical model is only suitable over a narrow range of porosity. The reasons for the limited applicability range and the disparity of the results predicted from theoretical models have been attributed to the simplifying hypotheses on foam microstructure and physics, and to the use of empirical parameters that cannot be generalized to all foams. Indeed, high-porosity foams do not consist of cubic or spherical cells. Moreover, dodecahedrons or tetrakaidecahedrons constitute a first step for a realist representation of foam cells, but they do not account for the foam irregularity and imperfections. Advanced studies were conducted over these last years for overcoming the weakness of theoretical approaches. Numerical approaches combining realistic (i) representation of the foam microstructure and (ii) modelling of the heat transfer physics appear more appropriate.

Wang and Pan [38] developed a random generation-growth method for modelling open-cell foam structures. They then solved the energy equations in solid and fluid phases over the numerical geometry by using the lattice Boltzmann method (LBM). They deduced the effective thermal conductivity of the equivalent homogeneous medium by applying Fourier's law. However, the random generation-growth method uses several (probabilistic) input parameters, which may be difficult to correlate with microstructural features of foams. Moreover, microstructures obtained from the random generation-growth method appeared qualitatively far from that of real foams, which are neither regular nor fully random.

During these recent years, the Voronoi-based tessellation methods have been applied for generating 3D cellular materials [39–41]. Among the various variants of Voronoi methods, the Laguerre–Voronoi tessellation is the most suitable method enabling a realistic modelling of foam microstructures [42,43]. Indeed, it permits generating large volumes having mean characteristics of cells (angle between linked edges, angle between linked faces, number of faces per cell, number of edges per face, volume or size of cells, etc.) very similar to those of real foams [42–44]. It also enables designing virtual foams with desired (i) cell size or volume distribution [45,46] and (ii) the variation law of strut cross-sectional areas [46,47]. Figs. 3a and 3b depict respectively closed-cell structures obtained by applying the Laguerre–Voronoi algorithm on a random

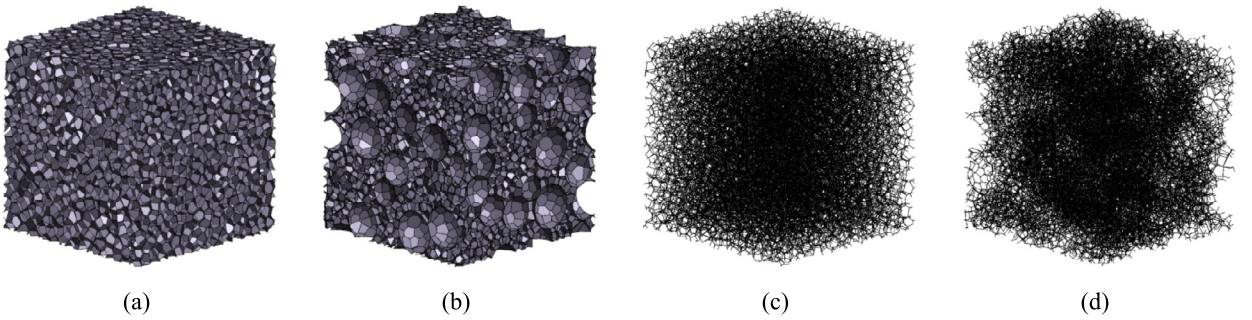

Fig. 3. Laguerre–Voronoi closed-cell and open-cell structures generated from (a) and (c) a random packing of identical spheres; (b) and (d) a random packing of bidisperse spheres of radius ratio of 4 and volume fraction of large spheres of 50%.

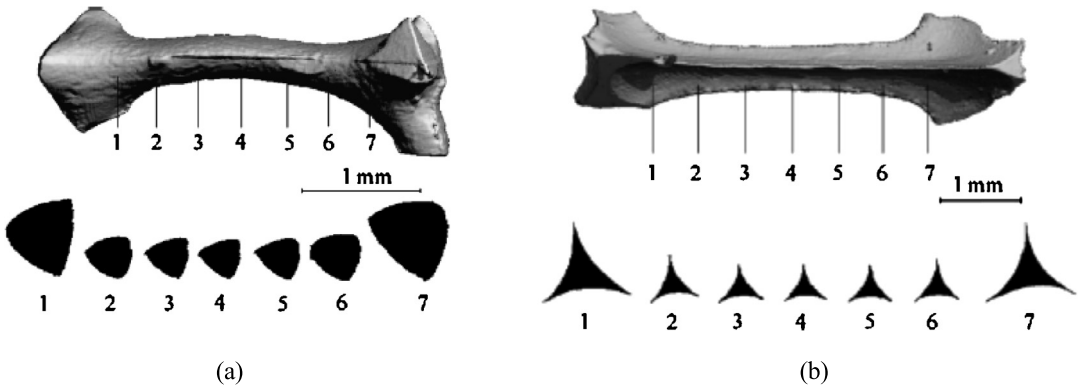

Fig. 4. Typical 3D shape of struts and cross-section views of (a) aluminium and (b) polyester urethane foams (taken from Jang et al. [48]).

dense packing of identical spheres and a random dense packing of bidisperse spheres. For the latter sphere packing, the following parameters were used: a ratio of sphere radius of 4 and a volume fraction of large spheres of 50%.

The skeleton of the open-cell structures consists of edges of Laguerre–Voronoi polyhedrons. Figs. 3c and 3d depict respectively the skeletons of open-cell structures corresponding to the two Laguerre–Voronoi polyhedrons shown in Figs. 3a and 3b. Note that the foam polydispersity is determined by the polydispersity of polyhedrons and, therefore, it can be controlled by playing on the distribution of sphere diameters of the packing. A quantitative image analysis of strut shape and variation of cross sections of open-cell polyester urethane (PU) and aluminium (Al) foams having different pore-per-inch ratios were performed by Jang et al. [48] and more recently by Liebscher et al. [49]. These analyses confirmed that the struts of both metal- (Fig. 4a) and polymer-based foams (Fig. 4b) present a plateau border shape. The cross-sectional area is nearly constant over the central half of the strut length, but increases rapidly when approaching the strut ends. The variation of the cross-sectional area between strut ends can be modelled by a polynomial function [46–49] with coefficients deduced from the image analysis on real foams.

The X-ray micro-computed tomography (μ-CT) imaging has been well developed in the field of materials science and enables nowadays an accurate reconstruction of heterogeneous materials and more particularly of foams (e.g., in [27,42, 50–52]). Fig. 5 illustrates open-cell NiCrAl (Fig. 5a), Al (Fig. 5b) [51], and closed-cell polymer foams (Fig. 5c) [52] obtained from μ-CT image analysis.

2.2. Voronoi-cell-based effective thermal conductivity

Li et al. [53] used the classical Voronoi diagram [54] to model two-dimensional (2D) and 3D closed-cell metallic foams. They then solved the thermal problem within the solid network by using the finite-element method (FEM). In that manner, the numerical model of Li et al. assumed that the thermal conductivity of the foam is only due to heat transfer through the solid phase, i.e. $k^*_{\text{eff}} \approx k^*_{\text{s}}$. The numerical model was applied for studying the effect of the irregularity of the cellular structure on their thermal conductivity. The non-uniformity of the thicknesses of cell walls was modelled by attributing random thicknesses to walls while the imperfection of cell walls was taken into account by deleting cell faces randomly.

At the same time, Randrianalisoa et al. [55] used also the classical Voronoi tessellation for generating 3D closed-cell and open-cell foams with regular and random microstructures. Regular foams consisting of Kelvin's cells were achieved by applying the classical Voronoi algorithm on seed points arranged in bcc lattice while foams with irregular cell shape were achieved by considering a random distribution of seed points. As in Li et al.'s works, they deduced the solid network thermal conductivity, k^*_{s} in Eq. (2), from FEM simulation results. The effective thermal conductivity of the cellular structure

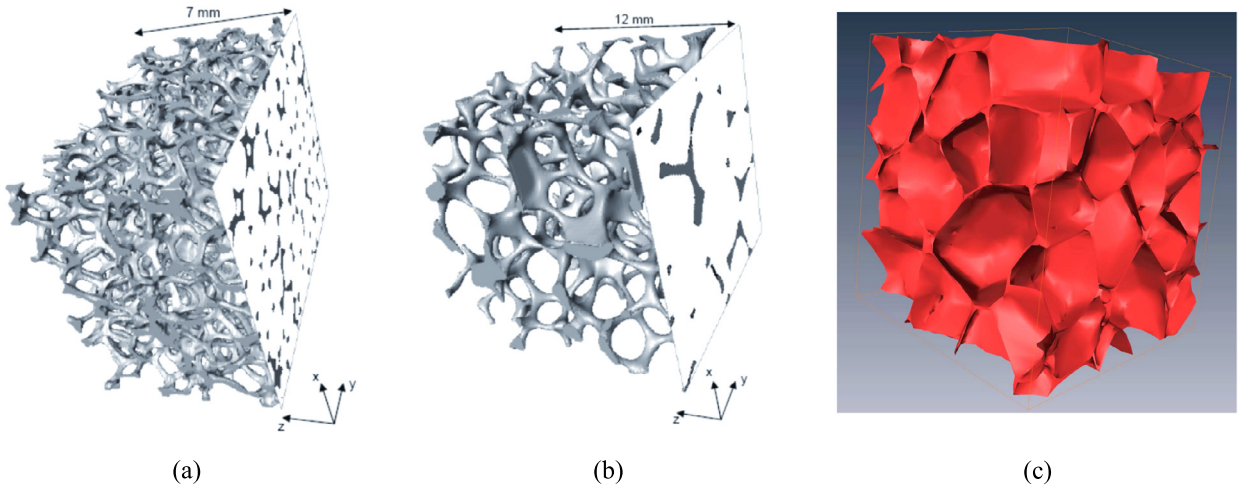

Fig. 5. (Colour online.) 3D open- and closed-cell foams obtained from μ-CT imaging. View and grey level tomographic slice of (a) NiCrAl and (b) Al foams (taken from [51]); and (c) closed-cell polymer foam (taken from [52]).

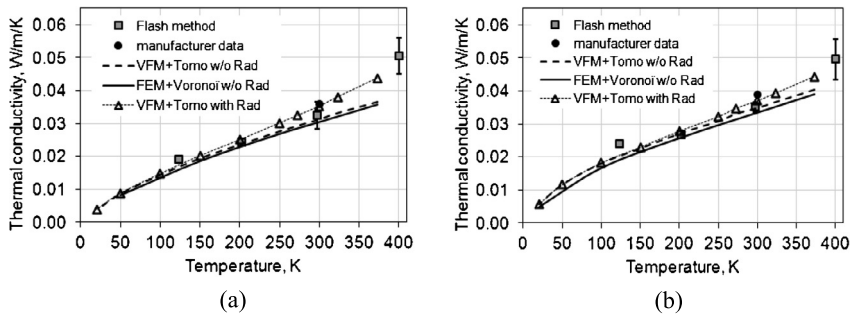

Fig. 6. Thermal conductivity of closed-cell PVC foams of (a) 94.3% and (b) 90.5% porosities. Symbols: measurements; dash curves: FVM computation on μ-CT samples without radiation contribution; solid curves: FEM computation on random Voronoi cells without radiation contribution; dot curves with symbols: FVM computation on μ-CT samples with radiation contribution.

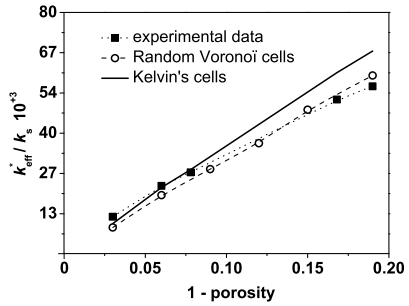

Fig. 7. Room-temperature thermal conductivity of open-cell metallic and ceramic foams versus relative density (= 1 – porosity). Experimental data contain those of Al [32], NiCrAl, mullite, and YSZ zirconia foams [27].

was obtained through Eqs. (2) and (3) by taking into consideration the contribution of the solid and fluid phases. The computed effective thermal conductivities of random closed-cell polymer foams of porosities 90.5 and 94.3% were found in good agreement with (i) measurements in the temperature range 50–300 K in which radiation's contribution is insignificant, and (ii) computation by using the FVM on μ-CT foam samples over a wide temperature range between 50 and 370 K (see Fig. 6). On the other hand, open-cell metallic foams were shown to be better modelled by random Voronoi cells than by Kelvin's cells, especially for the porosity range 80–85% (see Fig. 7).

Kanaun and Tkachenko [56] proposed a method combining the 3D Laguerre–Voronoi method for modelling open-cell foam structures and FEM for solving the energy equation within the solid phase. A similar approach was recently used by Randrianalisoa et al. [46] for investigating the effects of the microstructure on effective thermal conductivity of high-porosity open-cell foams (porosity greater than 80%). More particularly, they studied the effects (i) of the presence of two distinct

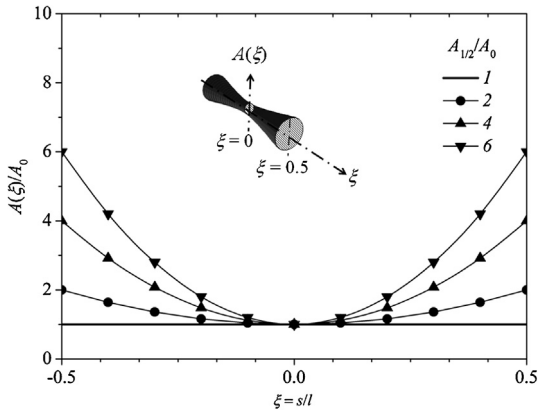

Fig. 8. Axial variation of strut cross-section for different ratios of areas at ends, $A_{1/2}$, and mid span, $A_{0,}$ considered in [46].

cell sizes within the samples, and (ii) of the non-uniformity of the cross-sectional area along the strut axis. Based on the finding of Jang et al. [48], the struts' cross sectional area variation was assumed symmetric with respect to the mid span and followed a quadratic law (Fig. 8).

2.3. Computed X-ray tomography structure-based effective thermal conductivity

Another advanced modelling approach uses 3D numerical microstructures obtained from μ-CT imaging. Coquard and Baillis [27] developed a numerical model combining 3D imaging of foam samples and FVM for solving heat flow within both solid and fluid phases. They stated that such approach permits to compute the effective conductivities of various closed-cell polymer foams and open-cell metallic foams by taking into account as faithfully as possible their porous structure. Later, the above numerical approach was applied by Coquard et al. [57] for deriving a new analytical model of effective thermal conductivity of open-cell foam materials by using only the topological information obtained from image analysis and the thermal conductivity of bulk components as input parameters.

Fiedler et al. [58] combined 3D numerical foams from μ-CT reconstruction and the Lattice Monte Carlo (LMC) method for predicting effective thermal conductivity of open-cell (M-pore) and closed-cell (Alporas) aluminium foams with stochastic pore morphologies. More recently, Fiedler et al. [59] applied the LMC approaches to study the effective thermal conductivity of the so-called "advanced pore morphology" (APM) foam, which was modelled as a periodic arrangement of spherical porous elements obtained from μ-CT.

Bodla et al. [60] developed a resistance network-based thermal conductivity model for metal foams. The model used essentially topological information obtained from μ-CT data. As an example, the nodes of the resistance network are the vertices of foam skeleton. The model was solved numerically by matrix method due to the large number of thermal resistances involved.

Mendes et al. [61] used an approach similar to that of Coquard et al. [27] (i.e. a combination of FVM and 3D μ-CT samples) to determine the effective thermal conductivity of open-cell ceramic and metal foams. They then derived four analytical models expressed as a weighting between minimum and maximum thermal conductivity bounds. The minimum and maximum bounds included respectively the serial and parallel models, Hashin and Strikmann's (HS) lower and upper bounds, effective medium theory (EMT) and parallel model, and EMT and HS upper bound. The weighting factor has been shown to be dependent on the structure of the porous medium, but was adjusted from full numerical simulation on μ-CT foams or measurements. These analytical models were recently extended by Mendes and co-workers [62] to a foam saturated with an arbitrary working fluid such as highly inflammable gas, products of partial oxidation, etc., or liquid metals, where performing measurements remains very challenging.

3. Radiative conductivity

A considerable effort of investigation has been concentrated on the radiative properties of porous materials over the decade. An overview of radiative properties of highly porous foams can be found in the monograph by Dombrovsky and Baillis [63], review papers such as those by Sacadura and Baillis [64], and recently by Baillis et al. [65]. The radiative properties may be theoretically predicted and/or identified from Fourier Transform Infrared (FTIR) measurements. However, this section focuses essentially on the predictive models of radiative properties of cellular foams with high porosities (porosity $>$ 80%).

3.1. Rosseland approximation

The radiative conductivity of a foam, k^*_{rad} in Eq. (1), is generally calculated by using the Rosseland approximation [34,66]:

$$k^*_{\mathrm{rad}} = \frac{16\sigma_{\mathrm{B}} T_{\mathrm{m}}^3}{3\beta_{\mathrm{R}}} \tag{4}$$

where T_{m} is the mean temperature in the medium, σ_{B} is the Stefan–Boltzmann constant and β_{R} is the Rosseland average extinction coefficient. As scattering of thermal radiation in foams is globally anisotropic, a spectral transport extinction coefficient, $\beta^{\mathrm{tr}}_{\lambda}$, is usually introduced in the calculation of β_{R} [63].

The Rosseland average extinction coefficient is defined as follows:

$$\frac{1}{\beta_{\mathrm{R}}} = \frac{\int_{\Delta\lambda} \frac{1}{\beta^{\mathrm{tr}}_{\lambda}} \frac{\partial I^0_{\lambda}(T_{\mathrm{m}})}{\partial I^0(T_{\mathrm{m}})} \mathrm{d}\lambda}{\int_{\Delta\lambda} \frac{\partial I^0_{\lambda}(T_{\mathrm{m}})}{\partial I^0(T_{\mathrm{m}})} \mathrm{d}\lambda} \tag{5}$$

where $I^0(T_{\mathrm{m}})$ and $I^0_{\lambda}(T_{\mathrm{m}})$ are respectively the total and spectral blackbody intensity at the temperature T_{m} · $\beta^{\mathrm{tr}}_{\lambda} = \kappa_{\lambda} + \sigma_{\lambda}(1 - g_{\lambda})$ is the "transport" extinction coefficient, in which g_{λ} is the spectral asymmetry factor of scattering. The integrals are performed over the meaningful wavelength interval of width, $\Delta\lambda$.

As a result, the radiative conductivity k^*_{rad} requires the determination of spectral radiative properties, κ_{λ}, σ_{λ} and $g_{\lambda} = \int_{-1}^{+1} P(\mu)\mu \mathrm{d}\mu$, which depend on the nature of the foam, the microstructure, the cell size, the solid wall roughness, and the porosity. $P(\mu)$ refers to the scattering phase function [63].

3.2. Radiative properties from classical independent scattering theory

First radiative properties models for open- and closed-cell foams were usually developed assuming idealized homogeneous cellular structures such as regular polyhedrons and independent scattering theory. The latter states that: (i) "*the radiative properties of the foam are the sum of the individual contributions of all walls and struts present in a unit volume*;" and (ii) "*walls and/or struts are randomly oriented and positioned within the foam volume*" [63–65].

Closed cell foam Different types of closed-cell polymer foam used for thermal insulation were studied in the literature, such as extruded (XPS) and expanded polystyrenes (EPS), and polyurethane (PUR) foams. Glicksman and Schuetz [67] modelled the radiative properties of XPS and PUR foams by representing their microstructure as pentagonal dodecahedrons (Fig. 1a). They apply independent scattering theory to the random arrangement of opaque struts with triangular cross-sections and partially transparent windows (the cell walls). They obtained an analytical expression of the transport extinction coefficient $\beta^{\mathrm{tr}}_{\lambda}$, which depends on the cell diameter and the porosity of the foam.

Placido et al. [3], Kaemmerlen et al. [5], and Kuhn et al. [66] proposed more complex radiation models for XPS and PUR foams. They modelled the porous structure by assuming dodecahedral cells composed of cylindrical struts and pentagonal windows. To tackle the extinction of the radiation by struts, they used the Mie–Kerker scattering theory or the discrete dipole approximation (DDA) applied to a circular cylinder (the strut). In this manner, the struts were not systematically treated as opaque to radiation, as in [67]. These authors considered the cell walls as optically smooth, infinitely large platelets of uniform thicknesses and randomly oriented throughout the entire porous volume. They then applied the geometric optics approximation (GOA) to compute the extinction by each platelet, since the dimensions of cell walls were much greater than the radiation wavelength considered. The radiative properties of the foam model were obtained by applying the independent scattering theory along with the radiative properties of each wall and strut present in the unit volume.

Coquard et al. [4,68] developed a predictive model for the radiative properties of EPS foams. In such materials, the amount of matter at the vertices of the cells was shown to be negligible and the solid phase was assumed entirely contained in the cell walls. The authors studied the influence of the cell shape and considered the porous materials made of cubic cells, dodecahedrons, or tetrakaidecahedrons. For cubic cells, square walls with constant thickness were assumed, whereas pentagonal windows and hexagonal/square windows were considered for dodecahedral and tetrakaidecahedral cells, respectively. The radiative properties of the foams (with square or pentagonal or hexagonal windows) were computed by analysing the interaction of a plane incident wave with a randomly oriented plane particle (the cell wall). The complete model was validated by comparing the spectral hemispherical reflectance and transmittance measurements with predicted values for various samples.

Open cell foams Baillis et al. [69–71] modelled the radiative heat transfer in open-cell carbon foams. They considered that these foams consisted of randomly oriented struts with triangular cross-sections and of heaps of matter corresponding to the volume intersection of four linked struts (Fig. 9). They computed the radiative properties of each strut and their juncture by using a combination of geometric optics laws and of diffraction theory. Then, the radiative properties of the foams were computed by applying the independent scattering theory.

Loretz et al. [72] predicted analytically the radiative properties of open-cell metal and ceramic foams by using similar simplifications of physics as those made by Baillis et al. [69–71], i.e. they used the independent scattering theory and GOA.

Fig. 9. Model of (a) strut and (b) strut juncture proposed by Baillis and Sacadura [69].

However, particular attention was paid to microstructure analysis and to its modelling. Indeed, these authors considered different kind of cells shapes and strut cross-sections, retrieved from scanning electron micrograph (SEM) and μ-CT image analysis. Coquard et al. [57,73] used the analytical model derived by Loretz et al. [72] for studying the coupled conductive and radiative heat transfer in metallic foams at fire temperature.

Zhao et al. [74] proposed an alternative approach based on the evaluation of radiosities. Radiation characteristics in open-cell metallic foams, in terms of emissivity, reflectivity and view factors, were described. Their model assumed a simple cubic cell as representative volume and predicted the trend of the experimentally measured apparent conductivity-versus-temperature curve, although the predicted conductivity was, in general, lower than that measured.

3.3. Radiative properties from ray-tracing Monte Carlo method

Previous theoretical studies include the determination of the radiative properties of highly porous foams (κ_λ, σ_λ and $P_\lambda(\mu)$) for simplified geometries composed of pentagon dodecahedron, tetrakaidecahedron or cubic unit cell. As already evoked in Section 2, the architecture of high porosity foams is more complex than that of these regular cells. Consequently, during the last ten years, there has been growing interest in drawing a more realistic morphology of the porous structure in modelling radiative transfer. It can be noted also that in models described in Section 3.2, it was assumed that independent scattering occurs even though the particles (walls and struts) forming the foam are close to each other. Thus, in order to increase the accuracy of the radiative models, the Monte Carlo (MC) method was developed. Indeed, the MC is well suitable to model the radiation propagation within complex structures, but also permits to overcome the possible failure of independent scattering assumptions.

Open cell foams In the works of Loretz et al. [51], highly porous metallic foams were characterized by using μ-CT. The images were processed to retrieve the morphological features required for the calculation of the radiative properties. The spectral extinction coefficient, $\beta_\lambda = \kappa_\lambda + \sigma_\lambda$, was then determined from two approaches. In the first one, the resulting morphological features were used as the input data of the conventional independent scattering theory. In the second approach, an alternative method is proposed in order to determine the extinction coefficient from the tomographic images without any calculation or assumption. The results show a good agreement with the extinction coefficient obtained from experimental measurements.

Zeghondy et al. [75,76] applied the ray-tracing Monte Carlo (RTMC) method to the porous structure of mullite foams obtained from μ-CT imaging in order to compute their radiative properties. They used the Radiative Distribution Function Identification (RDFI), originally proposed by Tancrez and Taine [77]. Petrasch et al. [78] applied the RTMC procedure to reticulate porous ceramics for calculating the extinction coefficients and scattering phase functions on the developed probabilistic distribution functions of the extinction path length and the directional cosine of the incident radiation. These pertinent previous studies, based on tomographic representations of foams, were intended to improve the accuracy of computation of radiative properties. However, in these works, some assumptions were still made concerning notably the nature of radiation reflections occurring at the solid/fluid interface. Moreover, the numerical models used in these studies were not experimentally validated. More recently, Coquard et al. [57,79], proposed to use an alternative RTMC method combined with tomography image analysis to compute the radiative properties of metallic open-cell foams. They evaluated and highlighted the improvements brought by the use of 3D tomography imaging in comparison with previous analytical models. Their RTMC method was initially proposed by Coquard and Baillis [80] and later improved by Randrianalisoa and Baillis [81]. This method, originally developed for particle beds, was extended to tomographic images of several Al foams. The reflection occurring at the solid/fluid interface is estimated using the measured roughness of the surface evaluated by stereoscopic SEM. A comparison between measured and computed hemispherical transmittances and reflectances has shown that the agreement is quite satisfactory and demonstrates the superiority of the numerical modelling combining μ-CT images and RTMC method.

Closed cell foams Few pertinent numerical studies on radiative properties of closed-cell foams were done. Coquard et al. [52] applied the RTMC method developed for open-cell foams [79] to 3D polymer foams obtained from μ-CT (Fig. 5c). For validating the radiation heat transfer models, the Rosseland conductivity was first deduced from computed spectral radiative properties of the foam samples. It was then combined with the effective thermal conductivity, computed using the FVM on

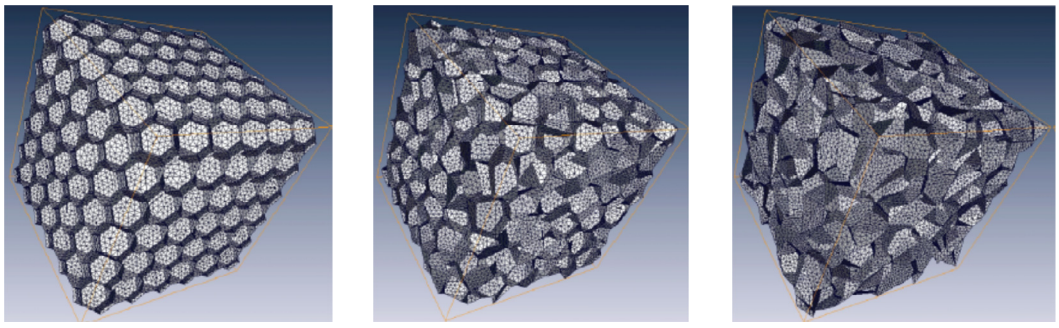

Fig. 10. (Colour online.) 3D renderings of meshed volumes generated using the classical Voronoi tessellation method and having different cell size distributions: (a) $\sigma_{cell}/D_{cell} = 0$, (b) $\sigma_{cell}/D_{cell} = 0.074$, and (c) $\sigma_{cell}/D_{cell} = 0.14$ (taken from Coquard et al. [82]).

the same numerical samples. The resulting apparent thermal conductivity was compared to measurements over a wide temperature range (50–373 K), as depicted in Fig. 6. The agreement between computed and measured apparent thermal conductivity is remarkable, proving the accuracy of the combined numerical conduction and radiation models.

Very recently, Coquard et al [57] applied the RTMC method to 3D volumes of closed-cell structures generated by using the classical Voronoi tessellation method (Fig. 10). They analysed the influence of structural parameters such as the porosity, the mean cell size (D_{cell}) and their standard deviation (σ_{cell}). Different foam morphologies were considered ranging from a periodic assembly of Kelvin's cells (Fig. 10a) to an assembly of random cells (Figs. 10b and 10c). The evolution of the radiative properties of samples was discussed in term of cell randomness. The calculated hemispherical transmittances and reflectances of plane parallel polymer samples are compared with (i) the corresponding measurements from FTIR spectrometer; and (ii) the literature data obtained from the calculation, but based on μ-CT images of the foam samples [82].

4. Discussion and current challenges

4.1. Foam structure modelling

Intensive researches in the fields of X-ray tomography and image processing have permitted a very realistic reconstruction of heterogeneous materials and especially foams. In heat transfer applications, 3D μ-CT samples enable obtaining accurate results in terms of thermal properties, and temperature and heat flux fields as well. However, those samples do not allow a parametric study of foam microstructure features (porosity, cell size, wall and/or strut thicknesses, etc.) on their thermal properties. Therefore the rescores to flexible foam structure models is necessary. The Laguerre–Voronoi tessellation carried out on a dense packing of spheres is a potential candidate for modelling and designing highly porous foams.

The analysis of reconstructed 3D samples obtained either from μ-CT or Laguerre–Voronoi method permits to determine the topological information needed to feed analytical thermal models.

4.2. Thermal modelling

Although analytical models with empirical parameters have been developed, they seem not appropriate for designing the foam's structure and thus optimizing the material's thermal behaviour. On the other hand, analytical models derived by using a thermal resistance network, for thermal conduction, and independent scattering theory, for thermal radiation, can be used as a straightforward approach for the material design and optimization provided that they use input parameters representative of the foam topology.

Concerning the numerical modelling of thermal conduction, FEM has been widely used. However, the finite element (FE) analyses have been globally carried out on the solid network alone due to unreasonable cost of computation when the high volume fraction of the fluid phase is also taken into account. In the presence of air or weakly conducting gas as fluid phase, the FE results combined with the contribution of fluid conductivity, given simply by Eq. (3), leads to accurate prediction. Alternatively, the FVM have been shown to be well suitable for analysing thermal conduction in the solid and fluid phases. This is probably due to the fact that FVM requires less computational resource than FEM to reach the same accuracy for thermal conduction problem in foams.

Concerning numerical computation of radiative properties, the RTMC method has been the most adopted due to its capability to handle complex geometries and to avoid the independent scattering hypothesis used in analytical models.

4.3. Current challenges

So far, the solid phase of most of produced foams was considered as a homogeneous medium in thermal models. Recently, production processes have permitted to produce foams with dual-scale porosities [83]. They have a classical foam

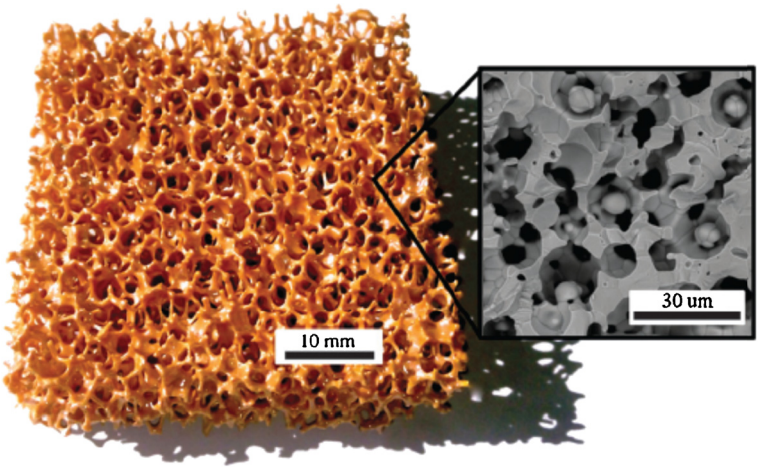

Fig. 11. (Colour online.) Cerium dioxide foam with dual-scale porosities (taken from Furler et al. [83]).

morphology, but struts are also porous, with open- or closed micro-porosity, as depicted in Fig. 11. Such ultra-high porosity materials are favourable for weight saving and for achieving intense chemical reactions, heat exchange, and solar radiation absorption.

Current challenges concern the foam modelling as well as thermal modelling. Indeed, 3D structure reconstruction requires high-resolution tomographs and/or a combination of transmission electron microscope (TEM) and X-ray tomography techniques.

Existing thermal models have to be extended to deal with this multi-scale high porosity foams. As far as the thermal conduction is concerned, the conductivity of struts and walls would differ significantly from that of the bulk material. So it has to be determined and then can be used in existing models. The determination of the conductivity of struts and walls can be done by an identification method based on measurements and numerical calculation, which assumes struts and walls as homogeneous medium.

For the radiative properties' calculations, the difficulty occurs when struts or walls are not opaque to the considered radiation, as with polymer or ceramic foams. In this case, struts and walls themselves are absorbing-scattering media. Existing analytical models may be questionable and improved analytical models and RTMC approaches have to be developed to determine the foam radiative properties κ_λ, σ_λ and g_λ.

5. Conclusion

Thermal conductive and radiative properties of highly porous foams have been investigated over the last decades. Theoretical models are very useful to investigate the influence of the topological parameters characterizing the microstructure of foams on their apparent thermal conductivity and to optimize the material thermal performances. The first studies, which tried to predict heat transfer in such complex materials, used drastic simplifications, and some noticeable improvements have been achieved in the recent years. The following conclusions can be drawn:

- analytical thermal models are straightforward approaches for material design and optimization but should be fed with topological information obtained from advanced foam modelling (μ-CT or Laguerre–Voronoi method as an example);
- although computationally expensive, numerical models (FVM, FEM, or RTMC method coupled with 3D foam structures) permit to obtain reference results. They constitute the alternative approach when analytical models are questionable or inexistent;
- further studies will focus on the improvement of existing approaches to deal with new kinds of foams, such as those with dual-scale porosities.

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