Journal of Quantitative Spectroscopy & Radiative Transfer 119 (2013) 53–66

Contents lists available at SciVerse ScienceDirect

Journal of Quantitative Spectroscopy & Radiative Transfer

journal homepage: www.elsevier.com/locate/jqsrt

Impact of morphological parameters onto simulated light scattering patterns

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ARTICLE INFO

Article history:
Received 21 August 2012
Received in revised form
7 December 2012
Accepted 12 December 2012
Available online 31 December 2012

Keywords:
Fractal aggregates
Light scattering
T-Matrix
DDScat

ABSTRACT

We have investigated the impact of the variation of various parameters of fractal aggregates on simulated light scattering patterns. Static light scattering is commonly used to measure soot in a flame and such a study could help to improve experimental approaches. Aggregate models, used for our light scattering simulations, are based on real soot structures that can be found under laboratory conditions in a premixed ethane/air flame (McKenna-type burner, equivalence ratio $\phi = 2.5$). Our work was not focused on modeling and analysis of aggregates that are typically encountered in the atmosphere, therefore the results might be of limited interest to climate scientists. In our study, the variation of all parameters that enter into the standard fractal equation were investigated. Additionally effects when varying the overlap of primary particles, the incident wavelength and the complex refractive index are discussed. For numerical simulations two different codes were used, the T-Matrix (when particles are in point contact) and the DDScat program (which is capable of performing light scattering simulations by overlapping spheres). Comparisons between these two methods show very good agreement. The results demonstrate that the radius of gyration is responsible for the amount of light scattered towards the back direction while the total volume of an aggregate defines the shape of the light scattering patterns. Small changes of the fractal dimension can be neglected (provided that the fractal prefactor is accordingly modified in a suitable way). The overlap level, if the radius of gyration is kept constant, introduces barely visible changes to the light scattering diagrams which suggest that a simple aggregate model, composed of particles being in point contact, can be used instead of a structure in early sintering stage when overlap of primary particles is not so high.

1. Introduction

Soot aggregates are products of incomplete combustion of organic fuels and are composed of a few, up to several hundred almost spherically shaped primary particles. Their morphology parameters are dependent on the individual combustion process, fuel composition, flame type and other technical aspects. Many techniques for determination of the morphology of soot aggregates have been developed. One of the most important techniques is

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0022-4073/$ - see front matter
http://dx.doi.org/10.1016/j.jqsrt.2012.12.014

TEM (transmission electron microscopy) analysis. However, it provides us with 2D projections of 3D fractal-like structures only. The lack of one dimension results in many difficulties and noticeable errors that persist in every method commonly used to derive morphological parameters from such images (e.g. [1,2]). Moreover, online-measurements are impossible to perform and the sampling process is considered to be quite invasive [3,4]. To overcome some of these problems, a new technique called ET (electron tomography) was created [5,6]. It is strictly associated with the TEM approach. A series of 2D images, collected from different viewing angles, are combined into representations which are capable of displaying morphological parameters of 3D structures. Because there is no simple formula for deriving 3D parameters from 2D images (e.g. the fractal dimension D_f), ET is considered to be much more accurate technique than TEM [5]. Optically based techniques, e.g. LII (laser-induced incandescence) [7–9] and ELS (elastic light scattering) [10] are capable of performing on-line measurements. The first one can be used to measure the size of primary particles and the second one to determine the fractal dimension as well as the soot volume fraction of aggregates. A new experimental approach, WALS (wide-angle light scattering), is capable of performing fast on-line measurements with a high angular resolution in the scattering pattern [11,12].

To model the light scattering by fractal-like structures many theories and programs have been developed so far. One of the most important is the Null-Field theory (also known as T-Matrix theory), proposed by Waterman [13], which is considered as numerically exact in its current implementation. However, it is limited to a single particle and cannot be used for more complex structures. For a more general treatment, the superposition T-Matrix method (capable of performing light scattering simulations by aggregates composed of particles being in point contact) has been developed. Another valuable theory is DDA (discrete dipole approximation), which was proposed by DeVoe [14,15] and improved by Purcell and Pennypacker [16]. It can overcome the point contact limitation. However, the computation time is significantly higher.

The main goal of this paper is to create a set of model aggregates similar to real soot structures, as inferred by TEM, and to investigate the impact of the standard fractal equation parameters and additionally the effect of the refractive index, the incident wavelength and the overlap between particles on the resulting light scattering diagrams. The aggregate model is based on the TEM analysis of 110 soot structures and the morphological parameters were calculated using the method proposed by Brasil et al. [1].

This study can help to develop effective soot characterization methods based on measuring static light scattering. We will first compare the T-Matrix program of Doicu et al. [17] with the DDScat program, written by Draine et al. [18] to show that both theories are capable of performing reliable light scattering simulations and provide us with almost identical light scattering results. Secondly, we create a soot aggregate model which is based on TEM analysis. The light scattering results by our model are compared to WALS measurements. Finally, we determine the impact of the variation of its parameters on the light scattering patterns.

2. Theoretical background

Soot aggregates are mostly described as fractal-like structures which are characterized by parameters which are considered to remain constant during the whole aggregation process. Fractal aggregates can be described by the equation:

$$N_p = k_f \left(\frac{R_g}{r_p}\right)^{D_f}, \tag{1}$$

where N_p is the number of primary particles, r_p is the particle radius and R_g denotes the radius of gyration of the structure. The fractal behavior is defined by two factors—the fractal dimension D_f and the fractal prefactor k_f. It was proven that the first one is independent of the size of the structure [19] and for freshly emitted particles its value is usually considered as $D_f \approx 1.8$ [20,21] what is typical for DLCA (diffusion limited cluster aggregation) process [10]. By the contrary, recent study (by the use of electron tomography measurements) suggests that this value might be underestimated and should be assumed as $D_f=2.4$ [20]. Moreover, due to the aging effect soot aggregates may collapse and form more compact clusters [22,20] with the fractal dimension even as high as $D_f=2.6$ [23]. The pseudo-fractal behavior of an aggregate cannot be described by a single parameter, also the fractal prefactor k_f is needed. Its origins and behavior are yet not fully clear. Moreover, its value changes from almost 1 to 3.4 according to various publications [24] and is dependant on the experimental set-up as well as on environmental conditions [21]. The radius of gyration can be expressed by the following equation:

$$R_g^2 = \frac{1}{N_p}\sum_{i=1}^{N_p}(r_i - r_0)^2, \tag{2}$$

in which r defines the position of a single particle in the structure and r_0 the geometrical center of the cluster. However, this is not the only suitable formula. For example Filippov et al. suggest the following correction [25]:

$$R_g^2 = \frac{1}{N_p}\sum_{i=1}^{N_p}(r_i - r_0)^2 + r_p^2. \tag{3}$$

They assume that the radius of gyration should also include all points on the spherule surfaces instead of the mean square of the distance between particles and the mass center. The resulting difference is only visible for aggregates that consist of very few spheres. In contrast, the overlap factor, defined by the equation:

$$C = 1 - \frac{l}{2r_p} = 1 - \frac{l}{d_p}, \tag{4}$$

where l defines the distance between centers of two particles, has an undeniable impact on the fractal prefactor [1]. It also affects the fractal dimension, but this change is less significant [26,27].

Soot particles are usually considered as non-overlapping spheres being only in point contact. Such approach has many advantages (e.g. can significantly decrease the computational