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# Experimental study of small aggregate settling

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## ABSTRACT

The drag coefficient and hydrodynamic radius of particles are important parameters needed in crystallisation science. Small aggregates of micronic primary particles are mainly produced in stirred crystallisers. We present experimental results on the drag coefficient of macroscopic aggregates consisting in glass beads in the number range [2,100]. The drag coefficient is calculated from settling measurements in glycerol in order to keep the Stokesian nature of typical flow around particles in a crystalliser. We show that the hydrodynamic radius of these aggregates is almost the radius based on the average projected area over all orientation. This result is extended to larger and more porous aggregates.

KEYWORDS : aggregate, sedimentation, Stokesian motion, drag coefficient, hydrodynamic radius

## INTRODUCTION

Aggregation of micronic particles, grains or crystals occurs in stirred tank due to turbulence. The higher the number of primary particles in aggregates, the looser the aggregates. Local shear, then, breaks large aggregates. Fragmentation competes with aggregation resulting in a steady state [1]. In this case it is characterized by a limit size for aggregates in terms of primary particles number. The limit size is often smaller than one hundred [2]. At the same time, sedimentation of aggregates occurs during stirring. Hence, information about aggregates sedimentation is needed in order to understand the evolution of their population in the tank. These data are particularly missing for small aggregates.

In this paper we present results about measurements for settling velocity of several small aggregates. This paper is organised as follows : after a brief review of the background connected with morphology and hydrodynamic resistance of aggregates, we introduce the experimental methodology to study the sedimentation of small aggregates. Then we show the experimental data followed by a short discussion based on comparison with other experimental results and existing theories.

## FUNDAMENTALS

### *morphology*

Large aggregates often have a fractal structure [1]. An aggregate containing  $i$  primary particles of radius  $a_1$  is characterised by its fractal dimension  $D_f$  , its outer radius  $a_i$  , its hydrodynamic radius  $a_{Hi}$  , its gyration radius  $a_{Gi}$  ; as the structure of the aggregates is non-uniform, their local volume density  $\phi_{i,a}(r)$  depends on the distance  $r$  from the centre of mass of the aggregate ; the average volume density is denoted  $\bar{\phi}_{i,a}$  . These different characteristics are correlated by the following equations :

$$a_i = a_1 \left( \frac{i}{S} \right)^{\frac{1}{D_f}} \quad (1)$$

$$\phi_{i,a}(r) = \frac{S}{3} D_f \left( \frac{r}{a_1} \right)^{D_f-3} \quad (2)$$

$$\bar{\phi}_{i,a} = S \left( \frac{a_i}{a_1} \right)^{D_f-3} \quad (3)$$

$$a_{Gi} = a_i \left( \frac{D_f}{D_f + 2} \right)^{1/2} \quad (4)$$

where  $S$  is a structure factor.

From computer simulations, Gmachowski [3,4] found the following correlation between  $S$  and  $D_f$  :

$$S = \left( \sqrt{1.56 - \left( 1.728 - \frac{D_f}{2} \right)^2} - 0.228 \right)^{D_f} \quad (5a)$$

or for  $D_f > 1.5$

$$S \approx 0.42 D_f - 0.22 \quad (5b)$$

The equation (4) is coming from equation (1) for high value of  $i$ .

According to Gmachowski equations 1 and 5b are compatible with equations 6a and 6b.

$$a_{Hi} = a_1 (i)^{\frac{1}{D_f}} \quad (6a)$$

$$a_{Hi} = a_i S^{\frac{1}{D_f}} \quad (6b)$$

This representation is quantitatively supported by simulation results from Chen [5], Kyriakidis [6], Sorensen [7], Rogak [8] and experimental ones from Pusey [9] and Takayasu [10].

Several authors have shown that only large aggregates ( $i > i_{lim}$ ) have fractal structure. For small aggregates, standard relations, which express the relationship between their size and the number of monomers in which they exist, have to be carefully used. However researchers do not agree on the limit value  $i_{lim}$  : for instance, this value, based on the radius of gyration, is nearly 50 for Kyriakidis [6], Sorensen [7], Adachi [11], while it is close to 1 from the hydrodynamic radius measurements of Takayasu [10] by using a technique of sedimentation in density gradient.

In aggregation and sedimentation theories, the relevant parameter is the projected area  $S_p$  of the moving object on a plane. We can thus define the radius  $a_{i,e}$  of the equivalent sphere for an aggregate through :

$$\pi a_{i,e}^2 = \langle S_p \rangle_o \quad (7)$$

Where  $\langle S_p \rangle_o$  is the average of the projected area on all aggregate orientations.

Meakin [12] used computer simulations to investigate collisions between point masses and fractal aggregates ( $D_f = 1.80 ; 1.95 ; 2.09 ; 2.12$ ). He showed that the projected area of aggregates, i.e. the collision cross section, can be written as :

$$\langle S_p \rangle_o = 4a_1^2 (\alpha i^\beta + \gamma i^\delta) \quad 1 \leq i \leq 10^4 \quad (8)$$

The positive parameters  $\alpha, \beta, \gamma, \delta$  depend on the fractal dimension. It can be seen that  $a_{ie} < a_{Hi} < a_i$  with  $a_{ie}, a_{Hi}$  and  $a_i$  respectively calculated from equations 8, 6a and 1.

The corresponding average volume density, then, is :

$$\bar{\phi}_{i,e} = i \left( \frac{a_1}{a_{i,e}} \right)^3 \quad (9)$$

Then, by using equation (3) for the relationship between  $a_{i,e}$  and  $\bar{\phi}_{i,e}$ , one can define a weak (in the mathematical sense of the word) fractal dimension  $D_{wf}$ . This new definition [2] can only be applied to aggregates with high (quasi spherical) symmetry. It is particularly suitable for small aggregates which contain accurately located primary particles. Hence this description is more realistic than an extrapolation of the fractal one to small aggregates.

It has been shown that aggregates in a stirred tank are small and slightly porous. Weak fractal dimension was found in the range [2.35-2.45]. In this case it will be expected that  $a_{ie}, a_{Hi}$  and  $a_i$  will have similar values.

#### *Hydrodynamic resistance and settling*

The forces balance applied to one settling particle (aggregate) takes into account the weight, the buoyancy and the drag force :

$$m \left( 1 - \frac{\rho_L}{\rho_S} \right) g = \frac{1}{2} \rho_L v^2 S_p C_D \quad (10)$$

$m, S_p, v, \rho_L, \rho_S, g, C_D$  are, respectively, the aggregate mass, projected area, velocity, liquid density, solid density, gravity and drag coefficient. The drag coefficient depends on the aggregate permeability, external rugosity and shape.

If the settling particle is a sphere with radius  $a$  and the motion is Stokesian, the drag coefficient obeys the classical relation :

$$C_D = \frac{24}{\text{Re}} \quad (11)$$

where  $\text{Re}$  is the Reynolds number. Equation 10 becomes :

$$m \left( 1 - \frac{\rho_L}{\rho_S} \right) g = 6\pi\mu a v \quad (12)$$

$\mu$  is the dynamic viscosity.

In the case of geometrically complex particles, equation 11 must be modified by introducing the drag corrective coefficient  $\Omega$  :

$$C_D = \frac{24}{\text{Re}} \Omega \quad (13)$$

Equations 12 and 13 can be rewritten as :

$$m \left( 1 - \frac{\rho_L}{\rho_S} \right) g = 6\pi\mu a_H v = 6\pi\mu a \Omega v \quad (14)$$

We shall briefly describe the different cases found in practice. For each case, relevant expressions for  $a$  and  $\Omega$  will be used.

- *non spherical impermeable particle* :

For any isometric sphere [13,14], the drag coefficient is calculated by the following procedure : The Reynolds number is defined as

$$\text{Re} = \text{Re}_v = \frac{\rho_L v d_v}{\mu} \quad (15)$$

where  $d_v$  is the volume equivalent diameter,  $V = \frac{\pi}{6} d_v^3$ .  $V$  is the solid mass volume.

For isometric particles the corrective factor  $\Omega$  is a function of the sphericity factor  $\Psi$ :

$$K_s = \Omega^{-1} = 0.843 \text{Log} \frac{\Psi}{0.065} \quad (16)$$

with  $\Psi = \frac{\pi d_v^2}{S} = \frac{d_v^2}{d_s^2}$  where  $S$  is the particle area..

Ro (14) expresses  $K_s$  as :

$$K_s = \frac{d_v}{d_s} \quad (17)$$



The two expressions for  $K_S$  give the same results if  $\psi > 0.5$ .

For other, non spherical shapes, the particle takes a preferential orientation depending on the Reynolds number. Then, the moving particle is characterized by its projected area  $S_p$  on the plane perpendicular to the sedimentation direction. Coulson [15] proposed to take :

$$\text{Re} = \text{Re}_p = \frac{\rho_L v d_p}{\mu} \quad (18)$$

$$\text{with } S_p = \pi \frac{d_p^2}{4}$$

Given equations 18 and 16, the drag coefficient can be expressed as :

$$C_D = \frac{24}{\text{Re}_p K_S(\psi')} \quad (19)$$

$$\text{with } \psi' = \frac{d_p^2}{d_s^2}$$

$$\text{or } F = \frac{3\pi\mu d_p v}{K_S(\psi')} \quad (20)$$

The equivalent diameter  $d_p$  being the relevant parameter.

- *spherical permeable particles* :

The drag corrective coefficient was calculated for large aggregates or porous spheres. Neale et al. [16] compared different models for aggregates with uniform porosity. Veerapaneni [17] and Kusters [18] consider fractal aggregates (consisting in  $i$  primary particles ;  $a = a_i$ ) for which  $\Omega = \Omega_i$  is a function of  $D_f$  and  $a_i/a_1$ . By using Neale and Veerapaneni's work, Vanni [19] shows that a good approximation for  $\Omega_i$  ( $D_f > 2$ ) is

$$\Omega_i = \Omega_{perm} = \frac{2\alpha^2(1 - \frac{\tanh \alpha}{\alpha})}{2\alpha^2 + 3(1 - \frac{\tanh \alpha}{\alpha})} \text{ with } \alpha = \frac{a_i}{\sqrt{k}} \quad (21)$$

where  $k$  is the aggregate permeability at the aggregate surface. Vanni proposes to use the permeability expression of a material with uniform porosity which is a function of the porosity or the inner solids volume fraction  $\phi$  :

$$k = \frac{2a_1^2}{9\phi} \frac{L(\phi)}{E} \quad (22)$$

where  $L(\phi)$  and  $E$  are respectively the Happel's function [20] and the shielding factor [19] :

$$L(\phi) = \frac{3 - 4.5\phi^{1/3} + 4.5\phi^{5/3} - 3\phi^2}{3 + 2\phi^{5/3}} \quad (23a)$$

$$E = 1 - 0.6e^{-10\phi} \quad (23b)$$

Rigorous models of Veerapaneni [17] and Vanni [19] rests on the same assumptions.  $\Omega_i$  values are slightly different because the description of aggregates by Vanni is more realistic than Veerapaneni's one. It is not possible to find such a simple expression for  $\Omega_i$  for  $D_f < 2$  ; Some authors assume that  $\Omega_i$  only depends on the fractal dimension. Thus, Gmachowski, [3], from different considerations suggests the following expression :

$$\Omega_i = \Omega = S^{1/D_f} \quad (24)$$

Johnson [21] carried out experiments on the sedimentation of large aggregates in water. Each aggregate contains at least  $10^5$  micronic primary particles. Settling velocity, projected area (and its equivalent diameter) and primary particles number per aggregate are measured. Results are expressed as drag coefficient :

$$C_D \approx \frac{c}{\text{Re}_p^b} \quad (25)$$

If  $D_f > 2$  (2.19 ; 2.25) then  $c = 0.6$  and  $b = 1.04$

If  $D_f < 2$  (1.79) then  $c = 0.14$  and  $b = 1.31$

It seems that errors appear in the Johnson's paper : figure 7 is not consistent with the data of table 3. We think that the  $c$  value for  $D_f > 2$  is 6 instead of 0.6. Thus, for  $D_f > 2$ ,

fluid resistance obeys Stokes' law with an  $\Omega$  coefficient equal to 0.25. However, calculations using equations [1,3,21,22,23] lead to  $\Omega = 0.98$ . Although the fractal dimension was incorporated into the calculations to account for the change of aggregate porosity with size, permeability functions are still based on the same assumption –that an aggregate is a porous object with an uniform distribution of small spherical particles. Thus, the single-particle-fractal approach underestimates the permeability of aggregates and fails to obtain a correct value for  $\Omega$ . The same research group [21] proposes an explanation to this experimental result. The single-particle-fractal model (Equations 1,3) is replaced by the cluster-fractal model where an aggregate is composed of primary particles separated into individual clusters that are less permeable than the aggregate. The overall permeability of the aggregate depends on the number  $j$  and sizes  $a_c$  of these clusters. Then equation 1 is replaced by :

$$a_i = a_c \left( \frac{j}{S} \right)^{D_f} \quad (1b)$$

Porosity and permeability are calculated in the cluster-fractal model by using (1b). Then,  $\Omega$  is obtained from equations (21-23). It should be pointed out that  $S$  is a constant, equal to 0.25 for Li [22].  $\Omega$  values predicted using the cluster-fractal model compare well to other experimental results for large aggregates [23] :  $\Omega = 0.37$  for  $D_f = 1.81$  and  $\Omega = 0.5$  for  $D_f = 2.33$ . For dense aggregates, improved predictions [24] of  $\Omega$  are obtained with Gmachowski's equation [25] for  $L(\phi)$  (23c) in place of Happel's function. Gmachowski's equation was proved to be suitable for small aggregates. It well approximates the Ergun equation in the porosity range characteristic for stationary beds and its limit value is 1 for very dilute disperse system.

$$L(\phi) = \frac{(1-\phi)^3}{(1-\phi)^3 + \frac{25}{3}\phi^{4/3}} \quad (23c)$$

The range of validity of all these theories [19] is  $\frac{a_i}{a_1} > 8$ , i.e.  $i > 150$  for  $D_f = 2.5$ ,  $i > 90$  for  $D_f = 2.3$ ,  $i > 50$  for  $D_f = 2.1$ .

Few researchers paid attention to the behaviour of small aggregates ( $i < 100$ ). As the impermeable sphere model seems more suitable as the aggregate porosity decreases, it is likewise for small aggregates, which are often compact. Gmachowski [26] shows that for small or embryonic aggregates,  $\Omega$  can be calculated from (1,21,22,23c) with a shielding factor equal to 1. Calculated values are in agreement ( $\pm 5\%$ ) with cited experimental results for aggregates with  $i < 14$ . The same agreement is found with equation (24) for  $i < 100$ . This calculation was used [26] to estimate  $\Omega$  values for aggregate with a self similar structure.

## EXPERIMENTAL PART

### *Principle*

In order to accurately and easily determine the friction coefficient for small aggregates with  $i$  primary particles, we build them at a macroscopic scale. However, in order to keep a similar flow around these obstacles ( $Re \ll 1$ ), the suspending medium for sedimentation experiments is a high viscosity liquid as glycerol. The forces balance applied to one settling aggregate takes into account the weight, the buoyancy and the drag force. One can easily deduce the drag corrective coefficient :

$$\Omega_i = \frac{m_i \left( 1 - \frac{\rho_L}{\rho_S} \right) g \tau_i}{6\pi\mu_i H} \quad (26)$$

where  $\tau_i$ ,  $H$  are respectively the settling time and the settling distance.

### *Construction of macroscopic aggregates*

Aggregates consist in one millimetre diameter glass beads (type mag : 40B, Merck Eurolab). In order to obtain a narrow size distribution, glass beads were sifted : the diameter is  $0.97 \pm 0.07$ mm. The density of glass is equal to  $2500 \text{ kg.m}^{-3}$  . In order to bind the beads together, two binding agents were used : an organic binder sticks and maintains the beads in place, while an inorganic agent strongly binds their after heating. The organic binder (type 400 – A) was purchased from E.S.L Europe. The inorganic binder (Fondant S334) was purchased from Cerdec France SA.

Glass beads are covered with a mixture of the two binders and then, placed on a grid in order to build 2D aggregates. 3D aggregates consist in several 2D aggregates. Rigidity is obtained by heating : the organic binder evaporates and the inorganic one sticks the glass beads to each other as the temperature exceeds  $565^{\circ}\text{C}$ . The mass ratio of inorganic binders in the aggregates is lower than 3%.

The heating parameters are the following :

- Heating rate :  $8^{\circ}\text{C} / \text{min}$
- temperature :  $650^{\circ}\text{C}$
- heating duration : 2 H
- cooling rate :  $5^{\circ}\text{C} / \text{min}$

1D, 2D and 3D aggregates containing a primary particles number in the range [2,100] was built by this procedure.

### *Calculation of the equivalent radius of aggregate*

Aggregate will be characterized by its equivalent radius  $a_{i,e}$  following Eq (7) or by the

dimensionless equivalent radius  $\beta_i = \frac{a_{i,e}}{a_1}$  .  $\beta_i$  is evaluated by direct calculation for very small

aggregate ( $i \leq 8$ ) and/or by image analysis ( $i > 8$ ). The weak fractal dimension [2] of intermediate aggregates ( $30 < i < 100$ ) was equal to  $2.5 \pm 0.05$ .

#### *Measurement of settling velocity*

Settling velocity is obtained from settling time measurements in a cylindrical container. Containers with two different diameters were used,  $D = 33\text{mm}$  and  $60\text{mm}$ . Each container was filled with glycerol. The temperature was measured with an accuracy of  $0.1^\circ\text{C}$ . Glycerol density is equal to  $1260 \text{ Kg/m}^3$ , as the temperature is in the range  $[20^\circ\text{C}-25^\circ\text{C}]$ . Dynamic viscosity  $\mu$  strongly depends on the temperature; in order to determine it, following relationship [27] could be used:

$$\ln \mu = -237.03 + \frac{16739}{T} + 31.734 \ln T \quad (27)$$

However, due to the high sensitivity of glycerol viscosity to temperature and contaminants, one proceeds by comparing the sedimentation velocity of an aggregate with that of a primary particle under the same operating conditions. Hence, the ratio  $\frac{\Omega_i}{\Omega_1}$  of drag corrective coefficients for the aggregate and primary particle will be calculated from Eq. (26). As  $\Omega_1=1$ ,  $\Omega_i$  is immediately deduced.

First, we made sure of the wetting of aggregates by glycerol and we verified that steady motion was reached.

Accuracy over the deduced drag corrective coefficient is about 7%.

## RESULTS

### *Preliminaries*

Most of the trajectories are rectilinear along the container axis. However, in a few cases, the aggregate trajectory is not rectilinear : aggregates may move towards the wall and settle along it. In this case the settling velocity is about 10% smaller than the ones along axis. It is not easy to foresee orientation of aggregate during settling. Trends are the following : the 1D aggregates axis is vertical during sedimentation, the 2D aggregate plan is horizontal, while 3D aggregates settle without preferred orientation.

The container walls exert a retarding effect known as the wall effect. This is taken into consideration in the case of rigid spherical particles by multiplying the terminal velocity as computed from Stokes's law by the factor  $K$ . The factor  $K$  depends on the ratio  $d/D$ . Francis [28] proposed the following expression :

$$K = \left( \frac{1 - \frac{d}{D}}{1 - 0.475 \frac{d}{D}} \right)^4 \quad (28)$$

Francis' equation produces predictions on the terminal velocity practically coincident with the more complicated relationship described by Clift [29]. The relative deviation is smaller than 0.5% in the range  $0 < \frac{d}{D} < 0.2$ .

Aggregates sedimentation velocity was measured in the two containers. Settling data are given after wall effect correction by taking  $d = 2a_{ie}$ . As the two  $\Omega$  values set are very close, only mean values will be given.

### *Validation*

Plenty of data are available for doublets. The resistance of a pair of contacting particles is less than twice that of a single particle. According to Batchelor [30], the reduction factor for the

resistance due to the motion of two touching spheres along the line of centres is  $Fr_{\parallel}=0.645$ , while for motion across such line is  $Fr_{\perp}=0.725$ . With this definition,  $Fr$  and  $\Omega$  are related by :

$$\beta_2 \Omega_2 = 2Fr$$

We found experimentally ( $D=60\text{mm}$ ) that  $Fr_{\parallel}=0.62$  and  $Fr_{\perp}=0.71$ . This confirms the validity of the experimental methodology.

### *Results and discussion*

Tables 1a and 1b contain, for the main oligomers, the normalized equivalent radius (from average projected area)  $\beta_i$  and the drag corrective coefficient  $\Omega_i$ .  $\Omega_i$  is the average value from several velocity measurements, during which any orientation of aggregate is possible.

Taking into account of the wall effect leads to  $\Omega$  values which do not depend on  $D$  and  $\beta_i$ . This in particular is equal to  $1.04 \pm 0.05$ .

So, the hydrodynamic radius will be expressed as :

$$a_{Hi} \square 1.04 a_{i,e} \tag{29}$$

In other words the hydrodynamic radius is about the equivalent radius (from the average projected area).

Figure 1 compares hydrodynamic radius values from this study (Equation (29) :  $a'_{Hi}$ ) and the ones of Gmachowski [26] (equations (1,21,22,23c) :  $a_{Hi}$ ) as the relative deviation  $(a_{Hi} - a'_{Hi})/a'_{Hi}$ . Except for linear hexamer a systematic relative deviation equal to about 0.06 occurs.

Linear aggregates need special attention. Table 2 compares normalized hydrodynamic radius values  $\frac{a_{H,i}}{a_1}$  for linear aggregates from different authors : Rogak [8], Gmachowski [26] and present authors (Eq (29)). Rogak presents theoretical results in case of cross ( $\perp$ ) flow and



parallel ( $\parallel$ ) flow around spheres chains. Gmachowski and the present authors do not consider a particularly oriented flow. Gmachowski's values are close to the  $\top$  values of Rogak. Our values are set in the middle of the  $[\top, \parallel]$  range.

For all aggregates considered in this study, the notion of porosity and permeability are not suitable, since they consist of a small number of primary particles and they are linear or rather compact. In this paper, we show that the diameter based on projected area plays a leading role. Then we propose to extend these results to any aggregate as follows. We have to take into account the geometrical cross section (projected area perpendicular to flow direction), roughness and permeability ; considering the previous relations (Eq (19,20,21,22)), we suggest to modify them as follows :

$$C_D = \frac{24}{\text{Re}_p K_S(\psi')} \Omega_{perm} \quad (19b)$$

$$\text{or } F = 6\pi\mu a_1 v \frac{\beta \Omega_{perm}}{K_S(\psi')} \quad \text{with } \psi' = \frac{d_P^2}{d_S^2} \text{ and } \beta = \frac{d_P}{2a_1} \quad (20b)$$

For a doublet,  $\Omega_{perm} = 1$ . When the flow is perpendicular to the doublet axis :  $\beta = 2^{1/2}, \psi' = 1$ ,

$\frac{a_{H,2}}{a_1} = 1.414$  in place of 1.45 (exact value). When the flow is parallel to the doublet

axis :  $\beta = 1, \psi' = 1/2$ ,  $\frac{a_{H,2}}{a_1} = 1.34$  in place of 1.29 (exact value). The experimental value

(average value over all orientations) is  $\frac{a_{H,2}}{a_1} = 1.37$ .

The results of this paper show that  $\frac{\beta}{K_S(\psi')} \approx \left\langle \frac{\beta}{K_S(\psi')} \right\rangle_o \approx \langle \beta \rangle_o$  (average value over all orientations).

For a porous aggregate, the contribution of  $\Omega_{perm}$  will decrease the normalized hydrodynamic radius. The permeability needed will be that of a porous sphere with radius  $\langle\beta\rangle_o a_1$ . To summarize, we suggest to use to express the drag force acting over an aggregate from a fluid :

$$F = 6\pi\mu a_1 v \langle\beta\rangle_o \Omega_{perm}(i, \langle\beta\rangle_o) \quad (30)$$

## CONCLUSION

We measured the settling velocity of macroscopic aggregates consisting of few glass beads in glycerol. The Reynolds number was always smaller than one. We show that the hydrodynamic radius of the aggregates is almost the radius based on the average projected area over all orientation. Porosity and permeability are properties without real meaning for the smallest chosen aggregates ( $i < 11$ ). As the so built largest aggregates ( $30 < i < 100$ ) are characterized by a weak fractal dimension close to 2.5, effect of porosity on drag coefficient is negligible. Hence the obtained results express the influence of external shape and roughness of the body on the drag coefficient. Effect of porosity, i.e permeability, should be only noticeable for large porous aggregates ( $i > 100$  ;  $D_f < 2.5$ ). Making of such aggregates, which is more difficult to do, is in progress. It will be followed by the measurement of their drag coefficient.










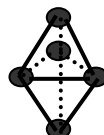






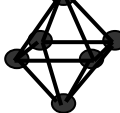


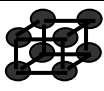
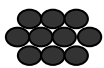
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## LEGENDS OF FIGURES

**Figure1 : relative deviation  $(a_{Hi} - a'_{Hi})/a'_{Hi}$  versus dimensionless radius  $\beta$  for aggregates with  $i < 11$ .**

| Aggregate structure   | $i$ | dimension | $\beta_i$ | $\Omega_i$ |
|---|-----|-----------|-----------|------------|
|    | 1   | 1         | 1         | 1.00       |
|    | 2   | 1         | 1.366     | 1.00       |
|    | 3   | 1         | 1.653     | 1.056      |
|    | 3   | 2         | 1.6       | 1.072      |
|    | 4   | 1         | 1.896     | 1.10       |
|    | 4   | 2         | 1.825     | 1.027      |
|    | 4   | 3         | 1.766     | 1.00       |
|    | 5   | 1         | 2.11      | 0.963      |
|    | 5   | 2         | 2.02      | 1.00       |
|    | 5   | 3         | 1.916     | 0.985      |
|   | 5   | 3         | 1.93      | 1.00       |
|  | 6   | 1         | 2.31      | 0.96       |
|  | 6   | 2         | 2.23      | 1.092      |
|  | 6   | 2         | 2.24      | 1.104      |
|  | 6   | 2         | 2.19      | 1.053      |
|  | 6   | 2         | 2.03      | 1.13       |
|  | 6   | 3         | 2.03      | 0.943      |
|  | 7   | 2         | 2.42      | 0.968      |
|  | 8   | 2         | 2.52      | 0.950      |
|  | 8   | 3         | 2.35      | 0.984      |
|  | 10  | 2         | 2.7       | 1.107      |

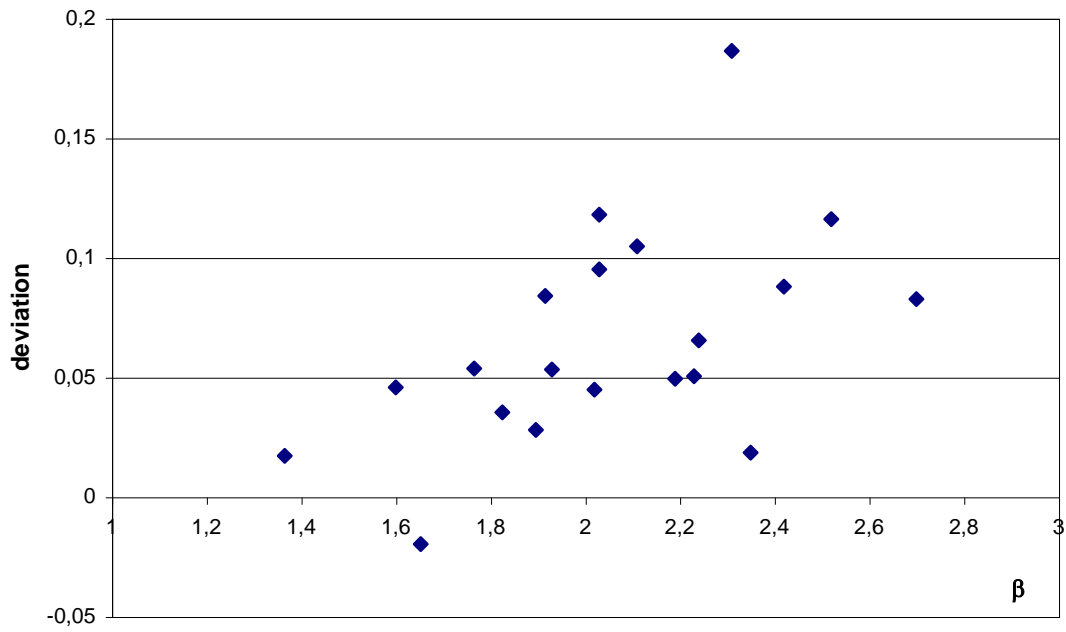
**Table 1a : structure, primary particles number, dimension, normalized radius (from projected area), drag corrective coefficient for  $i < 11$  aggregates**

| $i$ | Dimension | $\beta_i$ | $\Omega_i$ |
|-----|-----------|-----------|------------|
| 12  | 3         | 2.74      | 1.039      |
| 12  | 3         | 2.95      | 1.049      |
| 16  | 3         | 3.45      | 1.12       |
| 16  | 3         | 3.21      | 1.00       |
| 16  | 3         | 3.234     | 1.05       |
| 16  | 3         | 3.261     | 1.17       |
| 16  | 3         | 3.34      | 0.906      |
| 32  | 3         | 4.76      | 1.094      |
| 32  | 3         | 4.53      | 1.18       |
| 32  | 3         | 4.206     | 1.072      |
| 32  | 3         | 4.69      | 1.093      |
| 58  | 3         | 5.4       | 1.076      |
| 62  | 3         | 5.72      | 1.012      |
| 66  | 3         | 6.13      | 1.039      |

**Table 1b : structure, primary particles number, dimension, normalized radius (from projected area), drag corrective coefficient for  $i > 11$  aggregates**

| $i$ | ref. | [8] $\perp$ | [8] $\parallel$ | [26]  | Eq (29) |
|-----|------|-------------|-----------------|-------|---------|
| 2   |      | 1.45        | 1.26            | 1.445 | 1.42    |
| 3   |      | 1.80        | 1.50            | 1.685 | 1.719   |
| 4   |      | 2.11        | 1.745           | 2.027 | 1.976   |
| 5   |      | 2.44        | 1.98            | 2.424 | 2.195   |
| 6   |      | 2.78        | 2.18            | 2.85  | 2.402   |

**Table 2 : normalized hydrodynamic radius for linear aggregates from Rogak [8], Gmachowski [26] and present authors**



**Figure1 : relative deviation  $(a_{Hi} - a'_{Hi})/a'_{Hi}$  versus dimensionless radius  $\beta$  for aggregates with  $i < 11$ .**