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Anomalous Diffraction approximation for light scattering cross section: case of random clusters of non-absorbent spheres

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Abstract

We previously (Jacquier S. & Gruy F., Journal of Quantitative Spectroscopy & Radiative Transfer, 109(2008) 789-810) reformulated the anomalous diffraction (AD) approximation to calculate the light scattering cross section of aggregates by introducing their chord length distribution. It was applied to several ordered aggregates. This new method is entitled AD_r with the r for rapid because this one is at least one hundred times faster than the standard AD method. In this article, we are searching for an approximated expression for chord length distribution suitable all at once for ordered and disordered aggregates. The corresponding scattering cross section values are compared to the ones coming from the standard AD approximation.

Keywords: Aggregate, Light Scattering, Anomalous Diffraction, Scattering Cross Section, Chord Length Distribution

1. Introduction

Synthesis of inorganic material often leads to clusters of small particles. The successive steps of a precipitation are nucleation and growth of particles. When the particles reach a micronic size, growth stops and particles collide and aggregate. However, the particles are under the shear stress of the suspending medium. As a consequence, there is a competition between aggregation and break-up leading to aggregates with a little number of primary particles. The techniques planned to obtain information in real time on aggregates formation are the ones that use light-matter interaction. Turbidimetry is one of them. It is based on the measurement of the light intensity attenuation due to light scattering. Nevertheless, this kind of sensor does not allow, for the moment, to characterize aggregates by their shape, the number of primary particles that they contain their chemical nature. In fact, the analysis of the turbidity uses the Mie theory that is only suitable for spherical particle. However, there is an extension of the Mie theory to the case of aggregate: Kahnert [1] sums up some of them, one is entitled GMM (Generalized Multi-particle Mie solution) developed by Xu in 1995 ([2] for more information see the references therein). However, Xu's computer codes, making possible the calculation of the optical characteristics of aggregates, need consequent computing times. This limits the use of GMM theory because spectral turbidity analysis needs the knowledge of the scattering cross section of numerous different aggregates. Thus, in [3], we compared the exact method (GMM) against several approximated methods allowing the fast computation of the scattering cross section. Thereafter we [4] presented the improvement of one of these approximated

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method named rapid Anomalous Diffraction (ADr). This approximation makes easier the calculation of the scattering cross section for an aggregate consisting in optically soft primary particle whose size parameter is higher than 2. ADr was established for ordered aggregates but the ordered structure can support constructive or destructive interferences like a grating do. The topic of this paper is to extend the ADr approximation to the case of the random aggregates.

The background on AD and ADr approximations will be reminded in the section 2. Then, ADr approximation will be tested for random aggregates and an improved version will be proposed in the section 3. This section will be followed by conclusions.

2. Background on Anomalous Diffraction

The Anomalous Diffraction approximation is used to calculate the scattering cross section of large particles consisting in soft material. We consider an aggregate composed of N identical primary spherical particles. The radius and the dimensionless size parameter of the primary particles are respectively denoted r and $x=2\pi r/\lambda$ (λ is the wavelength of the incident light in the medium). Then, scattering cross sections depend on: the size of the primary particles, morphology of the aggregates, and relative refractive index. The relative refractive index (m) is the ratio between the material refractive index (n_p) and the refractive index of the medium (n_m).

2.1. ADr Formulation

Calculations based on anomalous diffraction approximation are time-consuming. Attempts were made to reduce the computation time [5]. As the chord length l appears in AD approximation, it is obvious that the scattering cross section contains morphological parameters such as the Chord Length Distribution (CLD) and the projected area. Then, one can show that the scattering cross sections averaged over all orientations can be written as:

$$\langle C_N^r \rangle = 2r^2 \langle S_p \rangle \int_0^{l_{\max}} (1 - \cos xl(m-1))D(l)dl \quad (1)$$

The projected area and the chord length are made dimensionless in Eq.1 by dividing the dimensional area and length by, respectively r^2 and r .

The benefit of this equation lies in the two quantities : the projected area $\langle S_p \rangle$ averaged over all orientations and the chord length distribution $D(l)$. The two quantities are only depending on the morphology of the aggregate and not on the orientation. This modification is called ADr, because the computational time is much shorter than the one of standard AD method.

CLD is presented as follows: $D(l)dl$ represents the number fraction of the chord length in the range $[l, l+dl]$. Thus, $D(l)$ obeys the normalization equation:

$$\int_0^{l_{\max}} D(l)dl = 1 \quad (2)$$

l_{\max} is the maximum chord length of an aggregate.

2.2. Analytical evaluation of CLD

With the aim of obtaining an analytical CLD, it is necessary to study the CLD got by a simulation using the algorithm presented in [4]. Figure 1 presents the CLD for a compact aggregate formed by 16 primary particles. The simulated CLD, as shown in [4], contains three contributions. The corresponding decomposition can be generalized to other aggregates.

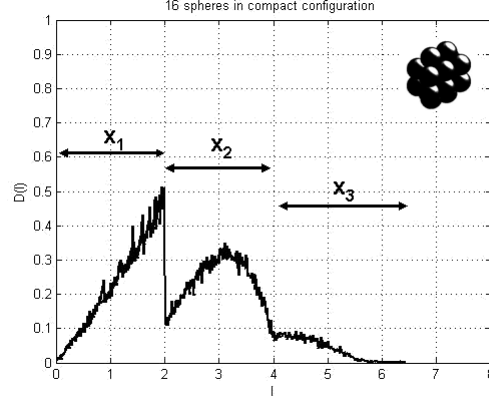


Fig. 1: The three sub-distributions represent a part of the total chord length distribution for aggregates. Case of an aggregate with 16 primary particles in compact configuration

2.2.1. CLD formulation

The aggregate chord length distribution will be written as follows :

$$D(l) = Da(l) + Db(l) + Dc(l) \quad (3)$$

The aggregate chord length distribution was divided into three sub-distributions corresponding to:

- the elementary structure (a sphere), for the chord lengths in the l -range from $[0;2]$:

$$Da(l) = x_1 D_1(l) \quad (4)$$

with

$$D_1(l) = l/2 \quad (5)$$

- the two-sphere aggregate characterized by $D_2(l)$, with a chord length range $[2;4]$:

$$Db(l) = x_2 \frac{D_2(l \in [2,4])}{\int_2^4 D_2(l) dl} \quad (6)$$

Note that the chord length distribution for a two-sphere aggregate is analytically available [4, 6]:

$$D_2(l \in [2,4]) = \frac{1}{16 \left(1 + \frac{8}{3\pi} \right)} \left[l \frac{16-l^2}{16+l^2} + \frac{3l^2-16}{8} \left(\arcsin \left(\left(1 + \frac{l^2}{16} \right)^{-1/2} \right) - \arcsin \left(\frac{l}{4} \left(1 + \frac{l^2}{16} \right)^{-1/2} \right) \right) \right]$$

(7)

- the equivalent object for aggregate, characterized by the last sub-distribution lying in the l -range $[4;l_{max}]$ where l_{max} is the diameter of the volume equivalent sphere ($l_{max} = 2N^{1/3}$):

$$Dc(l) = x_3 \frac{D_3(l \in [4, l_{max}])}{\int_4^{l_{max}} D_3(l) dl} \quad (8)$$

$D_3(l)$ corresponds to the reversed chord length distribution of the equivalent sphere in volume:

$$D_3(l) = \frac{2}{l_{max}^2} (l_{max} - l) \quad (9)$$

Each one of these sub-distributions represents a part of the total chord length distribution (e.g. Figure 1); their mutual contributions are peculiar to each aggregate. The values (x_1, x_2, x_3) of the normalized integral ($x_1 + x_2 + x_3 = 1$) are the weighted values of each sub-distribution for an aggregate.

Then, ADr method can be analytically performed if x_1, x_2, x_3 and $\langle S_p \rangle$ are expressed.

2.2.2. Previous relations for x_1, x_2, x_3 and $\langle S_p \rangle$

We previously were interested by a set of ordered aggregates and we expressed x_1, x_2, x_3 and $\langle S_p \rangle$ according to parameters that characterize the aggregate morphology: the number N of primary particles and the morphological parameter d_1 that is defined (Eq. 10) as the average distance between primary particle centres in the aggregate.

$$d_1 = \frac{1}{N(N-1)} \sum_{i,j} d_{i,j} \quad (10)$$

$d_{i,j}$ is the centre-centre distance between i and j particles. $d_{i,j}$ is dimensionless.

The expressions for x_1, x_2, x_3 and $\langle S_p \rangle$ lead to a scattering cross section value in agreement with the one coming from the exact method. In fact, for ordered aggregate the error is smaller than 15%.

3. Morphological Parameters for disordered aggregates

3.1. Introduction

New routes of particle synthesis lead to monodisperse suspensions consisting in monosized spherical particles. The possible aggregates are composed of a few monosized primary

particles. Depending on the locus of the aggregation process, aggregates with different spatial extent are observed. For instance, 1D, 2D and 3D aggregates are, respectively produced:

- in a channel into a microfluidics device or along the streamline in a laminar flow
- on a surface or an interface
- in a stirred chemical reactor.

The space dimension d_E of the aggregation locus is an important parameter.

Aggregation mainly leads to random aggregates. There is an extensive literature dedicated to random aggregation. Most of investigators consider computer simulations for building random aggregates from simple collision mechanisms. Among these models let us quote [7]: the Ballistic Particle-Cluster Aggregate model (BPCA), the Ballistic Cluster-Cluster Aggregate model (BCCA), the Diffusion Limited Aggregation model (DLA), the Reaction Limited Aggregation (RLA). At each mechanism and space dimension corresponds a fractal dimension of the so-built aggregates. Later on, more sophisticated models were developed (see, for instance, [8]). These models may represent some experimental observations as soot agglomerates formation in combustion processes [9, 10] or silica and titania agglomerate formation by flame aerosol processes [11]. However, it is not always the case. For instance, in industrial chemical reactors the produced aggregates contain a few primary particles (less than one hundred) and their morphology cannot be characterized by a fractal dimension. This results from a more complex formation mechanism (see, for instance [12]) than the above-mentioned ones. As the aim of this work is the search of expressions for scattering cross section of any aggregate without reference to their formation mechanism, fractal-like aggregates will not be particularly considered.

3.2. Description



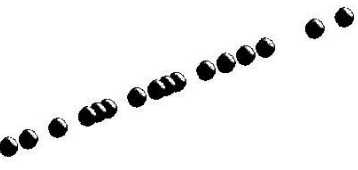
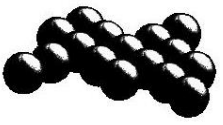
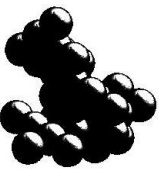


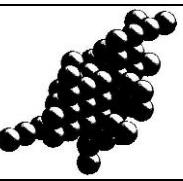
In this article, an aggregate is constituted by identical spherical primary particles. The disordered (random) aggregates are constructed according to the process of particle-cluster aggregation but with no condition on fractal dimension. Each new particle is added to one particle belonging to the aggregate either by a completely randomly way (procedure P') or randomly by checking the angle between triplets equal to 90° or 180° (procedure P). The added particle will be in touch with other ones in aggregate.

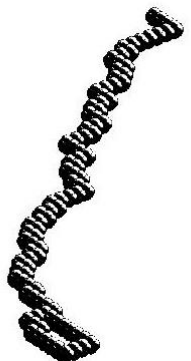

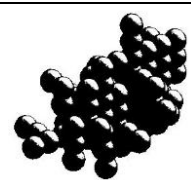
The two procedures are as follows:

- step 0: The space dimension d_E of the aggregate is fixed among the values (linear: $d_E=1$, plane: $d_E=2$, compact: $d_E=3$). The number N of primary particles in aggregate is chosen ($N>2$). An integer n (which will be the maximum co-ordination number for a primary particle in the built aggregate) is chosen.
- step 1 : a primary particle denoted A in aggregate is selected at random ; the number n_A of its linked neighbours (co-ordination number) is determined
- step 2 (procedure P) : if n_A is smaller than n , then a new primary particle B will be in touch with the previous primary particle A. if not, return to step 1. With procedure P, n is strictly smaller than 7.
- step 2' (procedure P') : if n_A is smaller than n , then a new primary particle B will be in touch with the previous primary particle A. if n_A is equal to n or greater than n or if the particle B overlaps another primary particle in aggregate, do not link the particle B and go to step 1.

Table 1 (first column) contains few examples among the 90 aggregates used in this study. The full random process (procedure P') may lead to "compact" aggregates (see figures e,h,k in Table 1). For getting a loose aggregate, it is necessary to limit the number of nearest neighbours of a given particle into the aggregate (see figures f,g,i,j in Table 1).

Table 1 : Examples of disordered aggregates with their geometrical characteristics (N , d_E , $d_1/2$, $\langle S_p \rangle$, x_1 , x_2 , x_3)

	Random aggregate	N	d_E	$d_1/2$	$\langle S_p \rangle$	x_1	x_2	x_3
a		8	2	1,93	2,53	0,82	0,16	0,02
b		8	3	1,81	2,54	0,81	0,18	0,01
c		16	1	11,8	3,89	0,948	0,05	0,002
d		16	2	2,46	3,46	0,8	0,17	0,03
e		32	3	2,57	4,39	0,593	0,31	0,097
f		64	2	9,1	7,2	0,84	0,14	0,02
g		64	3	7,39	6,9	0,76	0,2	0,04
h		64	3	3,32	5,76	0,52	0,32	0,16

i		100	2	23	9,03	0,84	0,14	0,02
j		100	3	11,6	8,65	0,77	0,195	0,035
k		100	3	3,7	6,8	0,45	0,3	0,25

3.3. Results

In this section, the new disordered aggregates depicted above are used to evaluate the accuracy of the ADr method. Firstly the relations previously proposed for ordered aggregates will be tested, then the scattering cross section obtained with ADr method will be compared to the one calculated with AD.

3.3.1. Expressions for x_1 , x_2 , x_3 and $\langle S_p \rangle$

The chord length distribution of an aggregate is obtained by the use of an algorithm introduced in [4] from a 10^6 chords set for a given orientation. 300 arbitrary aggregate orientations of a given aggregate were performed. Then, x_1 , x_2 , x_3 are deduced from the averaged CLD. At the same time, $\langle S_p \rangle$ was obtained as the averaged projected area over 300 aggregate orientations. These x_1 , x_2 , x_3 and $\langle S_p \rangle$ values will be called in the continuation of the text as simulated values. Table 1 collects these values for several disordered aggregates.

Given the number N of primary particles, the space dimension d_E and the maximum coordination number n of a primary particle in the aggregate, a set of aggregates are built. Then, x_1 , x_2 , x_3 and $\langle S_p \rangle$ are calculated for each aggregate. The mean value x_p and the standard deviation σ_p of x_1 , x_2 , x_3 and $\langle S_p \rangle$ were estimated for this set of aggregates. This represents the fluctuation amplitude A_f due to the weak description of aggregates (N , d_E and n). The procedure is performed for different number [4;100] of primary particles, space dimension

[1;3] and co-ordination number [2;6]. Table 2 contains the maximum value of the standard deviation σ_p (for x_1 , x_2 and x_3) and of the standard deviation σ_p over the mean value x_p (for $\langle S_p \rangle$). This two statistical parameters represent A_f .

Table 2: fluctuation amplitude A_f for x_1 , x_2 , x_3 and $\langle S_p \rangle$

	x_1	x_2	x_3	$\langle S_p \rangle$
A_f	0,01	0,01	0,001	0,01

The small observed amplitude allows us for asserting the existence of a relation between (x_1 , x_2 , x_3 and $\langle S_p \rangle$) and (N , d_E and n). However, we prefer to link (x_1 , x_2 , x_3 and $\langle S_p \rangle$) to (N , d_E and d_l) as d_l may represent the aggregate size.

Analytical expressions for x_1 , x_2 , x_3 , $\langle S_p \rangle$ in [4] were obtained from the study of 17 different ordered aggregates. The analytical expressions are summarized in Table 3. In these expressions, appear several morphological parameters depending on N and d_l :

- $N/(d_l/2)^3$ characterizes the compactness of the aggregate; one also defines a compactness index $CI = 3 + \log((N-1)/5(d_l/2)^3)$

- $\left(\frac{d_l}{2} - 1\right)$ shows that only large aggregates have high x_3 values.

Table 3: Analytical expressions obtained for x_1 , x_2 , x_3 and $\langle S_p \rangle$ in [4], ε_1 and ε_2 are the mean standard deviation between the analytical values and respectively the ones obtained by simulation for the 17 aggregates chosen in [4] and the aggregates used in this article (§ 3.2). Asterisk is the mean absolute difference.

	Analytical expressions [4]	ε_1 (%)	ε_2 (%)
x_3	$x_3 = 0.0031\beta^2 + 0.0182\beta$ with $\beta = \left(\frac{d_l}{2} - 1\right) \left(\frac{N}{(d_l/2)^3}\right)^{1.2}$ (11)	1*	1*
Intermediate step : $u = x_2/x_1$	$u = 0.96 \left(\frac{1}{320} \exp((d_E + CI)(1 - N^{-1})) + 0.05\right)$ (12)	18	24
x_2	$x_2 = (1 - x_3)u / (1 + u)$ (13)	13	19
x_1	$x_1 = 1 - x_2 - x_3$ (14)	6	11
$\langle S_p \rangle$	$\langle S_p \rangle = \pi R_p^2$ $R_p = \sqrt{1.25} \left(\left(\frac{d_l}{2}\right)^{1/3} N^{1/2}\right)^{5/8}$ (15)	8	17

Then, the relations for x_1 , x_2 , x_3 and $\langle S_p \rangle$ summarized in Table 3 were tested with all the new disordered aggregates. Table 3 contains ε_1 and ε_2 values. ε_1 and ε_2 are the mean standard deviations between the analytical values and, respectively the simulated ones for the 17 ordered aggregates [4] and for the disordered aggregates used in this article. The larger number of disordered aggregates should reduce the mean standard deviation. Figures 2,3 and 4 represent, respectively x_3 , x_2/x_1 and $\langle S_p \rangle$ values against morphological parameters. One observes that disordered aggregates basically behave as ordered aggregates. However, the representation of x_1 , x_2 , x_3 and $\langle S_p \rangle$ can be improved.

Figure 2 shows a good agreement between x_3 values and approximation (Eq. (11)) for ordered and disordered aggregates but the agreement is less good from Figures 3 and 4 where there is some discrepancy. Figure 3 permits to compare x_2/x_1 and equations 12-14, while Figure 4 shows the difference between $\langle S_p \rangle$ value and equation 15. The sequence of x_2/x_1 values (Figure 3) shows that the equations 12-14 cannot be applied to disordered aggregates. The x_2/x_1 values sequence presents a local maximum around $(d_E + CI)(1 - N^{-1}) \approx 4$, i.e. for aggregates with $d_E = 2$. The not monotonicity of the function indicates that $(d_E + CI)(1 - N^{-1})$ is not a so relevant single variable.

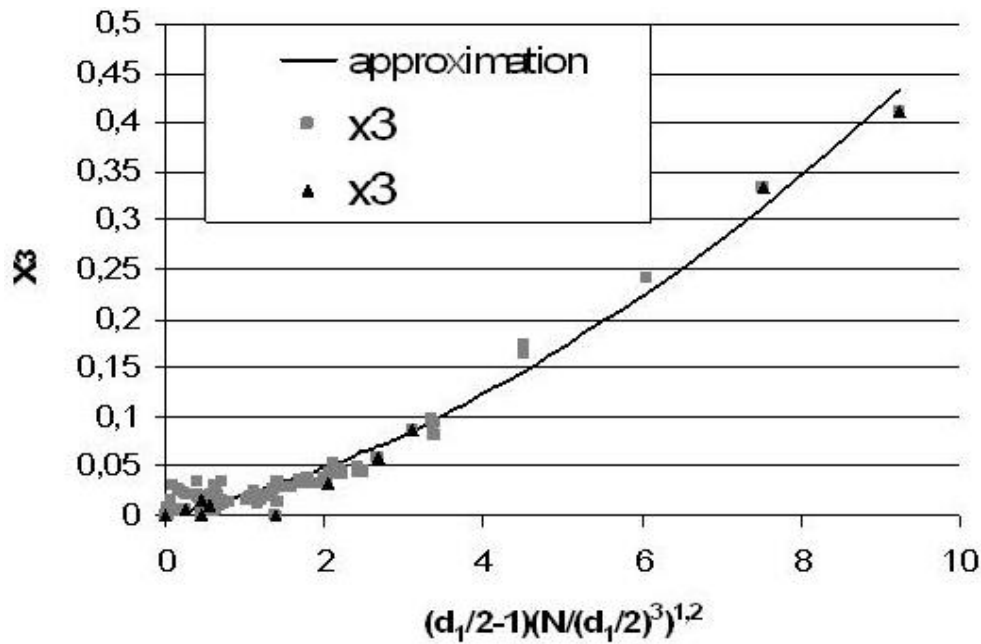


Fig. 2: Evaluation of x_3 according to the equation (11) (square for all aggregate set, triangular for ordered aggregates)

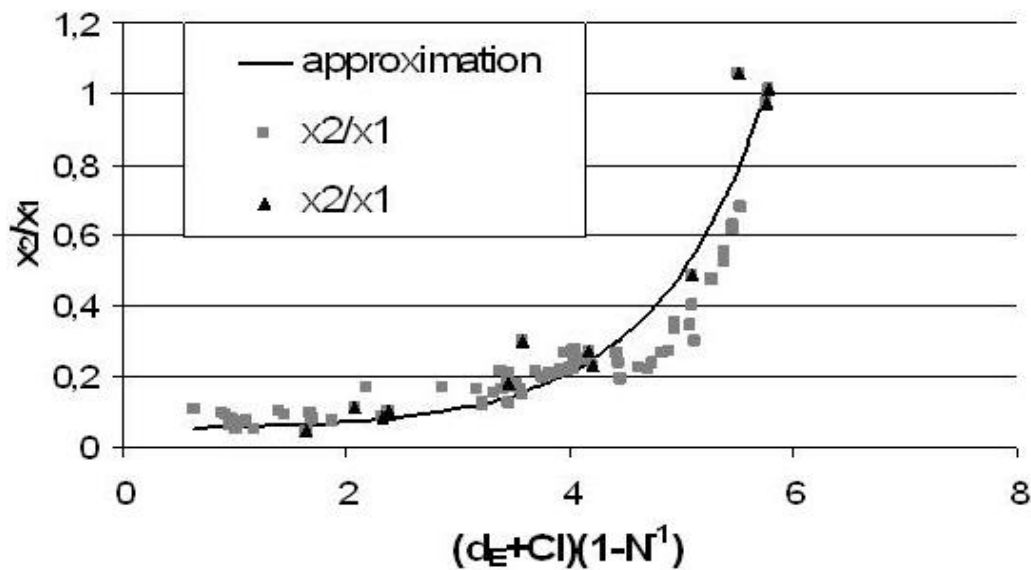


Fig. 3: Evaluation of x_2/x_1 according to the equations (12-14) (square for all aggregate set, triangular for ordered aggregates)

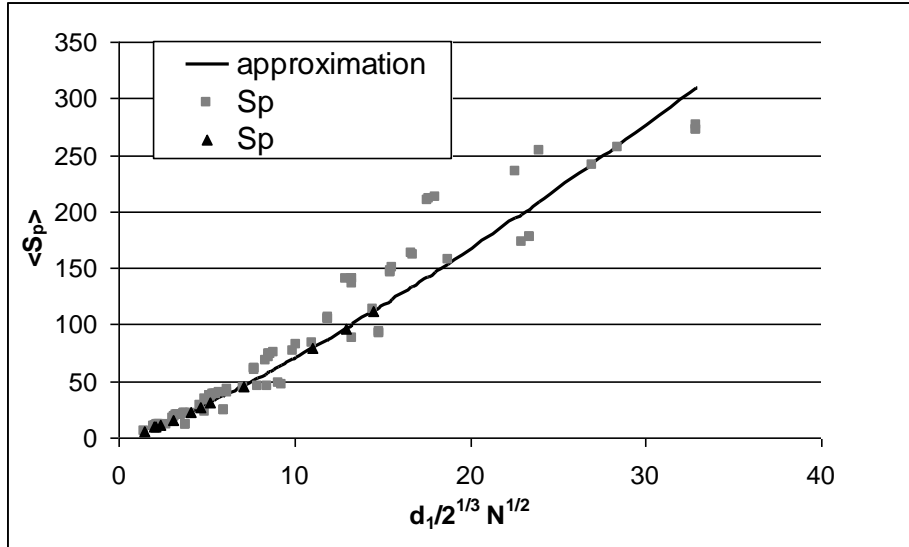


Figure 4: Evaluation of $\langle S_p \rangle$ according to the equation (15) (square for all aggregate set, triangular for ordered aggregates)

We suggest a new expressions set to evaluate x_1 , x_2 , x_3 and $\langle S_p \rangle$ (Table 4). These empirical expressions are simpler and more efficient. ϵ_3 is the mean standard deviation between the analytical values and the ones obtained by simulation.

Table 4: New analytical expressions obtained for x_1 , x_2 , x_3 and $\langle S_p \rangle$, ϵ_3 are the mean standard deviation between the analytical values and the ones got by simulation.

	Analytical expressions		ϵ_3 (%)
X3		(11)	1*
X2	$x_2 = -0.12d_E \ln \left(\left(\frac{d_1}{2} \right)^{1/3} \frac{1}{N^{1/4}} \right) + 0.08$	(16)	12
X1		(14)	3.7
$\langle S_p \rangle$	$\langle S_p \rangle = \pi R_p^2$ with $R_p = \left(\frac{d_1}{2} \right)^{1/5} N^{1/3} d_E^{1/8}$	(18)	9

Concerning the standard deviation values, it is preferable to use equations 11, 14 and 16 in place of expressions included in Table 3. However, we still recommend the use of equations in table 3 for ordered aggregates because the corresponding standard deviations are reduced by one third compared to those coming from equations (11, 14, 16 and 18). Figure 5 represents the x_2 values obtained by simulation and with equation (16); the points furthest away from the approximation correspond to high compactness aggregates with a symmetry centre.

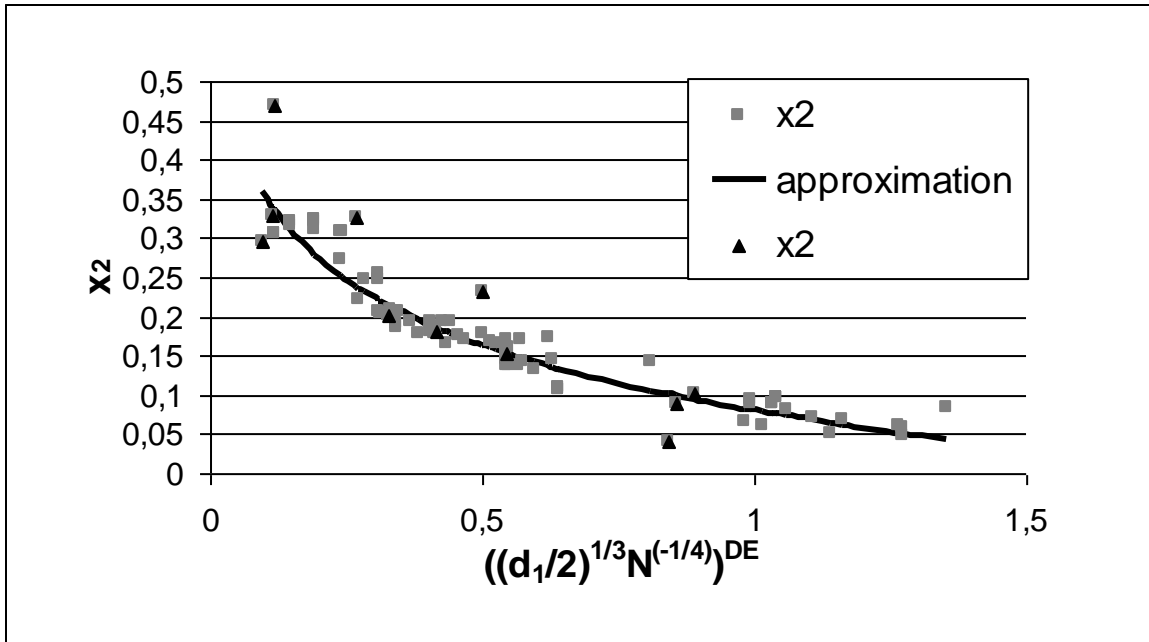


Fig. 5: Evaluation of x_2 according to the equation 16 (square for all aggregate set, triangular for ordered aggregates)

Figure 6 presents the $\langle S_p \rangle$ values obtained by simulation and with equation (18). The corresponding error on $\langle S_p \rangle$ is reduced to 9%. The points, which are the farthest from the curve describing the equation (18) correspond to the 1D non-continuous aggregates consisting in 4 or 8 primary spherical particles. The error on $\langle S_p \rangle$ is reduced to 7.2% if these aggregates are not considered. Thus, the expression (18) is satisfactory for physical aggregates.

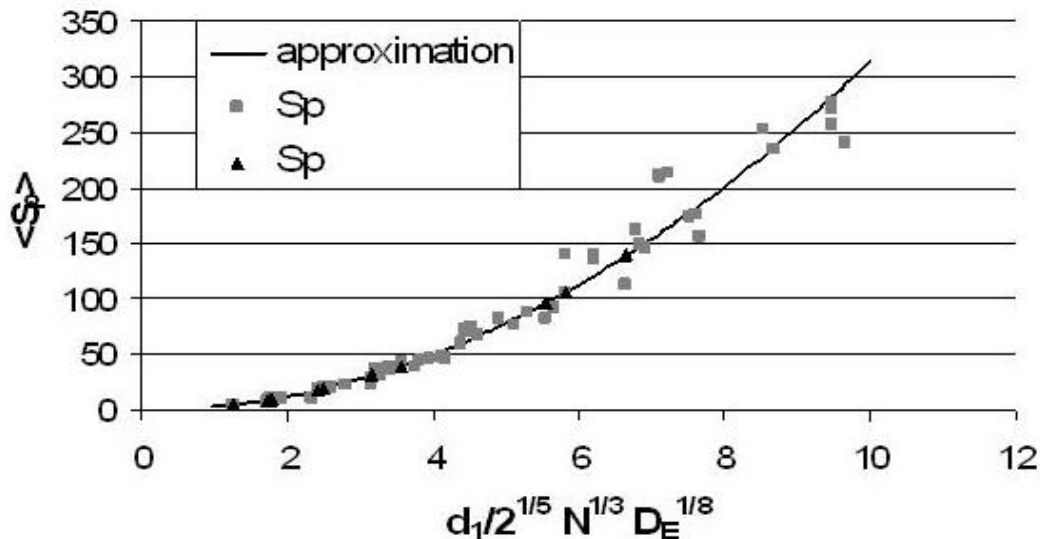


Fig. 6: Evaluation of $\langle S_p \rangle$ according to the equation 18 (square for all aggregate set, triangular for ordered aggregates)

The use of expressions (18), (14), (16) and (11), respectively for $\langle S_p \rangle$, x_1 , x_2 and x_3 leads to having an analytical equation for the aggregates CLD.

3.3.2. Aggregate scattering cross section with ADr method

We studied aggregates with N primary particles in the range [2,100]. The size parameter x of primary particles is in the range [2; 9.25]. For each N -aggregate, several morphologies or compactness were considered. The chosen material is silica SiO_2 . It is considered non-absorbent, i.e. the imaginary part of the relative refractive index m is equal to zero ($\text{Im}(m)=0$). The relative refractive index of silica in water is taken equal to 1.08.

In order to compare the approximated methods (AD and ADr), the R parameter, which is defined as the ratio of the aggregate scattering cross section obtained with the ADr method and AD method, was evaluated.

$$R = \frac{C_{ADr,N}}{C_{AD,N}} \quad (19)$$

The table 5 compares ADr to AD methods, by the mean value of R (over 500 x -values) and the relative standard deviation (σ) between ADr and AD . The results for three silica aggregate space dimensions d_E , are presented below.

Table 5 : Comparison between AD and ADr methods for the primary particle size parameter range $x \in [2,10]$ and different space dimensions d_E .

	d_E	1	2	3
R	mean	1.15	1.03	0.95
	σ	0.04	0.04	0.02

The mean value of R is close to 1 no matter what the space dimension. In the case of 1D space dimension an error is introduced by the presence of not connected primary particles in certain aggregates (see Table 1, c) : however, such configuration has no physical meaning for aggregation. The deviation between AD and ADr methods seems to be smaller than 4 %.

4. Conclusion

ADr is an efficient method making it possible to quickly calculate the scattering cross section of aggregates consisting in optically soft spheres. This method needs the chord length distribution, which can be written as a linear combination of elementary functions. Use of ADr requires the knowledge of the mean projected area $\langle S_p \rangle$ and the weighing coefficients x_1 , x_2 and x_3 . The expressions previously established for $\langle S_p \rangle$, x_1 , x_2 and x_3 in the case of ordered aggregates were tested in the disordered aggregates case (90 tested aggregates). These expressions are suitable for disordered aggregates, but the agreement between exact and calculated values is not so good. This suggests new expressions (x_2 , $\langle S_p \rangle$) and leads to an improved method by determining firstly x_3 , then x_2 and finally x_1 .

In the future, the equations for $\langle S_p \rangle$, x_1 , x_2 and x_3 will be tested for the case of fractal-like aggregates.

All the data can be accessible by sending a request to the authors

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