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Thermal Radiative Properties of Complex Media: Theoretical Prediction Versus Experimental Identification

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In many engineering applications and natural phenomena, thermal radiation interacts with complex media composed of dispersed phases that may be of different type: solid/solid, solid/gas, or liquid/gas. Most of them are semitransparent media that emit, absorb, and scatter thermal radiation. Heat transfer by combined radiation with conduction or convection in such media is a problem of high practical importance, mostly in situations where radiation is a dominant mode. Improvement of thermal performance of such materials or of the manufacturing processes that involve these media requires the availability of efficient methods (i) for radiative transfer modeling, and (ii) to predict and to experimentally determine the thermophysical properties intended to feed the models. This paper is focused on radiative properties assessment. After a brief overview of the materials and properties of interest, the emphasis is put on methodology of property investigation combining both theoretical prediction and experimental identification. Examples related to different particulate media are presented, showing recent advances and needs for further investigation.

INTRODUCTION

Thermal radiation is an important and even predominant mode of energy transfer in many engineering applications and natural phenomena. Most of them involve semitransparent media that may be not only absorbing/emitting materials but also scattering media due to their dispersed structure. Examples of these media are very common and numerous. They include solid/solid systems like nonporous ceramics, surface pigmented coatings, or solid/gas systems (media composed of fibers, foams, or micro-sized powders, as most insulating materials are, textiles, food products, fluidized and packed beds, combustors, catalytic reactors, and soot), or liquid/gas systems like bubbling media, and sprays. Examples are shown in Figure 1.

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Improving thermal performance of these media or efficiency and quality of the manufacturing processes involving them requires the capability of modeling the radiative heat transfer inside these media and the assessment of the radiative properties in order to feed the models. Owing to the significant progresses acquired in numerical methods over the two past decades, the prediction of heat transfer by combined radiation with conduction or convection in most industrial applications is nowadays possible through direct models handled by using commercial codes in which the radiative transfer equation is commonly solved by different families of methods, of which some are numerical, other ones semi-analytical [1]. A remaining problem is the assessment of radiative properties. It is an important topic that attracts continuously a great deal of interest from the radiative transfer community, as shown by the reviews of Viskanta and Mengüç [2] and Baillis and Sacadura [3] dealing with radiation transfer in particulate media. This paper is focused on theoretical prediction and experimental determination of semitransparent media radiative properties. After a brief recall of the properties of interest, the emphasis is put on methods of property investigation, both predictive and experimental. Some examples are presented showing recent advances and needs for
further investigation. The aim of this overview is not to provide a comprehensive reference list. Instead we prefer to focus on relevant works and reviews where the reader can find more references.

THEORETICAL BACKGROUND: RADIATIVE TRANSFER EQUATION AND RADIATIVE PROPERTIES

At the basis of radiative transfer calculations is the choice of a direct model that may be of a different kind and level of sophistication according to several criteria: complexity of the problem of interest (medium, radiation source, and boundary conditions), aim of the calculation and accuracy expected, time and effort allocated to the investigation, and also the experience of the investigator. Modeling radiation transfer inside particulate may be achieved by continuous or discrete approaches.

Continuous Approaches

The continuous approaches used in engineering applications are mostly based on the radiative transfer equation (RTE). The particulate medium is modeled as a random distribution of particles and the scattering and absorption properties of the medium are based on those of the discrete particles. In some cases the particles are embedded in a host medium that may significantly contribute to the absorption of the global medium. Independent particles are based on those of the discrete particles. In some cases the particles are greater than particle size and wavelength.

Practical data on scattering regimes are available in the literature in terms of the particle size to wavelength ratio $x = \pi d/\lambda$, called the particle size parameter or diffraction parameter, where $d$ is the particle diameter and $\lambda$ the wavelength, the ratio of particles clearance to wavelength $c/\lambda$, and $f_\nu$, the particle volume fraction, which is related to the ratio $cld$. It is commonly recognized (see the independent and dependent scattering map from Tien and Drolen [4]) that dependent scattering effects may be neglected when $f_\nu < 0.006$ or $c/\lambda > 0.5$.

This is the case in most engineering applications, excepting situations involving very small agglomerated particles such as soot, nanosphere insulations, and other nanoparticles that are currently receiving an increasing interest. Dependent scattering is also present in densely packed beds of particles larger than wavelength.

The radiative properties are used in RTE, which governs the conservation of the radiation spectral intensity $I_\lambda$ at wavelength $\lambda$, and along a direction $\Omega$. Excepting situations involving very fast radiative sources like in laser pulse experiments, which are beyond the scope of this paper, the RTE may be considered in its stationary form and written as:

$$\frac{dI_\lambda(\Omega)}{ds} = -(\sigma_\lambda + \kappa_\lambda) I_\lambda(\Omega) + \kappa_\lambda I_{\beta\lambda},$$  
$$+ \frac{\sigma_\lambda}{4\pi} \int I_\lambda(\Omega') P_\lambda(\Omega' \rightarrow \Omega) \, d\Omega'$$

where $s$ is the space coordinate, $\sigma_\lambda$ and $\kappa_\lambda$ are the scattering and absorption spectral volumetric coefficients, respectively, and $I_{\beta\lambda}$ is Planck’s blackbody emission function, which involves the real part $n_\nu$ of the refractive index $m_\nu$ of the medium. $P_\lambda (\Omega' \rightarrow \Omega)$ is the spectral scattering phase function and is proportional to the probability that the radiation propagating in a direction $\Omega'$ is scattered in a direction $\Omega$. $d\Omega'$ is an infinitesimal solid angle associated to the direction $\Omega'$. Note that, alternatively, the RTE may be and is often expressed in terms of two other spectral volumetric parameters, the extinction coefficient $\beta_\lambda = \kappa_\lambda + \sigma_\lambda$, and the single-scattering albedo $\omega_\nu = \sigma_\nu/\beta_\nu$, instead of $\kappa_\lambda$ and $\sigma_\lambda$.

This option does not affect $P_\lambda$. Thus, the radiation properties of the pseudo-continuous medium are four: $n_\nu$, $\kappa_\nu$, $\sigma_\nu$ (or $\beta_\nu$, $\omega_\nu$), and $P_\nu (\Omega' \rightarrow \Omega)$, and even more, depending on the number of shape parameters used in the phase function representation. The classical representation of the scattering phase function uses an expansion in Legendre polynomials series.

One should note that a significant complexity is added to the analysis in case of a nonisotropic medium, which means that its radiative properties depend on the direction of radiation incident onto each elemental volume of the medium. The phase function $P_\nu$ especially depends on both directions $\Omega'$ and $\Omega$. An important contribution to the subject is found in the pioneering papers by Houston and Korpela [5] and Lee [6–8] dealing with
fiber radiative properties. When the medium may be assumed as isotropic the phase function is expressed in term of the angle \( \Theta \) between the directions of incident and scattered radiation at a scattering location. In engineering applications the media are often assumed as isotropic. However, even with this assumption the phase function generally varies with \( \Theta \). In this case it is referred as anisotropic phase function.

As a number of particulate media exhibit anisotropic phase functions, often sharply peaked, this requires a high-order expansion and, subsequently, an impressive number of coefficients is needed to define the phase function from a polynomial series expansion. Therefore it is generally preferred to use an approximate phase function based on smaller number of shape parameters. Among useful models, the Henyey–Greenstein phase function, which only involves one shape parameter, as referred as anisotropic phase function.

\[
P_{HG}(\Theta, g_s) = \frac{1 - g_s^2}{(1 + g_s^2 - 2g_s \cos \Theta)^{1/2}}
\]

where \( \Theta \) is the angle between the directions of incident and scattered radiation at a scattering point and

\[
g_s = \frac{1}{2} \int_0^{\pi} P_{\lambda}(\Theta) \cos(\Theta) \sin(\Theta) d\Theta
\]

The \( P_{HG} \) model enables one to represent scattering patterns exhibiting just one peak, even sharp, forward or backward directed. For more complex patterns, Nicolau et al. [9] proposed a weighted combination of two HG functions plus an isotropic component:

\[
P_{HG}(\Theta, g_{1s}, g_{2s}, f_{1s}, f_{2s}) = f_{2s} [f_{1s} P_{HG}(\Theta, g_{1s}) + (1 - f_{1s}) P_{HG}(\Theta, g_{2s})] + (1 - f_{2s})
\]

where \( f_{1s} \) and \( f_{2s} \) are the weights associated to the scattering components \( P_{HG}(\Theta, g_{1s}) \) and \( (f_{1s} P_{HG}(\Theta, g_{1s}) + (1 - f_{1s}) P_{HG}(\Theta, g_{2s})) \), respectively. This model using four shape parameters is capable of representing a scattering pattern involving a forward peak and a backward peak and an isotropic component.

For applications requiring the use of less complicated scattering phase functions, simpler approximations are available. Among other simple descriptions of scattering phase functions, when usable, are delta-M, linear anisotropic, and isotropic scaling approximations (see for instance Siegel and Howell [10]). The last one is also called the transport approximation, as it was been first used in neutron transport theory. It is based on a phase function \( P_{\lambda}(\mu_0) = 1 - \mu_1 + 2\mu_1 \delta(1 - \mu_0) \) where \( \mu_0 = \cos \Theta \), \( \mu_1 = g_s \), and \( \delta \) is the Dirac function. This phase function is adequate for media exhibiting a forward scattering peak. Transport approximation provides a reduction of the RTE to the form corresponding to isotropic scattering, with a transport scattering coefficient \( \sigma_i^0 \), or a transport single-scattering albedo \( \omega_i^0 \), and a transport optical thickness \( \tau_{\lambda 0}^0 = \beta_i L \), instead of \( \sigma_i \) or \( \omega_i \), and \( \tau_{\lambda 0} = \beta L \), respectively. The forms of the scaled parameters depend on the reduced model adopted for the solution of the RTE [11–13]. A considerable amount of work was achieved by Dombrovsky [14–16] and by Dombrovsky and co-workers [17–19] aiming to develop analytical solutions that may be used in rather simple way for both radiative heat transfer calculations and radiative properties identification.

As an alternative to RTE, the radiation diffusion approximation, which is similar to \( P_{1} \) approximation, is used by Petrov and co-workers as a direct model for the identification of radiative properties [20].

Scaling correlations applied to radiative properties were suggested by authors aiming to extend continuous approaches to situations involving dependent scattering. This aspect is treated later (see Densely Packed Spherical Particle Beds section).

**Discrete Approaches**

In some cases like dense particulate media, when independent scattering may not be assumed, or for large-scale cellular media [21], the continuous approach of radiative heat transfer may not be appropriate and discrete approaches based on the geometrical optics approximation (see next section) are preferred. Among these, Monte Carlo methods are among the most popular [22]. They were initially used as a reference model aimed to validate other discretization based methods. Due to the increase of computing power and to the implementation of different techniques of variance reduction aimed to save computation time, like the radiation reciprocity principle, for instance, the use of MC as a solving tool for problems of radiative transfer in participating media is continuously increasing [23–26]. A recent review on the application of MC methods to radiative transfer in semitransparent media and on recent progress with reciprocal MC methods is available in [27]. Nowadays, MC methods are commonly used for theoretical calculations of radiative properties [28–32]. This aspect is developed in the next section.

**RADIATIVE PARAMETERS DETERMINATION METHODS: THEORETICAL PREDICTION VERSUS EXPERIMENTAL IDENTIFICATION**

The radiative properties of particulate media may be either theoretically predicted or experimentally determined. Theoretical prediction provides a better insight on the influence of morphology and optical constants of the particle and matrix materials on the radiative properties of the medium. Thus it opens tracks toward designing new material with customized properties. Conversely, experimental identification provides the knowledge of real material properties. Identification may also be used as a means to verify the validity of the choice of the particle property prediction model. Nevertheless, owing to the number of parameters involved, radiative properties identification probably ranks among the most intricate identification processes.
Theoretical Prediction

The theoretical prediction of the radiative properties of disperse media is based on solutions of Maxwell’s equations applied to the electromagnetic wave/particle interaction, which consists of elastic scattering (that means without change of wavelength) and absorption by the particle. An initial issue for the problem was the simple theory for spherical particles much smaller than the radiation wavelength, derived by Rayleigh during the second part of the 19th century. Then Mie published in 1908 a solution of Maxwell’s equations for an electromagnetic wave traveling through a nonabsorbing medium hosting a single embedded sphere. Mie’s scattering theory, which is also referred as Lorentz–Mie theory in recognition of Lorentz’s contribution to the field, remained for a long time the only issue available for the calculation of the radiative properties of dispersed media. Details of derivation of Mie’s solution and data on radiative properties of various particles derived on the basis of Mie’s theory can be found in textbooks by Kerkher [33], van de Hulst [34], and Bohren and Huffman [35]. Simpler solutions apply in some limiting regions depending on particle size parameter $x$ and complex refractive index $m$ values [36]:

$$x \ll 1 \text{ and } |m - 1| \ll 1 \quad \text{Rayleigh scattering.}$$

$$x \gg 1 \text{ and } |m - 1| \gg 1 \quad \text{Geometrical optics and diffraction theory.}$$

$$|m - 1| \ll 1 \text{ and } x |m - 1| \ll 1 \quad \text{Rayleigh–Debye–Gans (RDG) scattering.}$$

$$x \gg 1 \text{ and } |m - 1| \ll 1 \quad \text{Anomalous diffraction.}$$

Mie’s theory was then extended to several situations such as nonspherical particles (among which are infinitely long cylinders under oblique incidence [37], coated [15], inhomogeneous [17, 38], externally multilayered particles [39], absorbing host medium [40–45]). Several cases are treated in papers by Dombrovsky [14–16] and Dombrovsky and co-workers [17–19]. A recent paper by Wriedt provides a useful overview on the subject [46].

For a medium that hosts a population of embedded particles, if the independent scattering is assumed, the contributions of the different particles are simply additive and the radiation properties of a monodispersed population are deduced from the properties of a single particle multiplied by the number of particles per unit volume. For polydispersed particles the single-particle solution is averaged over the distribution of particle size, and in the case of fibrous media it should also be averaged over the particle angular distribution. Important contributions to such fiber properties calculations are found in papers by Lee and co-workers [47–50] and Jeandel and co-workers [51–53], among others. Thanks to its successive developments, Mie’s theory remains a most frequently used tool for particulate media property calculation.

Geometrical optics laws are used for the property derivation through approaches assuming independent scattering. The particle geometry is generally simplified: spheres, cylinders, and ellipsoids [54–57]. Another family of methods is based on Monte Carlo approaches that enable one to more easily account for the real morphology of particles and for some near-field dependent scattering effect, such as the so-called RDFI method by Taine and co-workers [28, 29, 58, 59] that was successfully used by Petrasch, Steinfeld, and co-workers [30, 31], and the approach proposed by Coquard and Bailleux for opaque particle beds [32]. The last authors extended their approach to semitransparent particles, as reported hereafter in section Densely Packed Spherical Particle Beds.

Situations in which independent scattering may not be assumed cannot be adequately treated by Mie’s theory. This is the case for densely packed beds of particles or particle aggregates like soot or nanoparticle thermal insulations, for instance. Nanoparticles, individually, satisfy the Rayleigh scattering approximation. But they combine into chains of aggregates (which, fortunately, may be described as mass fractals) that may contain several dozens or hundreds of primary particles. The modeling of the absorption and scattering by these aggregates requires that the interaction of the electric field components of electromagnetic waves scattered by each individual sphere be accounted for. Therefore, a number of theoretical and numerical techniques for handling the elastic scattering by mutually interacting particles of different sizes, including aggregates, started to be developed in the second part of the 20th century. More or less approximate techniques are available. Some are very simple, like the Rayleigh–Debye–Gans fractal approximation (RDG-FA) frequently used for soot aggregates [60–62]. Others, as in the pioneering work of Tien and co-workers [63–65], account for dependent effects through a near-field factor and a far-field coherent addition correction that are applied to the Mie’s solution. These corrections were derived by solving an equation proposed by Jones [66] for the internal field of a particle interacting with several other neighboring particles. This solution, which is approximate, assumes a statistical pair-particle distribution function. Several pair distribution functions are discussed in [63–65]. Methods based on direct numerical solutions of Maxwell’s equations are receiving increasing attention, due to their capability of handling particles of complex shape that may be separated or aggregated. Numerical methods in electromagnetic theory are reviewed in a recent paper by Kahniert [67]. Among the most currently used methods are the so-called T-matrix [68–70] and discrete dipole approximation (DDA) [71–74]. Note that a free DDA computing code, referred as DDSCAT, is available via the Internet [74]. Much effort has been expended in recent years by the electromagnetic scattering community in the development of tools that may also be used in the radiative heat transfer field. Recent books by Mishchenko and co-authors [75–77] and Mishchenko’s tutorial paper [78] are valuable sources of information on the subject. A review by Manickavasagam and Mengüc [79] is also available. In a recent article by Dombrovsky, various theoretical approaches for the determination of particle radiative properties are overviewed [80].
Experimental Determination

The experimental identification of the radiative properties is a stepped approach that requires appropriate choices of:

i. A direct model of radiative transfer and related solution model, for instance, RTE solved by DOM or another same level of sophistication method, or simpler equations as transport equation or diffusion equation that may be solved by semi-analytical methods.

ii. An experimental configuration.

iii. A set of parameters to be identified/theoretically predicted and the operating sequence.


Steps (i) to (iii) are obviously not independent. Making right choices is essential for a good identification strategy.

The experimental techniques available for the identification of the radiative properties consist of spectral and directional measurements of reflectance and/or transmittance. Emittance measurements have also been used.

The experimental configurations that may be used are schematically represented in Figure 2 (from Moura et al. [81]):

a. Collimated beam nearly normally incident onto the sample, bidirectional transmittance and reflectance measurements.

b. Collimated beam with different angles of incidence onto the sample, hemispherical transmittance measurements; hemispherical reflectance also possible for single nearly normal incidence direction.

c. Diffuse radiation onto the sample, bidirectional transmittance and reflectance measurements.

d. Measurements of bidirectional radiation flux emitted by a hot sample (self emission).

A few examples of such facilities are displayed in Figures 3 to 5. Figure 3 shows an apparatus enabling spectral bidirectional transmittance and reflectance measurements, developed by Nicolau et al. [9] and Moura et al. [81] from an initial concept by Sacadura et al. [82]. It consists basically of a Fourier transform infrared (FTIR) spectrometer coupled to a bidirectional attachment on which is the sample, with its front surface located on the rotation axis of the device, and the detector standing on the rotating arm. In situations where bidirectional transmittance measurements may be acquired with a relatively thin angular resolution in the neighborhood of the direction of the incident collimated beam, it provides a concentration of experimental data in a region, which is suitable for the identification of properties of forward peaked scattering materials [57].

Figure 4 provides a scheme of a golden-coated integrating sphere, also used as a FTIR spectrometer attachment, which enables directional–hemispherical transmittance or reflectance measurements. This sphere is an item of commercial equipment in which the direction of incidence is fixed to a configuration nearly normal to the sample. One should note that directional–hemispherical measurements are rather simple and faster to acquire than bidirectional. They may be performed by a moderately trained operator. On the other hand, bidirectional data acquisition involves long and careful measurements proceeded by a delicate alignment of the setup, both requiring a highly experienced operator.

Figure 5 shows the facility developed by Lopes et al. [83] for spectral directional emission measurements at high temperatures. Heating isothermally up to high temperature a sample that may be a poor thermal conductor, without placing it inside a furnace, is a real challenge. A possible issue consists of simultaneously heating with equal power incident fluxes both sides of a thin sample, by using lasers or arc lamps. This was adopted by Lopes et al. [83, 84] in the facility shown on Figure 5.

Making appropriate choices in the identification approach, mostly for steps (i) and (ii), requires some previous knowledge of the material composition and structure (the latter one may be acquired from microscopic observation, x-ray tomography, and other material investigation techniques) and of the behavior of the radiation parameters under investigation, in particular...
their rate of anisotropy, which may be obtained from the theoretical prediction models of radiation properties. Therefore, in practice both approaches, identification and theoretical prediction, are in fact frequently combined and may be considered as complementary.

The most commonly used experimental configurations consist of spectral measurements of bidirectional transmittance and/or directional–hemispherical reflectance and transmittance. Directional–hemispherical measurements are performed with commercial integrating sphere attachments for spectrometers, which are designed to operate at a fixed direction of incidence of radiation, generally nearly normal to the sample. Emittance measurements have also been used [85, 86]. Several methods enable one to achieve the identification, such as Gauss linearization, the Levenberg–Marquardt method, and genetic algorithms. Reviews on the subject are given by Baillis and Sacadura [3] and Sacadura and Baillis [87]. The last reference provides detailed description of the parameter identification procedure based on Gauss linearization, as well as of the experimental setups used that are illustrated here in Figures 3 and 4.

Whatever will be the identification procedure adopted, one should remember some principles that are summarized here:

The search is spectral, as the experimental tool of property investigation is the spectral radiation traveling through a sample in a narrow band around a given wavelength. The interaction of radiation with the material structure varies with the ratio of...
wavelength to structure size. This means that the contribution of micro- or nanostructure of the material to the radiative properties varies along the wavelength spectrum. Therefore, the choice of an adequate physical model depends on the wavelength range of interest. As shown in the preceding section, some physical models are only adequate for limited regions of the thermal radiation spectrum. As a consequence, different physical models may be necessary to cover the whole wavelength range of interest. Different experimental configurations may also be needed; in a spectral range where the sample is not too much optically thick, transmittance may be used. Conversely, configurations based on reflectance or emittance should be preferred for wavelength ranges in which the sample is optically thick and the radiation measured mostly comes from a thin surface layer.

The smaller are the number of parameters to identify, the easier will be the operation. The set of parameters governing the RTE is \( n_\lambda, \kappa_\lambda, \sigma_\lambda \) (or \( \beta_\lambda, \omega_\lambda \)) and \( P_{\lambda} (\Omega' \rightarrow \Omega) \), that is, three plus the number of scattering phase function parameters. Generally this is too many, owing to the ill-posed nature of the inverse problem to be solved and the uncertainty (noise) that affects the measurements, mostly the bidirectional ones. Thus, the probability of success of the identification depends on a well-suited strategy. Usually \( n_\lambda \) is available from separate measurements and is assumed as known, but this may not be the case for some materials. In order to reduce further the number of parameters to identify, one can combine theoretical prediction for some of them with experimental identification for others. In some cases and for given wavelength ranges the absorption of the material may be mostly due to the matrix and the scattering due to the imbedded particles, or conversely. This suggests the possibility of determining separately the absorption and scattering properties.

One should note that the choice of \( (\beta_\lambda, \omega_\lambda) \) instead of \( (\kappa_\lambda, \sigma_\lambda) \) as identification parameters is not without consequence. Although the first pair is frequently used, an identification based on \( (\kappa_\lambda, \sigma_\lambda) \) may be more convenient as it would retrieve directly and separately the absorption and the scattering parameters. This may have a consequence on the issue of the identification process. We believe that this subject requires further investigation.

\( P_{\lambda} \) is generally the most difficult to identify due to its possible complexity, which may require bidirectional measurements. But if some knowledge is available on the material and its morphology (thanks, for instance, to scanning electron microscopy (SEM)), the scattering phase function may be theoretically estimated from one of the above listed methods. If the shape of \( P_{\lambda} \) is compatible with a reduced model (requiring no more than one shape parameter), its identification may be tried with an increasing probability of success. Note that if the design problem for which the radiative properties are needed may be solved by a simplified RTE model, like the transport model, for instance, this means that a very simple definition of \( P_{\lambda} \)—isotropic in this case—may be used for the properties identification. However, in each case it is convenient to get an estimation of the inaccuracy introduced by the simplification. On the other hand, one should also estimate the experimental errors as they may drastically limit the use of more sophisticated models.

**EXAMPLES OF DISPERSED MEDIA AND PROPERTY DETERMINATION**

**Anisotropic Media: Fibers**

Most fibrous materials, like highly porous glass-wool insulation, for instance, show an anisotropic structure resulting in nonisotropic radiative properties. This is a source of complexity for property identification which:

- Requires experiments that allow one to vary the direction of incidence of radiation onto the sample (specially developed laboratory setups).
- Does not allow the use of simplified direct models of RTE.

Nevertheless, when the diameter and orientation distributions of the fibers as well as the refractive index of the material are available, which is often the case, the radiative properties may be computed from theoretical models based on an extension of Mie’s theory [33, 37]. Several papers by Lee and Cunnington [49, 50] and Jeandel and co-workers [51–53] provide a good background on the subject. In such situations the experimental identification of radiative properties should be rather considered as a validation means for the theoretical models established on the basis of an independent scattering assumption. Identification may also be useful when there is not enough data available for a theoretical computation of properties. As for sake of simplicity their identification is generally based on the assumption of isotropic properties, this may result in some inaccuracy when properties determined from normal incidence experiments are then used in design calculations involving diffuse irradiation of the material [14].

However, the question is, “When and for what purpose are detailed anisotropic properties really necessary?” Clearly, even in highly anisotropic materials, after traveling some depth inside the medium, anisotropy of radiative properties is attenuated due to an averaging effect from multiple scattering. As practical applications of insulating materials concern relatively thick layers, this means that predictions and/or identification of radiative properties based on simple models such as the transport model, for instance, or on experiments that do not allow varying the direction of incidence of radiation, probably are satisfactory for most engineering applications. A few situations, however, may require more detailed investigation of the real anisotropy of the properties. The radiative behavior of thin surface layers of material submitted to high external incident fluxes and/or internal radiative source profile showing important variation close to the surface layer are among these situations.

Radiative properties of fibrous media used as thermal insulation, mostly low-density media, have long been investigated through both theoretical modeling [6–8, 47–49, 88] and
experimental identification \[9, 82, 88, 89\] . These authors mostly used numerical solutions of the RTE by the discrete ordinates method combined with inverse approaches based on the Gauss linearization method to determine each of the unknown parameters.

The work of Moura et al. \[81\] provides an attempt at systematic methodology for this kind of media. The four experimental configurations shown in Figure 2 are discussed. In the first three cases, the emission term is not considered in the RTE since the experiments use modulated radiation incident onto the sample, combined with a phase-sensitive detection technique. The direct model is solved by a differential discrete ordinates method for a one-dimensional plane slab with or without azimuthal symmetry. The phase function model is a combination of two Henyey–Greenstein (HG) functions coupled with an isotropic component as suggested by Nicolau et al. \[9\] (Eq. 3). A study of condition number, as a function of the optical thickness, is carried out in order to determine the ability of each configuration to identify radiative properties. For each configuration and sample thickness \( l \), the direct model described earlier in which a set of data is assumed for the material radiative properties \( \tau_0 = \beta l \), \( \omega \), \( \varphi_1 \), \( \varphi_2 \), \( \varphi_3 \), \( \varphi_4 \) is used to calculate the theoretical transmittances, reflectances, or emittances, all referred as \( T_l(\theta, \varphi) \). The experimental bidirectional transmittances and reflectances, \( T_{ed}(\theta, \varphi) \) for an incident radiation, the experimental hemispherical transmittance \( T_{eh} \), and the experimental directional emittance \( \varepsilon_{ed}(\theta, \varphi) \), are defined by the following expressions:

\[
T_{ed}(\theta, \varphi) = \frac{i(\theta, \varphi)}{i_o d\omega_o}
\]

\[
T_{eh} = \frac{\int_{0}^{2\pi} i(\theta, \varphi) \cos \theta d\Omega}{i_o d\omega_o}
\]

\[
\varepsilon_{ed}(\theta, \varphi) = \frac{i(\theta, \varphi)}{i_b}
\]  

(5)

where \( i \) is the transmitted, reflected, or emitted intensity, \( i_o \) is the intensity of the beam incident onto the sample within a solid angle \( d\omega_o \), and \( i_b \) is the blackbody emission intensity.

For the cases (a) and (b), \( d\omega_o \) depends on the experimental device; for case (c), \( d\omega_o \) is the half-hemisphere \( (2\pi) \).

The identification of a radiative parameter \( \hat{\chi}_k \) is based on a minimization of the quadratic error between the measured, \( T_{m} \), and calculated, \( T_{n} \), transmittances, reflectances, and emittances over the measurements:

\[
F(\hat{\chi}_k) = \sum_{n=1}^{N} \left[ T_{m,n}(\hat{\chi}_k) - T_{n,n} \right]^2 \quad k = 1, \ldots, K
\]  

(6)

where \( K \) is the number of parameters to be identified.

In cases (a), (c), and (d) the summation, Eq. (6), is performed over the different bidirectional measurements. In case (b) the summation is over the hemispherical transmittance and reflectance measurements for different angles of incidence onto the sample.

The method adopted in the reported work to achieve this minimization is the Gauss linearization method that minimizes \( F(\hat{\chi}_k) \) by setting to zero the derivatives with respect to each of unknown parameters \( \hat{\chi}_k \). As the system is nonlinear, an iterative process is performed over \( m \) iterations \[9\]. It involves a matrix \( S \) that is composed of the sensitivity coefficient products, calculated from the theoretical model and does not directly depend on the experimental values. This matrix, \( S \), can be used in the sensitivity analysis to verify possible linear dependences between the sensitivity coefficients calculated for each parameter. The calculation of a condition number \( CN \) of this matrix can be used to determine the degree of ill-posedness of the identification problem \[90\],

\[
CN(S) = \|S^{-1}\| \cdot \|S\|
\]  

(7)

where the norm \( \|S\| \) is calculated from the elements \( S_{k',k} \), as:

\[
\|S\| = \max_{k'=1,K} \sum_{k=1}^{K} S_{k',k}
\]

Of course, the larger the condition number is, the more ill-conditioned the system is: Small changes in the measurements result in very large change in the solution vector, i.e., the increments \( \Delta \hat{\chi}_k \). It is then almost impossible to simultaneously determine all of the unknown parameters. Poor conditioning occurs when at least two of the sensitivity coefficients are quasi-linearly dependent or when at least one is very small or very large compared to the other.

The investigation was carried out for an optical thickness \( \tau_0 \) ranging from 0.1 to more than 20 and a set of data for the other radiative properties shown in Table 1.

These data correspond to the phase function represented in Figure 6, which is both forward and backward peaked.

This detailed investigation provided useful information on the capabilities and weaknesses of the different experimental configurations and inversion strategies for handling the radiative parameters identification.

Independent scattering assumed for the direct models of radiative transfer in fiber media is realistic for low-density insulation. But this may not be acceptable for high-density fibrous materials, such as those used for industrial furnace insulation. An issue was recently proposed by Coquard and Baillis \[91\], who extended successfully to dense fibrous media a model based on geometrical optics and Monte

<table>
<thead>
<tr>
<th>Radiative property data used in [81]</th>
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<tr>
<td>( \omega )</td>
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<td>( g_1 )</td>
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<td>( f_1 )</td>
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<td>( g_2 )</td>
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Due to the growing difficulty of the radiative transfer measurements when samples are heated, most experimental identifications of radiative property of fibrous media are performed at room temperature even for materials that may be used for high-temperature insulation. Recent reports by Zhang et al. [92] and Zhao et al. [93] are focused on the determination of high-temperature properties of fibrous insulation. Instead of radiative measurements, this approach is based on effective thermal conductivity obtained from thermal conduction setup allowing both steady-state and transient thermal tests on large samples (180 mm diameter, 20 mm thickness). An inverse approach accounting for the combined conduction and radiation heat transfer inside the sample is used to simultaneously estimate three "gray" radiative properties (extinction, albedo, coefficient of a linear assumed phase function) plus a thermal conductivity due to the gas and solid conduction. Obviously, the radiative properties determined by this approach are global and depend on the temperature profile inside the medium.

Recent contributions from Tagne and Baillis [94, 95] and Dombrovsky et al. [96] were dedicated to the development of methods of property investigation based on transport approximations or isotropic scaling that provide simple analytical means of property identification. The last group of authors developed a modified two-flux solution for the transport model aiming to account for collimated irradiation of the medium, as is the case in some specific radiative property identification experiments. They also investigated the limits of this solution by comparison with numerical solutions obtained from high-order DOM calculations. Application to fibrous media was examined by Tagne and Baillis [95]. Owing to their features, these simplified models are obviously not able to retrieve properties like the phase function shown in Figure 6 that displays both forward and backward peaks. But the real question is: Do the transport extinction coefficient and transport albedo experimentally retrieved by using such simplified models provide sufficiently accurate predictions of radiative transfer if the predictions are achieved through the same models? If the goal is just to provide a calculation of the divergence of the radiative heat flux to be input in the energy equation, as is the case for a number of engineering applications, the response probably is "yes." Conversely, if a good estimate of the radiative flux is expected, more accurate determination of the radiative properties may be necessary.

**Bubbles, Hollow Microspheres in A Host Semitransparent Medium**

A number of recent studies were motivated by bubbles generated during the industrial glass melting process or similar structures with bubbles or hollow microspheres hosted in a semitransparent matrix, as in some advanced thermal insulation materials. One of the first theoretical investigations on radiative properties of glass with bubbles is due to Fedorov and Viskanta [97], who used the anomalous diffraction approximation. Then Pilon and Viskanta [98] performed calculations based on the same approximation. A Mie's solution was then proposed by Dombrovsky [15]. For moderately anisotropic properties, including the phase function, the use of transport model for identification is not so critical. Therefore, it was frequently used in recent works dealing with experimental identification of the radiation properties of fused quartz containing bubbles [19, 99]. The last group of authors, Dombrovsky et al. [19] proposed an improved identification procedure of the infrared properties of this material from experimental measurements of directional–hemispherical transmittance and reflectance and by using the transport approximation. A modification of the two-flux approximation accounting for the Fresnel interface reflections at sample interfaces was used to analytically solve the transport equation [96]. Note that this study also provided an identification of the volume fraction of bubbles. The error of this modified two-flux approximation combined to the transport model of phase function has been estimated for an arbitrary scattering medium by comparison with numerical calculations using the discrete ordinates method combined to Henyey–Greenstein phase function. It appears to be not greater than 5% in the most important range of problem parameters. Again, measurements of hemispherical reflectance and transmittance performed on fused quartz with bubbles and radiation properties identified from these measurements were used by Randrianalisoa et al. [100] as tools for comparing predictions from CMT, FFA, and NFA models. The transport model was also used to identify the properties of porous zirconia ceramics [17].

**Foams**

Optimizing the thermal performance of foams is currently an important industrial challenge. A considerable effort of investigation has been concentrated on the subject over the decade; see for instance references [55–57, 101, 102], among others. These
investigations covered not only the radiative transfer through the structure but also the solid and conductive heat transfer, as heat transfer inside foam involves different coupled mechanisms. For other dispersed materials the radiative properties may be theoretically predicted and/or identified from FTIR measurements using techniques described in earlier sections. Polymer or metallic foams are structured into cells, SEM photos of these families of foams show reticular or cellular structures. In reticular structures the cells are open, the material is concentrated on struts and junctures, and the interstitial gas is the same as the external environment, which is typically air. Cellular structured foams have closed cells whose walls are thin membranes. Struts and junctures appear at cell edges and corners as membrane thickenings. The internal gas may be different from air and may contribute to increase the insulation effect. According to the bulk material used and to the manufacturing process, foams show more or less complex morphology and cells may have a single- or double-scale structure. Good descriptions of insulating foams as well as recent reviews on the topic may be found, for instance, in [102, 103]. As the mean diameter of cells is generally larger than the wavelength range of interest, the calculation of radiation properties may be performed by using geometric optics and/or Mie theory to describe the interaction of radiation with the elements of the foam structure.

Densely Packed Spherical Particle Beds

Beds of particles are commonly used in many industrial applications. Due to the weak clearance between particles, mostly in packed beds, the scattering phenomena are typically in the domain of dependent scattering. The modeling of radiative properties of beds of spherical particles has received much attention. Among other methods, correlations have been used by Kamiuto [104] and Kamiuto et al. [105], and scaling factors by Kaviany and Singh [106], and Singh and Kaviany [107], to approximate dependent scattering effects. Dombrowsky derived approximate models for the radiation properties of packed hollow microspheres. In one of these models the clearances are treated as randomly oriented spherical particles [16]. Coquard and Baillis [32] developed a Monte Carlo approach to predict the radiative properties of beds of large opaque particles that may be diffusely or specularly reflecting spheres. The bed is generated as a set of randomly positioned particles. The porosity is an adjustable parameter enabling one to represent particles from those that are very distant to touching ones. The procedure uses a large number of rays starting from random points in the bed. Each ray is tracked along its path in the bed; it may undergo several reflections and partial reflections at the surface of the particles before leaving the computation domain. All these phenomena are accounted for, and this provides, for a great number of rays, the radiative properties of the bed. This model, for which results are in good agreement with experimental measurements, is a new promising tool to predict radiative properties of beds of spheres independently from particle clearance. It also contributes to the assessment of the limits of independent scattering theories. Then Coquard and Baillis [108] extended this model to beds of semi-transparent spherical particles. The material inside each particle is assumed to be an absorbing and scattering medium. Independent scattering from particles is assumed. This model may be helpful to investigate the radiative behavior of complex particulate media. However, due to the time required by Monte Carlo calculations it does not seem appropriate for identification. The so-called radiation distribution function identification (RDFI) method is a Monte Carlo technique recently developed by Tancrez and Taine [28], and extended by Zeghony et al. [29], for the determination of radiative properties of high-porosity media made of solid particles embedded in a transparent fluid. The radiation propagates through the fluid medium and is partially reflected and absorbed when it hits a particle surface, according to the reflection model adopted and related laws. The radiative properties are identified from cumulated distribution functions of radiation free path inside the medium, and cumulated probabilities of absorption and directional reflection by the particle–wall interfaces. The strength of this method is that it only requires the knowledge of the real medium morphology and local radiative properties at the particle/host medium interface. This method was successfully applied to several porous materials, such as mullite foam, [58], rod bundles (for radiative transfer simulation in a nuclear reactor core [59]), and packed beds of reticulated porous ceramics that may be used as radiant absorbers for high-temperature solar thermochemical processes [30, 31].

Agglomerated Nanoparticles

Due to their promise for future applications, a growing interest is observed for the understanding of thermal radiation mechanisms involving nanoparticles that are generally agglomerated in more or less chained clusters, as is the case for soot or nano-powder thermal insulations. For other media, the investigation of radiative properties involves experimental identification and/or theoretical prediction, with the latter requiring one to account for possible dependent scattering effects. Revisiting the models of Tien and co-workers [63–65], Prasher derived two approximate solutions for Rayleigh regime that account for multiple scattering, or multiple plus dependent scattering, respectively [109]. Both derivations are based on the so-called Percus–Yevick pair distribution function, considered the most successful distribution for capturing the effects of depending scattering. The last approximation, referred as the quasi-crystalline-approximation (QCA), captures much better the right physics than the independent scattering approximation. In recent works the DDA method is used for radiative property calculation. Lallich et al. [110, 111] investigated the radiative properties of silica nanoporous matrices that are components of superinsulating materials. The samples contain agglomerates of approximately 80 to 200 primary particles of diameter ranging from 7 to 14 nm. The experimental identification of
the transport extinction coefficient and transport albedo was achieved from spectral directional–hemispherical transmittance and reflectance measurements, by using the transport equation as the direct model and the Newton–Raphson algorithm for the inverse problem. The authors also calculated theoretically the same properties by several methods: (i) Mie’s theory, (ii) Mie’s theory with three different dependent scattering corrections from Tien and co-workers and Prasher, and (iii) DDA, and they compared the identified with the theoretical results. As expected, Mie’s theory based on a proper representative size of the agglomerate appeared as predictive enough of radiative properties for wavelengths beyond 1 μm. For lower wavelengths (250 nm to 1 μm) the independent scattering model as well as the dependent scattering corrected models failed to match the experimentally identified parameters. Conversely, DDA predictions showed a better agreement with the experimental results. Note that in this work still in progress the DDA calculations were based on computer-generated aggregates rather than on the real aggregate structure as it may be investigated by transmission electronic microscopy (TEM) or other means of structure investigation. Another recent work deserves to be noticed [112]. It deals with the use of microwave analogy to experimentally characterize aggregated particles. It is not easy to perform light scattering measurements on arbitrarily shaped micro- and nanoscale particles due to the impossibility of tailoring them, but operating with larger particles, and subsequently larger wavelength, makes it possible, through analogy between thermal radiation and microwave ranges. Magnitude and phase of the field scattered by aggregates of dielectric spheres of diameter 1.59 cm (Figure 7) were both (which is original) measured and the results were successfully compared with theoretical predictions from DDA and T-matrix models [113]. This analogy seems to be a promising tool to investigate not only the scattering behavior of aggregated particles, especially small-sized ones, but also the validity of radiative property identification strategies.

**OPTIMIZATION ALGORITHMS**

The reader may be disappointed by the lack of information reported in this paper on the selection of optimization algorithms aimed to the radiation property identification. Several algorithms were used for that purpose, and sometimes compared. Some are deterministic (Gauss, Levenberg–Marquardt [114], conjugate gradient, and Tikhonov’s regularization); other ones are statistical, like genetic or particle collision algorithms, for instance, or hybrid methods coupling an evolutionary/stochastic approach with a deterministic one [115, 116]. In some works, two or three arbitrarily selected methods are compared in terms of accuracy/computational effort, but a lack of justification or strategy of choice is generally observed. In the case of identification of temperature, heat flux, radiative source, or conductive properties for semitransparent materials, a number of works are available in the literature that compare the efficiencies of different identification algorithms [117–119]. But their conclusions are not extensive for the radiative property identification, which certainly remains a subject of further investigation.

**CONCLUDING REMARKS**

As a closure of the overview presented in this paper, the following concluding remarks may be outlined:

- The determination of radiative parameters may be shared in variable proportions between theoretical prediction and experimental identification, with the choice being specific to each particular situation, depending on the properties to be determined. Theoretical prediction may be considered as a end method or, alternatively, as a first approach to an experimental identification, and this is valid for the whole or just part of the properties set.
- Theoretical prediction and experimental identification are both based on a physical model (direct model). Its choice in terms of sophistication should be appropriate owing to the final use of the parameters that are sought and to the accuracy of the data available for the model nurturing.
- A number of physical models are available allowing the determination of radiative properties with more or less details and accuracy, from the classical theory of Mie and its developments or simplified variants, to modern developments of electromagnetic scattering theory that lend themselves to treating media hosting particles of arbitrary shapes, or inhomogeneous particles, that may mutually interact—in a word, to account for more complex situations. Major advances were seen in the past decade in the development of methods for solving the electromagnetic scattering problem and free software packages are available in some specific websites.
- A similar diversity is also observed for the radiative transfer equation forms used for the property identification.
- It is not mandatory, even perhaps not convenient, to use a sophisticated direct model and/or RTE form when one is not able to feed the form with data of appropriate accuracy, or when the application that has motivated the property search may be satisfied with a simpler set of properties. In spite of the...
Further research is needed on the optimal choice and sequence of identification parameters to be adopted, such as the choice of the couple \((\kappa_\lambda, \sigma_\lambda)\) instead of \((\beta_\lambda, \omega_\lambda)\).

Identification of properties requires the choice of an optimization method. Although different methods have been used for radiative properties identification, some deterministic, other ones stochastic, or hybrid, a lack is observed of methodology aimed to operate an “optimal choice of the algorithm of optimization” well matched to radiative properties sought and related experimental configurations.

An important need is still observed in the assessment of radiative properties at high temperatures. Due to the difficulty of development and operation of experimental setups that can work at high temperatures to provide spectral and directional radiation measurements, most current experimental facilities only work at room temperature. Owing to the growing importance of the design of materials intended to efficiently work at moderate to high temperatures, such as thermal insulations, the development of such facilities and techniques is a remaining challenge.

**NOMENCLATURE**

- \(CN\) condition number of matrix \(S\)
- \(f_i\lambda\) weights associated to a phase function, \(i = 1, 2\)
- \(F\) function to be minimized
- \(g_\lambda\) spectral asymmetry parameter of a phase function
- \(i, I_\lambda\) spectral intensity of radiation, \(W \, m^{-3} \, sr^{-1}\)
- \(I_0\lambda\) spectral intensity of blackbody radiation, \(W \, m^{-3} \, sr^{-1}\)
- \(K\) number of parameters to be identified
- \(m\) complex index of refraction
- \(n\) refractive index (real part of \(m\))
- \(N\) number of data used in the identification process
- \(p, P\) spectral phase function
- \(s\) space coordinate, \(m\)
- \(S\) matrix of sensitivity coefficients
- \(S_{ij}\) element of matrix \(S\)
- \(T\) spectral transmittance, or reflectance, or emittance
- \(x\) particle size parameter

**Greek Symbols**

- \(\beta_\lambda\) spectral extinction coefficient, \(m^{-1}\)
- \(\epsilon\) spectral emittance
- \(\theta, \phi\) polar, azimuthal angle, rad
- \(\Theta\) scattering angle, rad
- \(\kappa_\lambda\) spectral absorption volumetric coefficient, \(m^{-1}\)
- \(\lambda\) wavelength, \(m^{-1}\)
- \(\mu_\theta = \cos(\Theta)\)
- \(\sigma_\lambda\) spectral scattering volumetric coefficient, \(m^{-1}\)
- \(\tau_\lambda\) spectral optical coordinate, \(m^{-1}\)
- \(\omega_\lambda\) spectral albedo = \(\sigma_\lambda / \beta_\lambda\)
- \(\omega, \Omega\) solid angle, \(sr\)

**Subscripts**

- \(b\) blackbody
- \(d\) directional
- \(e\) experimental (measured data)
- \(h\) hemispherical
- \(t\) theoretical (data calculated from a model)
- \(HG\) Henyey–Greenstein
- \(HGN\) Henyey–Greenstein–Nicolau
- \(\lambda\) spectral (at a given wavelength, per unit wavelength)

**REFERENCES**

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Jean-François Sacadura is Emeritus Professor of Mechanical Engineering and Thermal Sciences at INSA Lyon (National Institute of Applied Sciences) in Lyon, France. He received his mechanical engineering degree in 1963 from INSA Lyon, his Ph.D. in 1969 from the University of Lyon, and the doctor-ès-sciences physiques degree from INSA Lyon in 1980. He started his academic career in 1963 as a teaching and research assistant at INSA Lyon, where he reached the position of full professor of mechanical engineering with tenure in 1983. He has been an emeritus professor since 2005. Dr. Sacadura was dean of the master and PhD programs in thermal engineering at INSA Lyon (1998–2005) and director of the Thermal Science Centre of Lyon (CETHIL) from 1997 to 2003. His research interests include thermal radiation and combined mode of heat transfer, thermophysical properties, particulate media, semitransparent media, high temperatures, modeling and experimental characterization by direct and inverse methods, and radiative–convective design of water-spray fire protections for oil and chemical plants. He has co-authored more than 120 refereed publications, and is co-editor and/or co-author of four books. He serves on several editorial boards and is co-chief-editor of *High Temperatures–High Pressures* (2008–). He is a member of the boards of Société Française de Thermique (SFT, 1970–, chairman 1997–1999), Association Française de Mécanique (AFM), European Conference in Thermophysical Properties (ECTP, chairman 1993–1996), Eurotherm Committee (founding and honorary member), and ICHMT Scientific Council and Executive Committee. His efforts in engineering education and research have been recognized with several awards: Chevalier des Palmes Académiques, 1986; Prix Académies INSA as an outstanding PhD promotor and advisor, 1989; Officier des Palmes Académiques, 1998; and Commandeur des Palmes Académiques, 2005 (Palmes Académiques are French Ministry of Education awards for performance in higher education and research).