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MEASUREMENT OF AGGREGATE SIZE DISTRIBUTION BY INVERSION OF ANGULAR LIGHT SCATTERING

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ABSTRACT. The aim of this work is to propose a new method for determining the size distribution of submicronic particles by inversion of the measured angular scattering of light. This method relies on the determination of a function R_g^* by angular scattering. The variation of this function informs us about the polydispersity of the aggregates size. We show that, by supposing the nature of the size distributions (lognormal), it is possible to determine the governing parameters of these distributions.

1. Scientific Context

Some non-spherical particles, like soot particles produced by combustion processes, have a fractal morphology. These particles give rise to numerous researches for their modelling and due to their important implications in human health, flame radiative transfer and climatic impact. The soot size distribution is often determined by using ex-situ granulometers, after sampling of the particles. But the quenching of aggregation process in the sampling is difficult and raises the problem of representativeness of the results [1].

To avoid this issue, it is interesting to use in-situ angular light scattering measurement which is non-invasive. As aggregates are not spherical, their specific morphology has to be taken into account. Aggregates are fractal because there is a power-law relationship between the aggregate size (gyration radius R_g) and the aggregate mass (related to the number N_p of primary spheres in the aggregate):

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

In the so-called fractal law Eq.(1), D_f is the fractal dimension, R_p is the radius of a primary sphere and k_f is the prefactor. Thanks to a simple theory called Rayleigh-Debye-Gans for Fractal Aggregates (RDG-FA theory) [2], measurement of static light scattering can be interpreted for determination of the gyration radius. The limitations of RDG-FA theory has been studied by Farias et al.[3]. The main assumption is due to the size of the concerned particles relatively to the wavelength and to their optical index $\pi \frac{R_P}{\lambda} |m-1| < 1$ where λ is the wavelength of the laser and m the complex refractive index. Refractive index of soot has been recently determined in visible range of wavelengths [4] and the corresponding criteria for $R_p < 20nm$ (generally observed and admitted size) is lower than 0.75 for

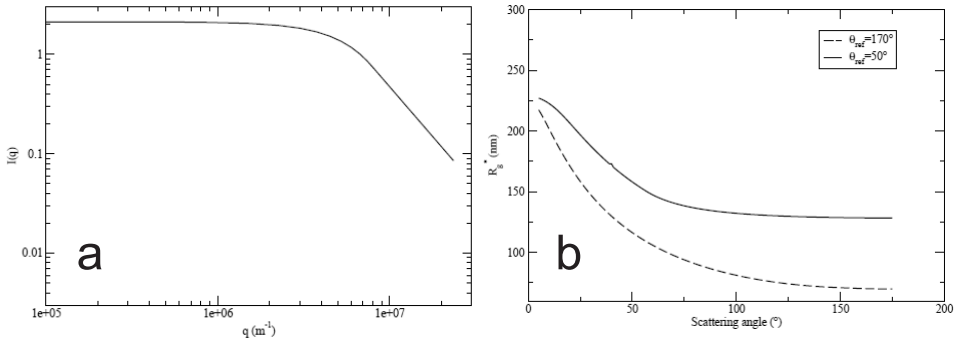

Figure 1. Calculation of R_g^* functions from the scattering angular signal. (a) Theoritical measurement of angular scattering and (b) Theoritical R_g^* for two θ_{ref}.

$0.1 < \lambda(\mu m) < 1.2$ making RDG-FA well adapted for soot particles. According to this theory, the intensity scattered by a single aggregate obeys the following equation:

$$I_{mono} = cN_p^2 f(q(\theta), R_g, D_f) \tag{2}$$

where θ is the scattering angle, c the coefficient of proportionality - due to the sensibility of the detector, R_p, λ and m. The function q is $q(\theta) = \frac{4\pi}{\lambda}\sin(\frac{\theta}{2})$ and f is defined by :

$$f \Leftrightarrow \begin{cases} \exp(\frac{-(q(\theta)R_g)^2}{3}) & \text{if} \quad (q(\theta)R_g)^2 < \frac{3}{2}D_f \\ (\frac{3D_f}{2e(q(\theta)R_g)^2})^{\frac{D_f}{2}} & \text{else} \end{cases} \tag{3}$$

For $(q(\theta)R_g)^2 < \frac{3}{2}D_f$ is the Guinier regime and else is the Power Law regime. The inversion by RDG-FA to infer the gyration radius of monodisperse aggregates has been recently validated [5]. Some authors ([2],[6]) have proposed to determine the gyration radius of the polydisperse population by this technique. However two distinct radii can be found if particles are suspected to scatter only in Guinier or in Power-Law regime and this assumption cannot be done without knowing the researched size distribution.

Koylu and Faeth [7] and Iyer et al. [8] proposed an inversion method to calculate the characteristics of soot size distributions by coupling scattering and extinction measurements. But these inversion methods rely on the knowledge of the refractive index of soot and morphological parameters. Moreover Burr et al [9] showed mathematically, by using Bayes' theorem, the ill-posedness of the inverse problem.

In the present work a new inversion procedure is proposed to determine the aggregate log-normal size distribution parameters. This method needs measurements at only three angles and does not need the knowledge of the refractive index of soot, monomer size or fractal prefactor. It will be shown that inversion is well posed. The main difference of this method in comparison to more direct ones (that compare the scattering properties to one another) is the use of an intermediate function of aggregate size, representative of the polydisperse population.

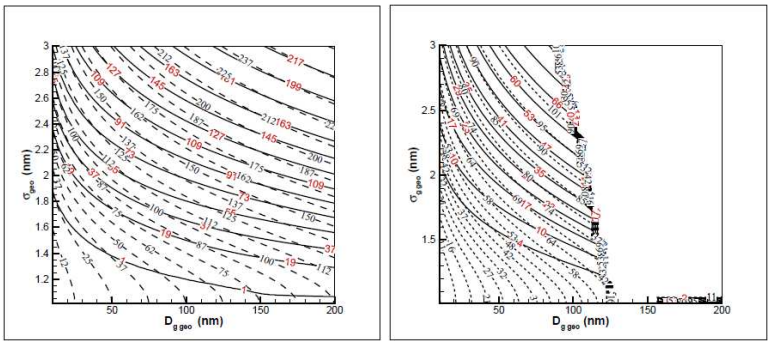

Figure 2. Cartographies, β is plotted in dotted lines and α in full lines. With (left) θ_{ref}=50° θ_1=15° θ_2=130°, and (right) θ_{ref}=170° θ_1=40° θ_2=130°.

2. Principle of the inversion for determining the soot size distribution

Our method consists in determining for each analized angle, by using the RDG-FA theory, a gyration radius $R_g^*(\theta)$ of a monodispersed population which have the same optical behaviour as the real polydisperse population. By integrating the expression (2) over the entire population described by the size probability density function $P(R_g)$, we find the scattered light intensity I as function of the scattering angle expressed as $q(\theta)$, illustrated in Fig.(1a). This intensity corresponds to the theoretical signal predicted in the case of polydisperse aggregate size population:

$$I(\theta) = c \int N_p^2 f(q, R_g) P(R_g) dR_g \tag{4}$$

As c is difficult to be determined experimentally, the theoretical signal Eq.(4) is normalized by the measured intensity at a reference angle. We define a normalized function Ψ:

$$\Psi(\theta) = \frac{I(\theta)}{I(\theta_{ref})} = \frac{\int N_p^2 f(q, R_g) P(R_g) dR_g}{\int N_p^2 f(q_{ref}, R_g) P(R_g) dR_g} \tag{5}$$

Our method consists in searching the equivalent size R_g^* of a monodisperse population having at each scattering angle the same scattering properties as normalized function Ψ. This leads to solve the following equation:

$$\Psi(\theta) = \frac{I(\theta)}{I(\theta_{ref})} = \frac{f(q, R_g^*)}{f(q_{ref}, R_g^*)} \tag{6}$$

Figure 1b shows the theoretically obtained R_g^* for a log-normal size distribution calculated with $D_f = 1.7$, the geometric mean gyration diameter $D_{ggeo} = 118nm$, the standard geometric deviation $\sigma_{geo} = 1.98nm$, $\lambda = 532nm$ and $\theta_{ref} = 50°$ and $\theta_{ref} = 170°$. The R_g^* curve depends on the reference angle chosen.

When the size distribution is monodiperse, R_g^* is constant for all θ. The inversion can be done by using only three angles : θ_{ref} and two other θ_1 and θ_2. R_g^* values associated to this two angles can be used to define two parameters : $\alpha = R_{g1}^* - R_{g2}^*$ and $\beta = \frac{R_{g1}^* + R_{g2}^*}{2}$. Figure 2 presents diagrams (α, β) versus (D_{ggeo} σ_{geo}) for given (θ_1, θ_2, θ_{ref}). The choice

of θ_1, θ_2 and θ_{ref} has an important effect on the range of D_{ggeo} and σ_{geo} which can be found by inversion. The figure 2 (right) shows a bad choice of these three parameters. On the good diagram 2 (left) we observe only one intersection between α and β curves. That indicates the uniqueness of the solution and the well-posedness of the inverse problem. One particle size distribution gives one couple (α, β).

Acknowledgments

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