EFFECT OF FRACTAL PARAMETERS OF SOOT AGGREGATES ON THEIR ABSORPTION AND SCATTERING PROPERTIES SIMULATED BY DISCRETE DIPOLE APPROXIMATION

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Effect of Fractal Parameters of Soot Aggregates on their Absorption and Scattering Properties Simulated by Discrete Dipole Approximation

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Abstract

The objective of this study is to determine the impact of the fractal properties of methane soot issued from incomplete combustions on their absorption and scattering properties. The fractal properties are selected according to the bibliography. Sample numerical aggregates are generated accordingly. The absorption and scattering cross-sections, as well as the asymmetry factor are determined by our in-house developed DDA (discrete dipole approximation) code. Simulation results indicate a higher sensitivity of the radiative properties to the fractal dimension than to the prefactor. This sensitivity analysis aims to guide the choice of relevant characterization experiments of soot samples about the necessary precision on measurements and data collection, and also to decide on the extent of soot morphological properties to be considered in heat transfer simulations.

1 Introduction

The interaction of soot with radiation is of interest for the control of heat transfer within combustion chambers and the improvement of their design. The overall size of soot aggregates formed by incomplete combustions may reach several hundreds of nanometers. In high temperature applications, radiation is the main heat transfer phenomenon, and a strong interaction is expected between an incident radiation and soot aggregates reaching the size of the incident wavelength. Under such conditions, the complex morphology of these aggregates cannot be ignored in the radiation-aggregate interaction because the incident wave is able to probe the spatial arrangement of the material within the aggregate. In this work, the electromagnetic theory "DDA" is used to account for the complex shape of the aggregates in the computation of their radiative properties i.e. the features of their interaction with the electromagnetic radiation.

2 Soot aggregates

Inside a combustion chamber, the morphology, the composition and the number of monomers of a flame soot depend strongly on the operating conditions like the temperature, type of fuel, stoichiometry, type of flame.

This research focuses on soot aggregates from the combustion of methane with air and oxygen. These aggregates are of complex shape formed by hundreds of sphere like carbonaceous monomers.

A complete 3D description of the aggregate is needed as input for the electromagnetic modelling. Unfortunately, the current microscopy techniques do not allow access to this information, which is one of the fundamental input data for DDA simulation.

Studies found in the literature propose the application of the fractal theory for the 3D geometry description of soot aggregates [1,2]. It is therefore possible to correlate the geometrical parameters N (number of monomers in the aggregate), r (monomer radius), R_g (radius of gyration of the aggregate) by use of the dimensionless parameters k_f (prefactor) and D_f (fractal dimension) specific to the material and the conditions of its elaboration:

$$N = k_f (R_g / r)^{D_f} \quad (1)$$

Once the D_f, k_f and N parameters are decided, different aggregation algorithms like DLA (Diffusion Limited Aggregation) or DLCCA (Diffusion Limited Cluster Cluster Aggregation) are able to generate aggregates in a specific range of D_f, k_f and N.

Reference	r	N	D_f	kf
Zhang et al. (1998)	10	5-200	1.62	-
Koylu et al. (1995)	-	-	1.65	2.71
Cai et al. (1995)	-	-	1.74	1.23
Cai et al. (1995)	-	-	1.74	2.45
Sorensen et al. (1992)	-	-	1.73	-
Lee et al. (2000)	5 - 10	10-600	1.75	6.46

Table 1 *Bibliographic data on methane soot fractal properties*

A partial list of r, N, D_f and k_f values found in the literature for our study on methane soot is presented in Table 1. D_f values are all around 1.7 while the range of k_f values is much larger (from 1.2 to 6.5) depending on the experimental conditions. In order to quantify the impacts of these fractal parameters on the radiative properties, we generate, for each couple (D_f , k_f) examined, several samples of soot aggregates and calculate their interaction cross-sections with the DDA theory.

The reference case selected for methane soot has the parameters D_f = 1.7 and k_f = 2.0. The fractal properties are slightly modified around the reference values to observe the variations in the absorption and scattering cross-sections. The number of monomers per aggregate N ranges from a few tens to several hundreds and the monomer radius r from 5nm to 10nm according to the literature data. To represent correctly the methane soot and to ensure a high aggregate-radiation interaction, higher limits are selected as N = 500 and r = 10nm. Numerical soot aggregates are generated using the DLCCA algorithm of Mackowski [2], as represented in Figure 1.

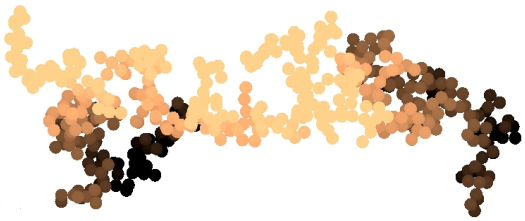

Figure 1 *Numerical aggregate with the reference parameters.*

3 DDA simulations

The DDA theory, originally proposed by Purcell and Pennypacker [3], is now a well-known modeling tool and the solution procedure is well established [4]. The method consists of material discretization into a sufficient number of dipoles where each dipole is polarized under the effect of the incident light. Each dipole contributes to the material-light interaction depending on its composition and its position in the discretized geometry. The limiting condition proposed by Draine [5] ensures a sufficient discretization under an incident wave :

$$|m|kd<1 \qquad (2)$$

where m is the complex refractive index of the material, k is the wavenumber and δ is the characteristic size of the volume elements of the discretization.

Our research is focused on the radiation in combustion chambers at 1500-2000K. The DDA simulations are performed for an incident wavelength of λ = 1µm ; this wavelength is in the range of high temperature radiation and is of the same order of magnitude as the characteristic size of the soot aggregates, so that unconventional interaction effects can be expected. The non-polarized thermal radiation is modeled by averaging the simulations with 2 orthogonal polarizability directions for the same incident wave direction [6].

For our monomers of radius r = 10nm, the spatial discretization of one dipole per monomer satisfies the condition given in Eq. (2) under λ = 1µm and with a typical methane soot refractive index of m = 2.0+0.5i [7].

Soot being a highly absorbing material, errors may arise in DDA simulations due to the badly explained surface interactions (surface dipole polarizabilities) and the shading effect on inner dipoles at high refractive indices, even when the discretization condition is satisfied [8,9]. In a previous work, we have compared the Mie cross-sections of a soot monomer with the DDA solutions using different discretization finesses, and decided to consider one dipole per monomer in order to avoid such discretization errors [10]. Furthermore, the final aim is to obtain a qualitative comparison of the sensitivities to the fractal parameters.

4 Absorption and scattering from soot aggregates

It is to be reminded that the scope is to find the impacts of the morphology and fractal properties of numerically generated aggregates on their radiative properties, all other variables (material composition, r and N) being fixed. In the first part, a comparison with the Mie solution shows the relevance of considering complex aggregate shapes. In the second part, the evolution of the radiative properties is presented as a function of D_f and k_f.

4.1 Effect of complex aggregate geometry

Our present in-house heat transfer simulations consider Mie solution properties for the radiative properties of soot. The DDA results for the reference aggregate presented in Figure 1 are compared to the Mie solution for a sphere having an equivalent material volume.

Cross-section	*DDA (m²)*	*Mie (m²)*	$\lvert\Delta C\rvert/C_{DDA}$
$C_{Extinction}$	7.52e-15	9.18e-15	23 %
$C_{Absorption}$	7.10e-15	8.08e-15	14 %
$C_{Scattering}$	0.42e-15	1.10e-15	160 %
$g_{asymmetry}$	0.411	0.061	-

Table 2 *DDA compared to Mie solution for the reference case*

The results presented in Table 2 show the relevance of DDA simulations for the determination of the radiative properties of soot. Despite the well-known overestimation tendency of absorption by DDA [11], the DDA solution for absorption is more than 10% below the Mie solution. Also the asymmetry factor predicted by the DDA indicates a strong forward scattering unlike the Mie solution, which proves the sensitivity of the DDA to the morphology of the scatterer.

4.2 Effect of fractal properties

Aggregate samples are simulated first by varying D_f around its reference value with all other parameters fixed. The trends of the DDA results as a function of D_f are represented in Figures 3 and 4. As can be seen on these figures, several numerical aggregates are generated for the same fractal parameter in order to account for the random nature of the aggregation process.

The same procedure is applied to study the sensitivity of the radiative properties to the prefactor k_f. The trends extracted for k_f are globally similar to the ones presented in Figures 3 and 4. However, the sensitivities on D_f and k_f are slightly different, as summarized in Table 3.

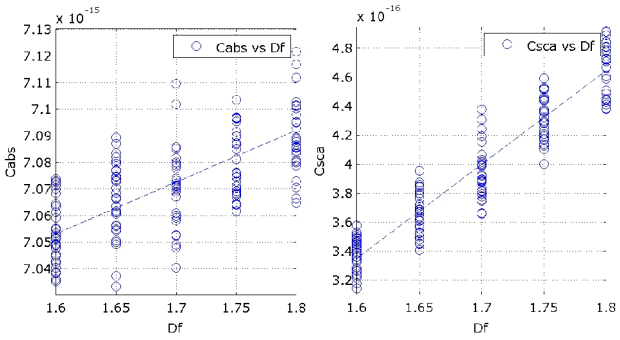

Figure 3 *Absorption (left), scattering (right) cross-sections of methane soot as a function of D_f (k_f, N and r are fixed).*

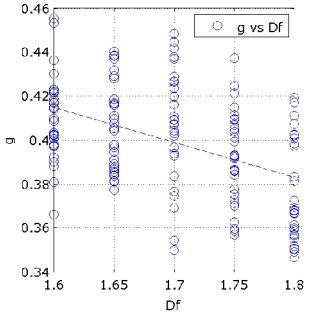

Figure 4 *Asymmetry factor for methane soot as a function of D_f (k_f, N and r are fixed).*

Results indicate that the interaction of soot with radiation is more affected by the value of the D_f parameter. In particular, varying D_f from 1.6 to 1.8 modifies the absorption by only 1.5% but the scattering by 46%. But as the absorption phenomenon dominates the scattering by soot, the sensitivity of soot radiative properties to the fractal parameters can be considered relatively small in the range of study if no particular interest is shown for scattering in the radiative heat transfer simulations.

	% change for 1% modification of D_f	% change for 1% modification of k_f
$C_{Absorption}$	0.12	0.04
$C_{Scattering}$	3.93	1.63
$g_{asymmetry}$	2.56	0.31

Table 3 *Variation of the radiative properties of methane soot aggregates due to modification of their fractal parameters.*

5 Conclusion and perspectives

In this study, the effects of the fractal properties on the radiative properties of methane soot are investigated. Methane soot is represented by numerically generated aggregates and the radiation-soot interaction is simulated using the DDA theory. Results indicate a small dependence for absorption but an important one for scattering versus the D_f and k_f parameters. If absorption is considered as the dominating phenomenon, the uncertainties on D_f and k_f around the reference values can be ignored.

The choice of our future characterization techniques and the necessary precision in data extraction depends on an extended version of the presented sensitivity study. Also, numerical soot aggregates will be gathered in groups of identical radiative properties merely dependent on the fractal parameters ; these groups are to be inserted as input for heat transfer simulations.

6 Acknowledgement

The authors kindly acknowledge the support of this research within the industrial chair OXYTEC of Ecole Centrale Paris, jointly conducted and funded by the laboratory EM2C, Air Liquide, CNRS and ANR.

7 References

[1] Megaridis C.M., Dobbins R.A., Morphological Description of Flame-Generated Materials, Combustion Science and Technology 71:95-109 (1990).

[2] Mackowski D.W., A simplified model to predict the effects of aggregation on the absorption properties of soot particles, J. Quant. Spectrosc. Radiat. Transfer 100:237-249 (2006).

[3] Purcell E.M., Pennypacker C.R., Scattering and absorption of light by nonspherical dielectric grains, Astrophys. J. 186:705-714 (1973).

[4] F. Enguehard, Mie Theory and the Discrete Dipole Approximation Calculating Radiative Properties of Particulate Media with Application to Nanostructured Materials, in S. Volz, *Thermal Nanosystems and Nanomaterials, Topics in Applied Physics* 118:151-212 (Springer Verlag Berlin Heidelberg, 2009).

[5] Draine B.T., Flatau P.J., The discrete dipole approximation for scattering calculations, J. Opt. Soc. Am. A 11:1491 – 1499 (1994).

[6] C.F. Bohren, and D.R. Huffman, *Absorption and scattering of light by small particles* (Wiley, New-York, 1983).

[7] Van-Hulle P., Caractérisation des Aérosols Emis par la Combustion des Hydrocarbures : Application à la Mesure de l'Indice de Réfraction des Suies, thèse de doctorat, INSA Rouen (2002).

[8] Collinge M., Draine B., Discrete-dipole approximation with polarizabilities that account for both finite wavelength and target geometry, J. Opt. Soc. Am. A 21 :2023-2028 (2004).

[9] Rahmani A., Chaumet P.C., Bryant G.W., On the importance of local-field corrections for polarizable particles on a finite lattice : Application to the discrete dipole approximation, Astrophys. J. 607:873-878 (2004).

[10] Okyay G., Enguehard F., Modélisation des propriétés radiatives des suies issues de combustions incomplètes au moyen de l'approximation dipolaire discrète, *Congrès Français de Thermique,* Lyon, 2014. [abstract accepted, paper in progress]

[11] K.A. Fuller and D.W. Mackowski, Electromagnetic Scattering by Compounded Spherical Particles, in Mischenko et al., *Light Scattering by Nonspherical Particles*, 225-272 (Academic Press, 2000).