RADIATIVE TRANSFER MODELING FOR RADIATION-CHEMISTRY COUPLING ANALYSIS

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ABSTRACT
This study is devoted to the prediction of pollutant formation inside combustion chambers. In order to control minor species emissions, considering the high sensitivity of kinetic chemistry to temperature, it is required that physical analysis include detailed description of all heat transfer processes . As far as radiation is concerned, one may state that no numerical model is available at date that meets the requirements in terms of geometrical description and accuracy. The present work discusses an alternative approach that combines an accurate Monte Carlo algorithm with a simple Taylor expansion of radiative exchanges as function of temperatures and concentrations. Accurate models can be designed on this basis that can be easily coupled to detailed kinetics models.

Key words : gas-soot radiation, Monte-Carlo method, laminar diffusion flame.

1. INTRODUCTION

The steadily increasing severity of emission regulations has motivated strong research efforts concerning the pollutant formation mechanisms. It is well established that radiation is an important heat transfer mode in industrial large-scale flames. Then, the coupling of radiation and flame chemistry is a subject of renewed interest, with the acceptance of the fact that reacting flow models are not complete without radiation effects.

Our interest here goes beyond the ability to predict heat transfer to enclosure walls. It is known, for example, that substantial fractions of flame energy can be converted to radiation, and that the gas cooling resulting from the non-adiabatic loss can affect flame chemistry and leads to inaccuracies in prediction of pollutants like nitric oxide.

In the most recent literature, among emerging questions in this frame, the question of coupling mechanisms between soot processes and radiation in luminous flames has been widely considered.

Dagusé (1996) may be mentioned with his work on non-luminous laminar flames. Computations were performed with detailed chemical kinetics and narrow-band statistical models to study interactions between radiation and flame properties.

Hall (1994) has shown the effect of radiative cooling on NO production in a sooting opposed jet diffusion flame.

The results of Sivathanu and al. (1994) indicate that local radiative source term are essential when modeling soot kinetics in strongly radiating flames (where gas radiation can be neglected).

Recent work by Sivathanu and al. (1997) shows that even for weakly radiating flames (where soot radiation is dominated by gas radiation), the local radiative heat source strongly influences soot formation.

In intermediate cases, coupling between soot formation processes and radiative transfer can be expected to be significant and both gas and soot radiation need to be considered.

The high sensitivity of kinetic chemistry to temperature makes it necessary to design high quality radiation models with detailed geometrical and spectral aspects description.

Such a model is detailed hereafter using an adapted methodology for coupling radiate transfer with kinetic models.

Illustrations will be given with the analysis of a sooting counterflow diffusion flame.

2. PROBLEM FORMULATIONS

In terms of radiative transfer modeling, it's difficult to take into account spectral gas effects within the real world of geometrically intricate combustion chambers.

On the one hand, accurate gas radiation models are available but required such high computational powers that they can't be used in conjunction with complex combustion models. On the other hand, fast three-dimensional models satisfy all engineering needs in terms of energetics constraints but do not include detailed gas spectral property descriptions and therefore cannot allow accurate enough temperature predictions in vue of pollution studies. Considerable efforts are made towards the design of accurate and efficient radiation models and among alternative approaches, we retain an integral formulation and a numerical resolution with the Monte-Carlo Method.

2.1 The Net Exchange Formulation (N.E.F.)

The present formulation was first introduced by Green (1967) for atmospheric studies. It has some common features with the zone formulation, Hottel and Sarofim (1967), and was recently revisited by Cherkaoui et al. (1996a and 1996b) and Dufresne et al. (1998 and 1999). One of its main features is to allow an intrinsic satisfaction of both the energy conservation and reciprocity principle. In the Net Exchange Formulation, radiative interaction is expressed between pairs of system subdivision elements. Without entering in any details, this formulation can be illustrated in the case of two volumes V_i

and V_j within a black wall cavity filled with non-scattering medium :

$$\psi(V_i, V_j) = \int_0^\infty d\eta \int_{V_i} dV_i(P) \int_{Vj} dV_j(Q)\, k_\eta(P)\, k_\eta(Q)\, \tau_\eta(PQ) \frac{\left[L_{b\eta}(P) - L_{b\eta}(Q)\right]}{l_{ij}^2} \qquad (1)$$

Where η is the wave number,

$$k_\eta = k_\eta^s + k_\eta^g \qquad (2)$$

Is monochromatic absorption coefficients due to both soot and gas contributions and $\tau_\eta(PQ)$ is the monochromatic transmittance function along straight line between two points *P* and *Q*.

In most combustion configurations, soot particles have dimensions of the order of 1 µm, which allows scattering to be ignored. According to Lee & Tien (1981) and Dalzell and Sarofim (1969), the monochromatic absorption can be related to the soot volume fraction f_v according to:

$$k_\eta^s = 5.5\ \eta\ f_v \qquad (3)$$

For standard combustion gases, absorption coefficient and transmission functions can be computed from modern spectral databank, in particular that proposed by Soufiani and Taine (1997). This databank provides average quantities on narrow spectral bands that can be used for description of line spectrum properties without a full description of all line profiles. Equation (1) is therefore not used as such, but a k-distribution approach can be used that leads to very much equivalent expressions.

2.2 An approximate Taylor-like formulation

Exact computation of radiative exchanges can be performed on the basis of the previous integral formulation using the Monte-Carlo Method (M.C.M.). But despite of all recent developments in this field, such computations remain numerically expensive and this procedure could not be coupled with any accurate chemical kinetics model. However, one major advantage of the M.C.M. is that sensitivities to all problem parameters can be computed without any additional cost. The reason, is that in the M.C.M. algorithm, integrals are computed as discrete sums of analytical functions evaluated at randomly generated values. In this sum, the analytical function can simply be replaced by its partial derivative with respect to any parameter to get an estimate of the derivative of this integral.

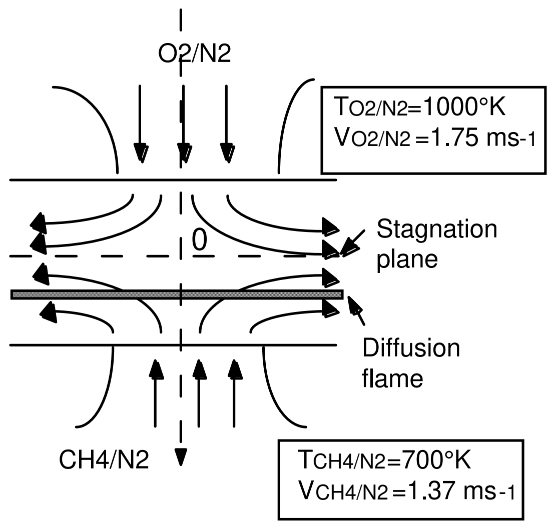
For instance, let us consider that the system is subdivided in *N* volume elements and that in each volume V_i, soot fractions can be assumed uniform with the value $f_{v,i}$. Then, when computing the radiative exchanges with M.C.M. for a given set of temperature and concentration profiles, one can simultaneously estimate all derivatives of this exchanges with respect to all parameters, in particular to the N soot volume fractions :

$$\frac{\partial\psi\left(V_i, V_j\right)}{\partial f_{v,m}} = \int_0^\infty d\eta \int_{V_i} dV_i(P) \int_{V_j} dV_j(Q)$$

$$[-k(i)\, k(j)\, l_m\, \tau_\eta\ 5{,}5\eta\ f_{v,m} + \delta_{im}\, k(j)\, \tau_\eta\ 5{,}5\eta\, f_{v,i} + \delta_{jm}\, k(i)\, \tau_\eta\ 5{,}5\eta\ f_{v,j}]\ \frac{L_{b\eta}(i) - L_{b\eta}(j)}{l_{ij}^2} \qquad (4)$$

where l_m is the path-length inside V_m and δ the chronecker symbol.

Figure 1 – Schematic of the counterflow



Having access to all sensitivities allows to design a simple coupling procedure for the Monte-Carlo code with an unstationary chemical kinetics model. At time t_0 the temperature and concentration profiles are used to make a first Monte-Carlo computation to accurately estimate all radiate exchanges and all sensitivities. At further time steps of the kinetic model, while temperature and concentration profiles remain close enough to the initial profile, a Taylor expansion model can be used to rapidly estimate radiative sources terms. For instance, if only the soot volume fraction was submitted to changes, this approach would lead to the following simple formulation :

$$\psi_t = \psi_{t_0} + \sum_{k=1}^{N} \left.\frac{\partial\psi}{\partial f_{v,k}}\right|_{t_0} \left[f_{v,k,t} - f_{v,k,t_0} \right] \qquad (5)$$

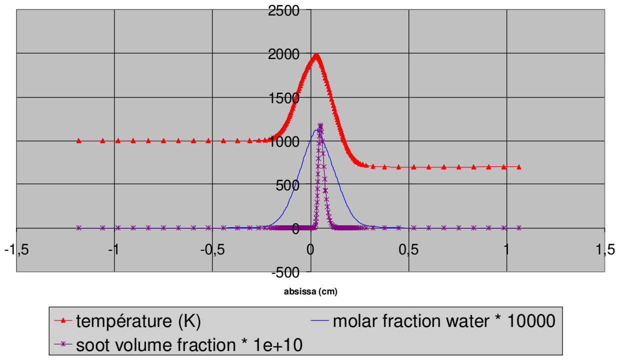
New Monte-Carlo computations are only performed when significant profile perturbation have occurred, therefore that leads to save large computational expenses and allows fast radiation-kinetic coupling.

3. SAMPLE RESULTS

The configuration consists of two opposed fuel and oxidiser jets at 1 atm pressure, leading to a one-dimensional structure

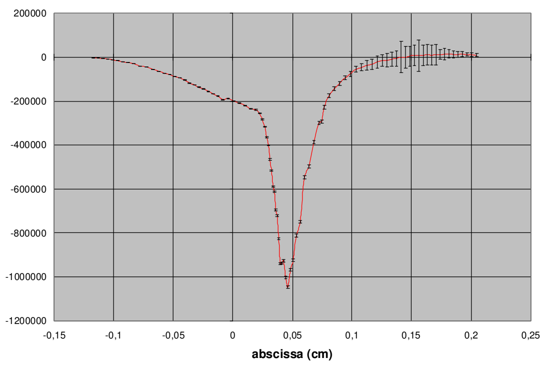
near the stagnation streamline (Figure 1). The strain rate is about 232 s^{-1}.
Temperature and gaseous species are computed by the model developped by F. Egolfopoulos (1994) and soot volume fractions (acethylene concentration) are obtained in the first step of a simplified reaction mechanism proposed by Leung et al. (1991). Soot transport is not implemented. Stationary solution profiles are reported in Figure 3 (temperature, water molar fraction and soot volume fraction).

Figure 2 – Concentrations and temperature profiles



These reference profiles are used to perform a first M.C.M. computation of soot-H_20 radiating transfer under a plane parallel assumption (CO_2 radiation is omitted). Corresponding radiate source terms are reported in Figure 4.
Despite of weak soot concentrations, maximum heat losses correspond to soot radiation. Closed to the region of maximum temperature, gas radiation represents only 25% of the total radiation heat loss.
But soot being formed only in very restrictive area, the gaseous absorption is dominant outside the temperature tip vicinity (Figure 3).

Figure 3 – Source term radiation (W/m^3) with error bars



In order to test the validity of the linearized solution, a 10% perturbation is applied to the soot volume fraction profile and two separate computations are performed :
(i) an exact M.C.M. computation
(ii) a computation with the Taylor-like model based on the first reference profile.

Figure 4 displays both solutions indicating that for such perturbation, the approximate and exact solution are in agreement within 0.07% accuracy. Making a zoom in Figure 5, allows to observe the difference between the perturbated and non-perturbated radiative source term.

Figure 4 – The compared results of Source term radiation (W/m^3)

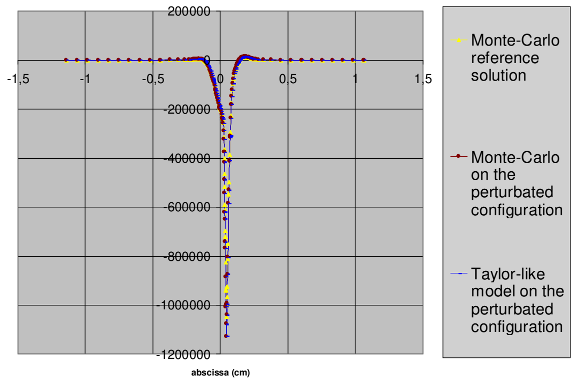
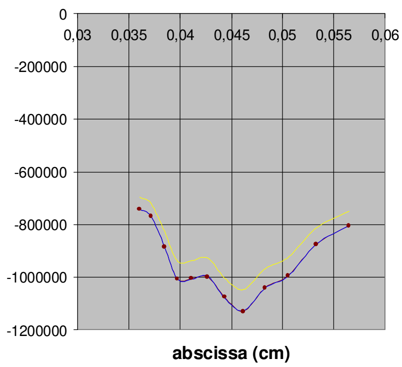


Figure 5 – Zoom of the compared results of Source term radiation (W/m^3)



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NOMENCLATURE

$f_{v,m}$ soot volume fraction in the m^{th} volume

k_η monochromatic absorption coefficient of gas-soot (m^{-1})

k_η^g monochromatic absorption coefficient of gas (m^{-1})

k_η^s monochromatic absorption coefficient of soot (m^{-1})

$L_{b\eta}$ Spectral black body intensity (W/(m^2.sr.m^{-1}))

N number of volume inside the system

V_m mth volume of the system (m^3)

$\psi\ (V_i, V_j)$ Radiative net exchange between two volumes (W)

η wave number (m^{-1})

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