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To cite this version:

Marthe Lagarrigue, Sandra Jacquier, Johan Debayle, Jean-Charles Pinoli, Frédéric Gruy. Multi-scale agglomerates: relationship between morphology and optical properties.. Stéphane Veesler. Proceedings World Congress of Particle Technology, Apr 2010, Nuremberg, Germany. 2010. <hal-00967501>

HAL Id: hal-00967501
https://hal.archives-ouvertes.fr/hal-00967501

Submitted on 3 Apr 2014

Multi-scale agglomerates: relationship between morphology and optical properties.

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1) INTRODUCTION

Multi-scale solid particle aggregates designate aggregates of clusters, which are composed of smaller clusters, themselves composed by aggregated solid particles. There are usually 3 or 4 relevant scale levels of agglomeration. The final aim of this study is to define the relationships between the geometrical and optical characteristics of different aggregates. Indeed, a previous work of Gruy and Jacquier in 2008 [1] and [2] showed the existence of a narrow link between these two aggregate characterizations. The first step consists in modeling multi-scale aggregates with specific geometrical shape and optical properties. Thereafter, the aggregate characterization is performed. Following the Xu theory called GMM (Generalized Multiparticle Mie solution) exposed in his paper [3], the studied optical parameter is the scattering cross section, denoted, Csca in (μm^2). In addition, the optical analysis is focused on aggregates with a high optical index.

Concerning the geometrical characterization, several parameters are studied: the compactness, the number of the primary particles and the volume of the aggregates, successively.

Using different 2D joint (geometrical/optical) representations, the correlation between the Csca values and the geometry of the aggregate is analyzed.

2) AGGREGATES MODELING

a. Geometrical shape

Two scale levels are used for modeling the aggregates. For the first level, particles are modeled by spheres (imposed by the knowledge of the theoretical optical method). Their radius will be equal to 10, 15 and 30 nm, respectively.

On the second scale level, aggregates will have a known overall shape (Figure 1): sphere, cube, cylindrer, oblate and prolate convex hulls respectively. The sizes of these hulls are chosen so that they have the same volume of an equivalent sphere whose diameter is 300 nm. The cylindrical type depends on the size of its basis diameter and its height, k-proportional to the basis diameter, $k>1$. For the ellipsoidal cases, shape axes a, b and c are so that $a=b$, and $c=a/k$, with $k>1$

for the prolate type, and for the oblate type: $0<k<1$. In this paper, we use $k\in\{2, 20\}$ for the prolate case and $k\in\{1/2; 1/20\}$ for the oblate type.

We also consider four types of filling ratios of the hulls (Figure 2): filling at 100% (convex hull filled with particles with the most compact way), 75% and 50% (corresponding to the conservation of respectively 75% and 50% particles of the 100% filling, randomly and uniformly selected), and hollow filling, where only particles of the hull edge of the 100% filling case are conserved.

b. Optical properties

As previously mentioned, this study is focused on multiscale aggregates with a high optical index. For these two reasons, zinc sulfide (ZnS) aggregates are studied, because of the work of Mekki-Berrada *et al.* in 2009 [3], with an absolute optical index equal to 2.35. They are considered in water, whose optical index is 1.765.

The wavelength of the incident beam used is the Helium Neon laser (HeNe): 0.6328 µm in the air.

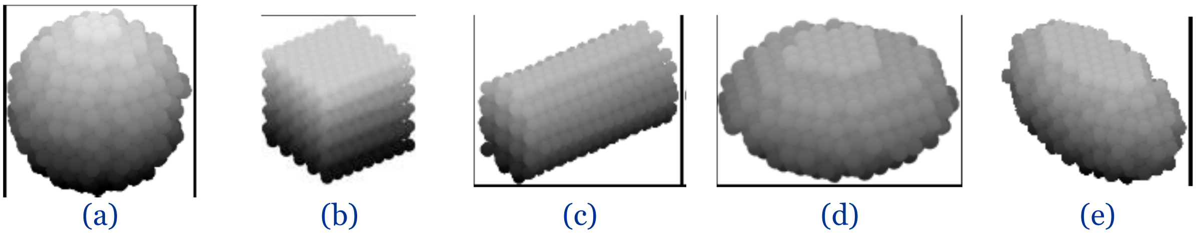

Figure 1: Representation of aggregates with full convex hull, constituted by 750 to 854 primary particles of radius equal to 15nm: (a) spherical, (b) cubic, (c) cylindrical with k=2, (d) oblate with k=2, (e) prolate with k=2, (f) cylindrical with k=20, (g) oblate with k=20, (h) prolate with k=20.

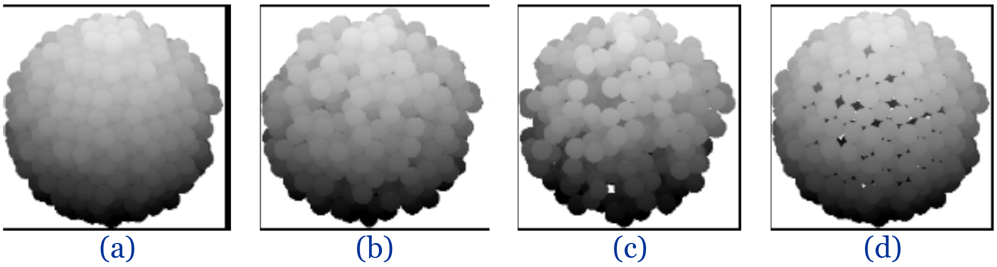

Figure 2: Representation of aggregates having a spherical convex hull with different filling ratios: (a) 100% filling, (b) 75% filling, (c) 50% filling, (d) hollow filling.

3) GEOMETRICAL AND OPTICAL CHARACTERIZATION

On each graph are represented all the aggregates with different convex hulls, filling ratios and radius of primary particles. The average Csca represented corresponds to the mean Csca value for each aggregate. The markers of the legend correspond to the convex hull aggregate geometry.

a. Influence of the filling ratio

Figure 3 shows the average Csca value in the relation with the compactness for the aggregates.

We can see that the aggregates filled at 100%, 75% and 50% are grouped in relation with of their compactness (volume of the primary particles of an aggregate divided by the convex hull volume), independently of the primary particles radius. For hollow filling, it is more chaotic. But it is important to note that, for a same compactness value, several Csca can be obtained: from nearly 0 to 0.16. We can deduce that this geometrical criterion is not decisive for Csca value.

Figure 4 shows the average Csca in relation with the primary particles number constituting the aggregates. It results that the size of the primary particles is a factor that strongly affects the Csca value. Indeed, the aggregates are arranged along curves following the primary particles radius. Besides, it appears that the aggregates are arranged along another criterion: a morphological factor which induces a different behavior above the Csca values. Indeed, for each size of primary particles, we can distinguish two curves of arrangement following the aggregate anisotropy. So, it is shown that the size of primary particles and the anisotropy of the aggregate are decisive geometrical parameters.

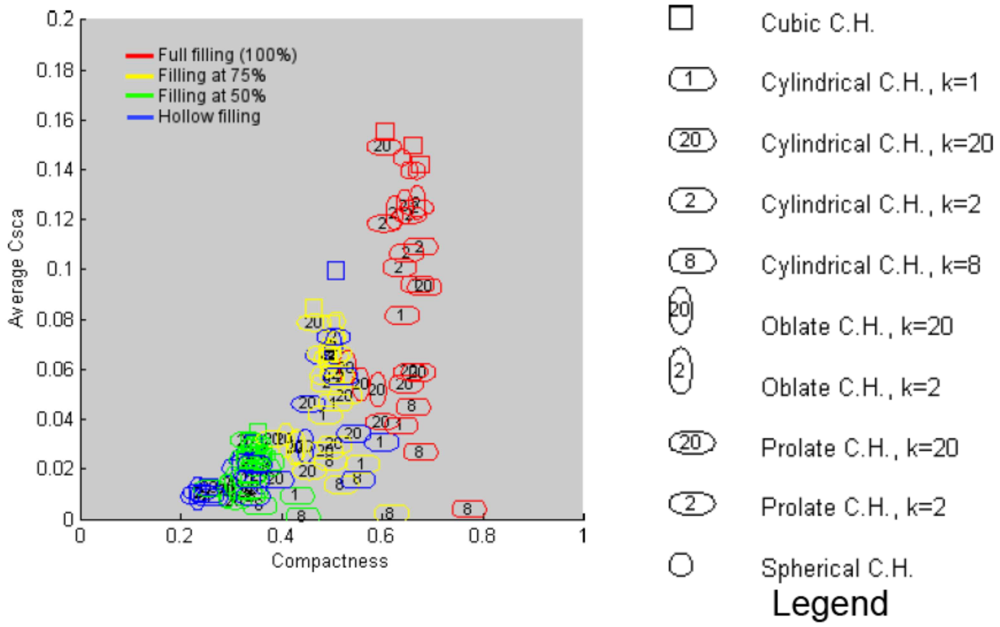

Figure 3: Joint representation of compactness and average Csca for each aggregate. A color distinction is made in function of their filling ratios.

b. Influence of the size of primary particles

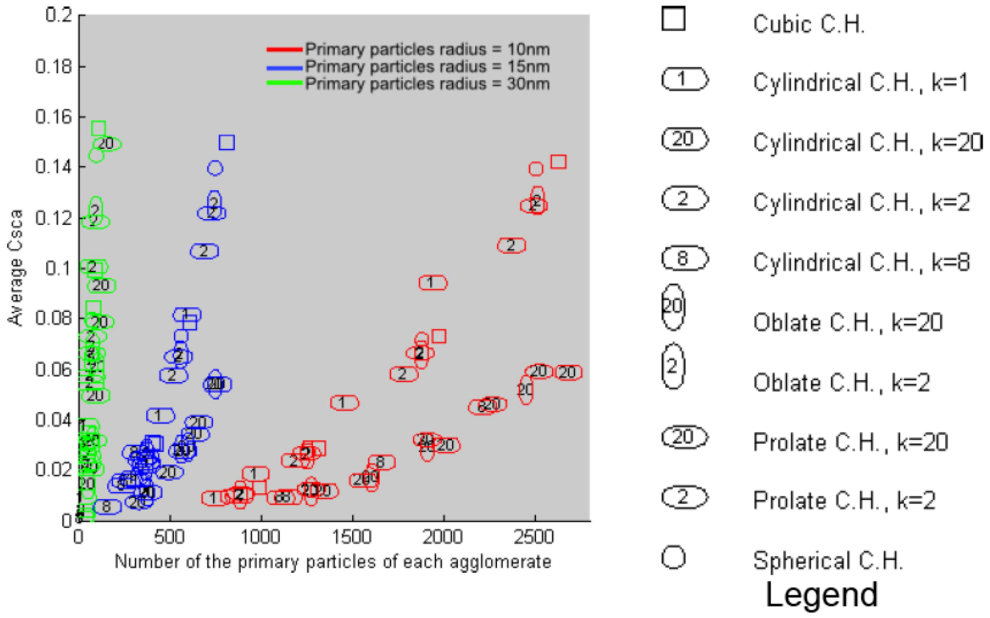

Figure 4: Joint representation of the number of particles and average Csca of for each aggregate. A color distinction is made in function of the size of the primary particle.

c. Influence of the hull anisotropy

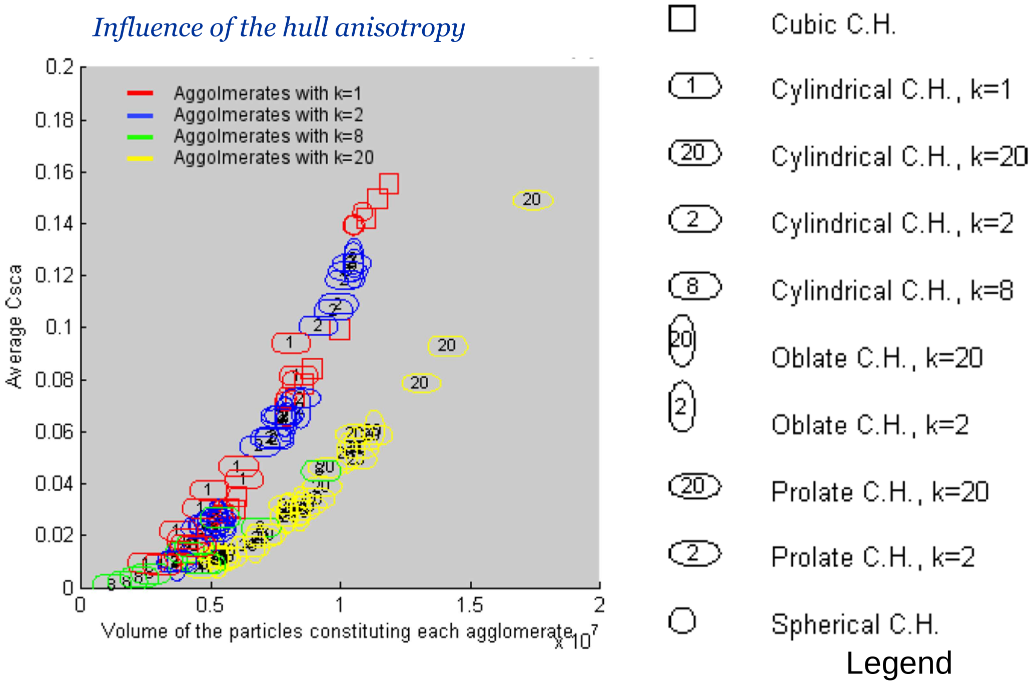

Figure 5: Joint representation of particles volume and average Csca of each aggregate. A color distinction is made in function of the anisotropy of aggregates.

In this graph (Figure 5), the aggregates follow two distinct curves in relation with average Csca and the particles volume. For the first one (which the highest slope), we find all aggregates whose hull is rather isotropic. For the second one: all the anisotropic aggregates. Nevertheless, this graph is not discriminative enough to correlate the anisotropy of aggregates to their average Csca.

4) Conclusion

Using different 2D joint (geometrical/optical) representations, it results that several geometrical parameters affect the mean Csca value of an aggregate to be studied: compactness, number of primary particles, and volume of the articles of each aggregates. A more detailed statistical analysis is needed to correlate the optical and geometrical parameters, by involving a spatial arrangement.

5) References

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