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SYMBOLIC MONTE CARLO METHODS : AN ANALYSIS TOOL FOR THE EXPERIMENTAL IDENTIFICATION OF RADIATIVE PROPERTIES AT HIGH-TEMPERATURE

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ABSTRACT. A new Symbolic Monte Carlo (SMC) method, involving null collisions, was recently developed. It allows expressing radiative quantities as polynomials of absorption and scattering coefficients in a single simulation. This approach is applied here for giving insight on the experimental identification of radiative properties at high-temperature. SMC polynomials are used to analyze if the identification of absorption and scattering coefficients is achievable from measurements of radiative intensities in different directions. In particular, spectrometric emission measurements at high-temperature are investigated.

INTRODUCTION

Inverse measurements problems in radiative transfer, such as experimental identification of radiative properties, concern many applications: from the characterization of complex media (porous or fibrous media [1], medical imaging [2], *etc.*) to combustion diagnosis [3]. In those problems, radiative properties, such as absorption and scattering coefficients and/or phase function parameters, are determined from radiative fluxes measurements. In particular, spectral measurements of transmitted and reflected fluxes at room temperature have been used to identify the effective scattering and absorption properties of various media [4, 5, 6, 7].

The radiative properties of materials, such as ceramics used for thermal barrier coatings [8, 9] or for thermal protection systems [10], are generally strongly dependent on temperature. For high-temperature processes, it is therefore relevant to identify radiative properties of the considered materials at their operating temperature. However, the development of an experimental bench for the measurements of transmitted and/or reflected radiative fluxes remains a challenging issue at high-temperature since the own emission of the medium is predominant. In this context, an experimental bench has been specifically designed in [11] for measuring spectrometric emission of materials heated from 800K up to 2000K.

The approach proposed in [11] has been applied to identify the spectral radiative properties of SiO_2 fibrous material at temperature of 1248K and 1722K. It was shown that identifying directly the absorption and scattering coefficients from spectral emission measurements, using the radiative transfer equation (RTE) as direct model, was not always achievable because of correlations between absorption and scattering coefficients. In [11], input parameters of dielectric function models have been identified, and the radiative properties determined using the Mie theory. However, these models are restricted to the considered materials (fibers of SiO_2). One question remains from this work: in which case is it possible to directly identify radiative properties at high-temperature using the RTE, and which measurements are most relevant to obtain information about absorption and scattering inside the medium, regardless of the considered material.

The aim of the paper is to introduce Symbolic Monte Carlo (SMC) and illustrate how it may be used as an analysis tool for answering the preceding question. The SMC method has been introduced by Dunn in [12]. In SMC, Monte Carlo algorithms are applied with parameters retained as symbolic variables [13]. The output of SMC simulations is therefore a functional in which the requested parameters appears explicitly. SMC was investigated in radiative transfer by Dunn in [14], and Subramaniam and Menguç in [15]. In [15], the scattering albedo together with the asymmetry factor of the step phase function has been retained as symbols to obtain a functional of these parameters. However, the knowledge of the optical thickness was required in these studies.
In a recent work [16], the authors propose a new SMC approach based on null-collision algorithms which overcome the required knowledge of optical thickness. It allows expressing radiative quantities as polynomials of absorption and scattering coefficients. In other words, a single simulation allows to evaluate the considered radiative quantity over a wide range of absorption and scattering coefficients values. Therefore, it turns out to be a very practical tool for physical and inverse analysis.
In this work, it is shown how SMC polynomials can be used for analysis in the context of identification of radiative properties at high-temperature.

SYMBOLIC MONTE CARLO METHOD

SMC algorithm and standard MC algorithm are very similar. In standard MC, scalars are affected to all parameters and a numerical value of the radiative quantity is estimated. In SMC, some parameters are retained as symbolic variables, and therefore undetermined all along the simulation process. As a result, SMC directly outputs the radiative quantity as a function of the retained symbolic parameters. As shown later, the functional expression may be relevant for physical analysis and identification approaches.

Principle of SMC using temperatures as symbols In order to illustrate the principle of SMC, the case of a non-scattering and homogeneous absorbing medium is considered. The radiative intensity $I_\eta(\mathbf{x}_0,\mathbf{u}_0)$ at location $\mathbf{x}_0$ in the direction $\mathbf{u}_0$ is then given by:

$$I_\eta(\mathbf{x}_0,\mathbf{u}_0) = \int_0^{+\infty} \kappa_\eta \exp\big(-\kappa_\eta l\big) B_\eta\big(T(\mathbf{x})\big) dl \tag{1}$$

where l are the free path lengths and $B_\eta\big(T(\mathbf{x})\big)$ the blackbody intensity at location $\mathbf{x} \equiv \mathbf{x}(l) = \mathbf{x}_0 - l\mathbf{u}_0$.

The estimation of $I_\eta(\mathbf{x}_0,\mathbf{u}_0)$ by a standard direct Monte Carlo algorithm would consist in performing a large number N of independent samples (indexed i) composed of the following steps:

1) *Sample a free path according to the pdf* $p_L(l) = \kappa_\eta \exp\big(-\kappa_\eta l\big)$
2) *Compute the absorption location* $\mathbf{x}_i = \mathbf{x}_0 - l\mathbf{u}_0$
3) *Compute the Monte Carlo weight* $w_i = B_\eta(T(\mathbf{x}_i))$.

Finally, the radiative intensity is approximated by averaging the N samples according to:

$$I_\eta(\mathbf{u}_0,\mathbf{x}_0) \approx \frac{1}{N}\sum_{i=1}^{N} w_i = \frac{1}{N}\sum_{i=1}^{N} B_\eta(T(\mathbf{x}_i)) \tag{2}$$

In the case where the considered medium is composed of N_T isothermal columns $\mathscr{V}_j$ for $j \in \{1, N_T\}$, the radiative intensity is approximated by:

$$I_\eta(\mathbf{x}_0,\mathbf{u}_0;T_1,T_2,\ldots,T_{N_T}) \approx \frac{1}{N}\sum_{i=1}^{N}\Big[\sum_{j=1}^{N_T} \mathrm{H}\big(\mathbf{x}_i \in \mathscr{V}_j\big) B_\eta(T_j)\Big] \tag{3}$$

where $\mathrm{H}(a)$ is an Heaviside function, equal to 1 if the condition a is verified, to 0 otherwise.

In practice, standard MC algorithms provide a scalar value estimating the quantity of interest. However, Eq. 3 can be seen as a function of temperatures $T_j, \forall j \in \{1, N_T\}$ and can be slightly reformulated as follows:

$$I_\eta(\mathbf{x}_0, \mathbf{u}_0; T_1, T_2, \ldots, T_{N_T}) \approx \sum_{j=1}^{N_T} \left\{ \left[\frac{1}{N} \sum_{i=1}^{N} \mathrm{H}\left(\mathbf{x}_i \in \mathscr{V}_j\right) \right] B_\eta(T_j) \right\} = \sum_{j=1}^{N_T} \overline{w_j} B_\eta(T_j) \tag{4}$$

Using temperatures T_j as undetermined variables, it is possible to implement a SMC algorithm allowing the estimation of I_η as a tractable function of all temperatures. The only difference with regard to the direct algorithm described previously is that N_T weights (instead of a single weight) $w_{i,j} = \mathrm{H}\left(\mathbf{x}_i \in \mathscr{V}_j\right)$, for $j = \{1, N_T\}$, have to be computed at each sample and averaged as $\overline{w_j} = \frac{1}{N}\sum_{i=1}^{N} w_{i,j}$.

SMC using the absorption coefficient as symbol If the evaluation of radiative quantities as functional of temperatures is straightforward, dealing with radiative properties (absorption and scattering coefficient) leads to some difficulties. Indeed, starting again from Eq. 1, one could try to compute with SMC a function I_η with respect to the absorption coefficient. The first stage would be to introduce an arbitrary probability density of free-paths $\tilde{p}_L(l)$ independent of absorption coefficient (considered as unknown), rewriting Eq. 1 into:

$$I_\eta(\mathbf{x}_0, \mathbf{u}_0) = \int_0^{+\infty} \tilde{p}_L(l) \frac{\kappa_\eta \exp\left(-\kappa_\eta l\right)}{\tilde{p}_L(l)} B_\eta\left(T(\mathbf{x})\right) dl \tag{5}$$

Consequently, the estimation of I_η is expressed by the non-linear function of κ_η:

$$I_\eta(\mathbf{x}_0, \mathbf{u}_0; \kappa_\eta) \approx \frac{1}{N} \sum_{i=1}^{N} \frac{1}{\tilde{p}_L(l)} \kappa_\eta \exp\left(-\kappa_\eta l_i\right) B_\eta\left(T(\mathbf{x}_i)\right) \tag{6}$$

The non-linearity makes this sum hardly tractable since it is composed of as many non-linear terms as Monte Carlo samples. Therefore, SMC with κ_η (and/or with the scattering coefficient σ_η) cannot be handled with classical approaches and explains why the knowledge of the optical thickness was up to now required [14, 15].

It has been shown that null-collision algorithms [17, 18] allow to overcome the non-linearity issues encountered in SMC [16]. Indeed, the principle of null-collision algorithms consists in defining an arbitrary extinction coefficient $\hat{\beta}_\eta$ (independent of κ_η and σ_η) by adding fictitious collisions that do not change the overall radiative transfer. These collisions, of arbitrary coefficient $\gamma_\eta = \hat{\beta}_\eta - \kappa_\eta - \sigma_\eta$, corresponds to pure forward scattering events. In non-scattering medium, the radiative intensity $I_\eta(\mathbf{x}_0, \mathbf{u}_0)$ is then deduced from the recursive expression:

$$I_\eta(\mathbf{x}_j, \mathbf{u}_0) = \int_0^{\infty} \hat{\beta}_\eta \exp\left(-\hat{\beta}_\eta l_{j+1}\right) dl_{j+1} \left\{ \frac{\kappa_\eta}{\hat{\beta}_\eta} B_\eta(\mathbf{x}_{j+1}) + \frac{\hat{\beta}_\eta - \kappa_\eta}{\hat{\beta}_\eta} I_\eta(\mathbf{x}_{j+1}, \mathbf{u}_0) \right\} \tag{7}$$

where $\mathbf{x}_{j+1} = \mathbf{x}_j - l_{j+1}\mathbf{u}_0$.
Thus, the exponential attenuation term only depends on the arbitrary extinction coefficient $\hat{\beta}_\eta$. A symbolic Monte-Carlo algorithm can therefore be developed using the absorption coefficient as symbols. For a given sample i, the symbolic weight is given by the following expression:

$$w_i(\kappa_\eta) = \sum_{j=1}^{M_i} \frac{\kappa_\eta}{\hat{\beta}} B_\eta(\mathbf{x}_{j,i}) \times \left[\frac{\hat{\beta}_\eta - \kappa_\eta}{\hat{\beta}_\eta} \right]^{j-1} \tag{8}$$

where M_i is the number of null-collisions events in the generated path, and $\mathbf{x}_{j,i}$ the successive locations of null-collisions. w_i can now be developed under a polynomial form of order M_i: $w_i(\kappa_\eta) = \sum_{j=1}^{M_i} a_{j,i}\kappa_\eta^j$. The radiative intensity is estimated according to :

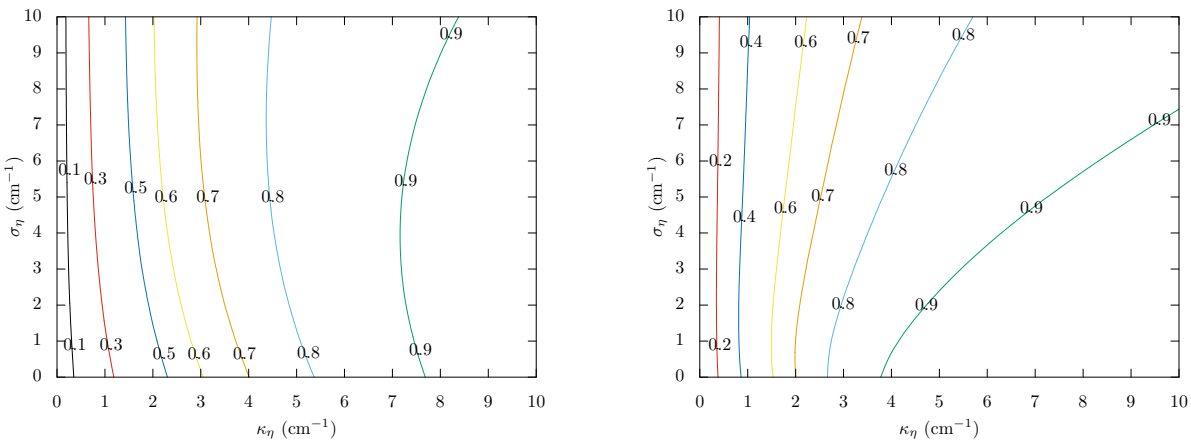
$$I_\eta(\mathbf{x}_0,\mathbf{u}_0) = \frac{1}{N}\sum_{i=1}^{N}\sum_{j=1}^{M_i} a_{j,i}\kappa_\eta^j = \sum_{j=1}^{M_{max}}\left[\frac{1}{N}\sum_{i=1}^{N} a_{j,i}\right]\kappa_\eta^j = \sum_{j=1}^{M_{max}} \overline{a}_j\kappa_\eta^j \tag{9}$$

where M_{max} is the maximum number of null-collision events overall the N realizations ($a_{j,i} = 0$ if $j > M_i$).

SMC using the absorption and scattering coefficients as symbols As shown in [16], all the preceding developments can be extended to both absorbing, emitting and scattering medium. The radiative intensity in an homogeneous absorbing and anisotropic scattering media is then obtained, following the same steps as described previously, by:

$$I_\eta(\mathbf{x}_0,\mathbf{u}_0) \approx \sum_{j=0}^{M_{nc,max}+M_{s,max}-1}\sum_{k=0}^{M_{nc,max}} \overline{b_{j,k}}\sigma_\eta^j\kappa_\eta^k \tag{10}$$

where the coefficients $\overline{b_{j,k}}$ are calculated during the SMC simulation, and where $M_{nc,max}$ and $M_{s,max}$ are the maximum number of null-collision and scattering events encountered during the N random generations of optical paths. An example of results is provided in Fig. 1 where an isothermal uni-



(a) (b)

Figure 1: $I_\eta(\mathbf{x}_0,\mathbf{u}_0)/B_\eta(T)$ computed with SMC as a function of κ_η and σ_η. In Fig. 1a, the angle θ between $\mathbf{u}_0$ and the outgoing normal at $\mathbf{x}_0$ is 0, while $\theta = 60°$ in Fig. 1b.

form absorbing and scattering medium is considered in a rectangular box (described in figure 2). Isolines that lead - according to the RTE - to a given value of $I_\eta(\mathbf{x}_0,\mathbf{u}_0)/B_\eta(T)$ are displayed for the infinity of couples $(\kappa_\eta,\sigma_\eta)$. The polynomial, obtained with one SMC simulation, allows in this case to evaluate $I_\eta(\mathbf{x}_0,\mathbf{u}_0)$ for $\kappa_\eta \in [0,10]$ cm^{-1}and $\sigma_\eta \in [0,10]$ cm^{-1}. Figures 1a and 1b show that measurements (represented by isolines) of radiative intensity leads to relevant information for identification purposes. For instance, for a given value of $I_\eta(\mathbf{x}_0,\mathbf{u}_0)/B_\eta(T)$, the identification of κ_η for small absorption optical thickness may be achievable, whereas a wide range of σ_η values remains possible.

RESULTS

In this section, it is shown how SMC polynomials can be used as an analysis tool in the context of experimental identification of radiative properties in high-temperature media.

SMC benefits from all the advantages inherent in standard Monte Carlo method [16]. In particular, it can be easily applied to complex three-dimensional geometries. A three-dimensional rectangular box with an homogeneous absorbing and scattering medium is considered here (see Fig 2). The size of the box (0.003m $\times$0.02m $\times$0.02m) corresponds to the typical size of samples tested in the experimental bench described in [11]. An Henyey-Greenstein phase function is assumed with an asymmetry factor of 0.3. The surrounding medium is cold.

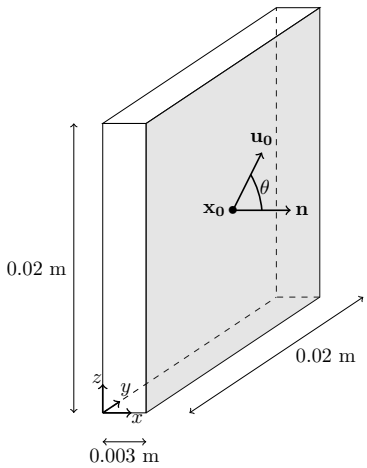
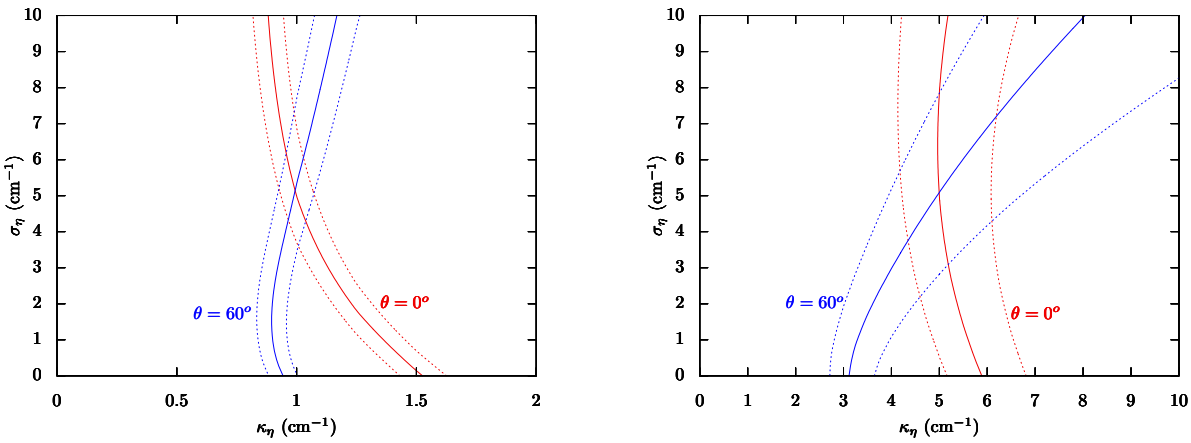


Figure 2: Scheme of the test case. The radiative intensity is measured at the center of one side of the sample : $\mathbf{x}_0 = (0.003, 0.01, 0.01)$ and the angle θ is defined between $\mathbf{u}_0$ and the outgoing normal $\mathbf{n}$ at $\mathbf{x}_0$.

Measurements in isothermal media As shown in Fig. 1, an infinity of couples $(\kappa_\eta, \sigma_\eta)$ lead to a particular solution of RTE. Therefore, several experimental values of radiative intensities are required to identify the couple $(\kappa_\eta, \sigma_\eta)$. Here, for the previous rectangular box assumed as isothermal, two different directions of measurements ($\theta = 0°$ and $\theta = 60°$, see Fig. 2), leading to different depths along the line of sight, are considered.
In Fig. 3, two cases are considered. The first one (Fig. 3a) corresponds to a medium where $\kappa_\eta = 1$ cm^{-1} and $\sigma_\eta = 5$ cm^{-1}, the second one (Fig. 3b) to $\kappa_\eta = \sigma_\eta = 5$ cm^{-1}. In both figures, SMC results are only depicted in solid lines for pseudo-measured values $I_{\eta,meas}$, *i.e.*, only the isolines $I_\eta(\mathbf{x}_0, \theta = 0) = I_{\eta,meas}(\mathbf{x}_0, \theta = 0)$ and $I_\eta(\mathbf{x}_0, \theta = 60) = I_{\eta,meas}(\mathbf{x}_0, \theta = 60)$ are displayed. These pseudo-measured intensities have been actually computed for each direction and case with a standard MC. For each case, the intersection of the curves indicates the desired couple of radiative properties $(\kappa_\eta, \sigma_\eta)$.
However, in practice, experimental errors must be taken into account. The dashed lines depict an interval of $\pm 5\%$ experimental errors around the pseudo-measured values. When this confidence interval is considered, the SMC polynomials give regions that cover couples of properties that are potential solutions of the inverse problem. In Fig. 3a, it is shown that for small absorption optical thicknesses, the region of potential solution is relatively small, and consequently, the identification may be carried out with small uncertainties. In Fig. 3b, where the absorption optical thickness is more important, the region covering the potential solutions is much larger. Consequently, from two measurements at different directions, the identification of absorption and scattering coefficients remains difficult when



(a) (b)

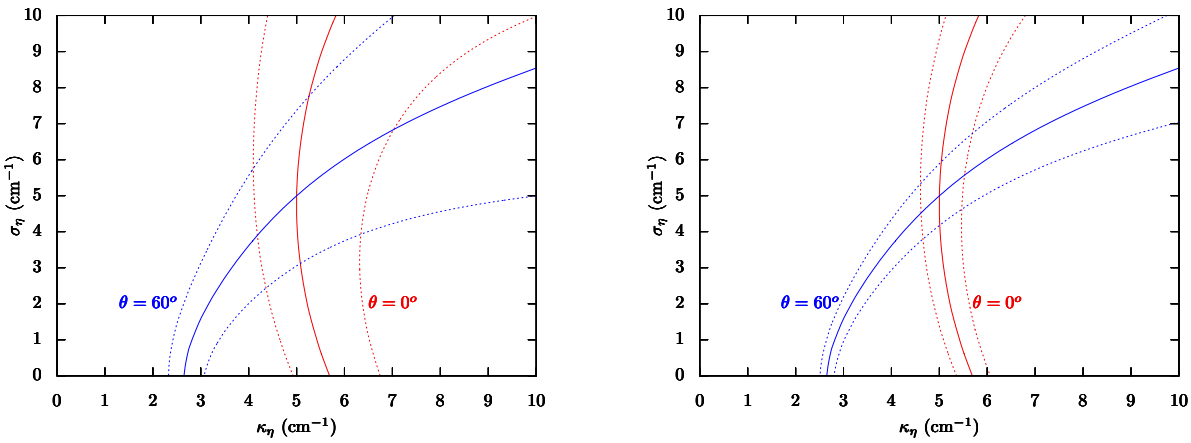
Figure 3: $I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T)$ computed with SMC as function of κ_η and σ_η. In Fig. 3a, $I_{\eta,meas}(\mathbf{x}_0, \theta = 0)/B_\eta(T) = 0.368$ and $I_{\eta,meas}(\mathbf{x}_0, \theta = 60)/B_\eta(T) = 0.429$. In Fig. 3b, $I_{\eta,meas}(\mathbf{x}_0, \theta = 0)/B_\eta(T) = 0.829$ and $I_{\eta,meas}(\mathbf{x}_0, \theta = 60)/B_\eta(T) = 0.849$. For each pseudo-measured radiative intensity, two isolines corresponding to $0.95 I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T)$ and to $1.05 I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T)$ are displayed to present the effect of $\pm 5\%$ experimental errors.

large absorption optical thickness are involved. Other experimental values are required to reduce the region of potential solutions.

Measurements in anisothermal media The experimental heating of materials could lead to a temperature gradient that needs to be considered when attempting to identify radiative properties. As shown in [16], SMC approaches are perfectly adapted to deal with such non-uniformities.
Here the sample described in Fig. 2 is considered with a linear profile of temperature along the x-axis ($T(x = 0) = 1500$K and $T(x = 0.003) = 1200$K). Again, the choice of measurements and sample temperatures is made according to typical experiments conducted in [11]. The variation of absorption and scattering coefficients with respect to temperature is neglected.
Pseudo-measured values of $I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T_{max})$ are assumed, resulting from a medium with $\kappa_\eta = \sigma_\eta = 5$ cm^{-1} at wavenumber $\eta = 1250$ cm^{-1} (wavelength $\lambda = 8\mu$m). The SMC results are displayed in Figs. 4. As previously, $\pm 5\%$ experimental errors are depicted in Fig. 4a. By comparison with Fig. 3b, it is observed that the temperature gradient increases the region of potential solutions.
An analysis of the influence of experimental errors may be required for inversion purposes. In Fig. 4b, $\pm 2\%$ experimental errors are considered. Assuming 2%, the region of potential solutions is strongly reduced. κ_η is between 4.8 and 5.8 cm^{-1}, and σ_η is between 3.8 and 6.5, while an infinite region of possible solution ($\kappa_\eta > 4$ cm^{-1} and $\sigma_\eta > 2.2$ cm^{-1}) is observed with errors of $\pm 5\%$. In this particular case, comparisons allowed by SMC show that the feasibility of the identification is very sensitive to the experimental uncertainties if two measurements in two directions are available.

CONCLUSION

The introduction of null-collisions in the SMC approach allows expressing radiative quantities as polynomials of absorption and scattering coefficients. For a given configuration, one single SMC simulation (very similar to standard MC simulation) allows to estimate the radiative quantity over a wide range of absorption and scattering coefficients values. The resulting functional expression offers several advantages:



(a) (b)

Figure 4: $I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T_{max})$ computed with SMC as function of κ_η and σ_η. Only isolines corresponding to the pseudo-measured intensities (for $\theta = 0$ and $\theta = 60°$, see Fig. 2) are represented. In Fig. 4a, two isolines corresponding to $0.95 I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T_{max})$ and to $1.05 I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T_{max})$ are displayed to present the effect of $\pm 5\%$ experimental errors. In Fig. 4b, two isolines corresponding to $0.98 I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T)$ and to $1.02 I_\eta(\mathbf{x}_0, \mathbf{u}_0)/B_\eta(T)$ are displayed to present the effect of $\pm 2\%$ experimental errors.

- It can be used as direct model in inversion procedures. Indeed, the functional is an estimation of the radiative quantities for all sets of parameters values. It consequently avoids the costly resolution of the RTE at each iteration. Initially, SMC has been developed for this purpose [13, 15].
- It is a valuable tool for inverse analysis with regard to the uniqueness of solutions, and to develop efficient sensitivity analysis (thanks to its polynomial form) [16].
- It can be a precious help for experimental design. It allows to know if the identification is achievable from the available measurements, and to determine which measurements are most relevant.

In this work, identification of radiative properties in high-temperature media is investigated with SMC. In particular, it is shown how the polynomials can be used to study the limitations of spectrometric emission measurements for identifying absorption and scattering coefficients. Indeed, SMC polynomials allow to gather very practical information about the feasibility of the inversion, and about the influence of experimental errors.

This study shows that the identification of radiative properties from spectrometric emission measurements may be hardly achievable when large optical thicknesses are involved as in several high-temperature processes. In these conditions, the development of an identification strategy is a challenging question that will be investigated in future work.

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