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A new description of the fractal dimension of particle aggregates in liquid medium

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Abstract

The possible existence forms of particle aggregates in liquid medium are classified into four different types according to their morphological characteristics, including the single particles that are separated from each other, the linear aggregates in which all component particles are located in a line, the planar aggregates where all particles are arranged on a plane, and the volumetric aggregates where all particles forms a three-dimensional space. These particle aggregates with different space morphologies have different fractal dimensions and different influence on the rheological phenomena of the solid-liquid system. The effects of various aggregates on the suspension viscosity are analyzed and related with the particle concentration, and then a mathematical model is presented to determine the fractal dimensions of various aggregates by measuring the apparent viscosity of the solid-liquid system. In the model, the viscous fractal dimension is developed as a new concept, the fractal dimensions of different aggregates can be obtained separately and then the relative components of various aggregates experimentally analyzed.

Keywords : particle aggregates; space morphology; viscous fractal dimensions; apparent viscosity.

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1. Introduction

The aggregates constructed by fine solid particles in a liquid medium could be formed due to the compression of double-electric-layer around particles named as coagulation or due to the bridging of polymer compound among particles called as flocculation. A model of aggregate obtained in computer by Vold(1963) many years ago is diagrammatically presented in Fig.1 that not only shows the complicated morphology of the aggregate but also implies the potential difficulty to analyze the aggregate quantitatively. Fortunately, the establishment of fractal theory in 1980's (Mandelbrot, 1977) and its development later in many applied fields (Kaye, 1994) provide a kind of very important approach, i.e. the fractal dimension, to the qualitative and quantitative description of the sophisticated geometry such as aggregates which possess self-similarity in statistics (Gregory, 1997).

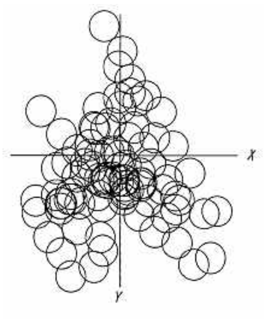


Fig.1 Vold's computer model of aggregates

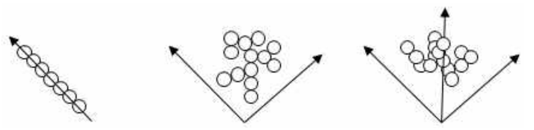
The methods presented so far to determine the fractal dimensions of aggregates can be divided into two classes according to the directly measured objectives in these methods. The first type is that where the measures parameters are only the geometry characteristics of the aggregates. For example, the one-, two- and three-dimensional fractal dimensions (Wang, 2009) obtained in an image analyzer are related respectively with the boundary, area and transformed volume of the aggregate projection, the surface fractal dimension measured by adsorption analyzer is collected with the void distribution inside aggregates, and the size fractal dimension gauged by a particle-size analyzer is dependent on the size distribution of aggregates (Logan and Kilps, 1995). The second type is that the fractal dimension has a bearing on both the size and mass of aggregates, such as the mass fractal dimension gauged with sediment method (Meakin, 1998; Liaoetc., 2005) or light dispersion system (Bushell, 2006; Gregory, 2009).

It is necessary to point out that the aggregate fractal dimension determined by the methods mentioned above is not reflection of all the aggregates in a solid-liquid system although there are a lot of aggregates sampled and analyzed, meanwhile a bigger measurement error may be unavoided because there are two parameters such as both aggregate size and effective density or both aggregate size and projection area to be surveyed at the same time (Fettweis, 2008).

In this paper, the aggregates in a suspended system is firstly classified into different types according to their space mophorlogy, a new concept of viscous fractal dimension presented by authors previously (Xing etc., 2014) is discussed more profoundly, and then a mathematical model in that the viscous fractal dimensions of aggregates can be determined by surveying the apparent visosity is derived in a simplified and easily-unstandable style. Lastly, some experimental results are given to testify the derived model.

2. The types of aggregates in a solid liquid system

In the previous researches (Ding et al., 2013), authors divided the aggregates in a suspended solid liquid system into four types according to their morphological characteristics. One is the single particles separated from each other, they are the left primary particle and (or) the particles broken from other aggregates after flocculation, second is the linear aggregates that look like a line, they are composed of at least two primary particles and the mass centers of all the component particles are located in a straight line, third one is the planar aggregates in that there are at least three primary particles and all the component particles are located in a plane, the last one is named as volumetric aggregates in which there are at least four primary particles and all the component particles are arranged in a three-dimensional space. These aggregates with various morphologies can be demonstrated as Fig.2 where the single particle is looked as a special aggregate and it is easy to find that all the solid particles exist in one of the four types.



Linear aggregate Planar aggregate Volumetric aggregate

Fig.2 Aggregates with different morphologies

The topological dimensions of the various aggregate illustrated in Fig.2 are 0, 1, 2 and 3 for single particles, linear, planar and volumetric aggregates respectively, and the corresponding fractal dimensions of these aggregates should be 0, interval 0~1, 1~2 and 2~3. The purpose of classifying all aggregates into the types shown in Fig.2 is to investigate the influences of different aggregates on the rheology of the suspended system separately and then to make it potential to examine the aggregate composition and further control the composition in practice.

Maybe it is easy to understand that the fractal dimensions of a planar and a volumetric aggregate are between 1~2 and 2~3 respectively, however some explanations about the fractal dimension of 0-1 for a linear aggregate need to be made. Apparently, if all the primary particles in a linear aggregate are linked closely each other, the fractal dimension of the aggregate should be one, otherwise the fractal is less than one. In fact, when two primary particles are coagulated together, their hydrated shells link or overlap each other, if the hydrated shell thickness is much less than the particle size, the fractal dimension of the aggregate is approximately one, and if the hydrated shell is comparable to the particle in size, a fractal dimension less than one should be given. As for the linear aggregate formed by flocculation mechanism of polymer bridging, the aggregate's fractal dimension is obviously less than one.

In the aggregation progress, the four types of aggregates transform from one to another at all times, however when the progress is stable, the composition of different aggregates can be thought of in a state of dynamic equilibrium, in other words, the formation and breaking of various aggregates are in balance. As a result, the apparent viscosity of the suspended system is a function of time but trends to be unchangeable at last as the aggregation reaches a stable stage, and the following analysis on the viscosity and aggregate composition is limited at the stable or balance situation.

3. The model of apparent viscosity and fractal dimensions of aggregates

In the previous work, authors (Shiqiang Ding et al., 2013) have set up a mathematical model of the apparent viscosity of a suspended system and the fractal dimensions of various aggregates by considering the mechanisms of particle collision and aggregate breaking. However, the introduction of fractal dimensions was more or less unnatural in the previous work. In this paper, authors present a new approach to the model by considering the resistance of various aggregates to fluid. It is found that the new approach leads to same destination but looks more simple and understandable.

Denoting the primary particle size for mono-dispersed system or the average size of all primary particles for poly-dispersed system as L_0, the average characteristic size of linear, planar and volumetric aggregates as L_i with $i = 1, 2, 3$ respectively, the primary particle number concentration as n, the number concentrations of single particles, linear, planar and volumetric aggregates as n_i and the average primary particle numbers contained in a single particle, a linear, planar and volumetric aggregate as $n(i), i = 0, 1, 2, 3$ respectively (specially, $n(0) = 1$ for single particles), we introduce following assumptions:

$$n_i \propto n, \quad i = 0, 1, 2, 3 \qquad (1)$$

and $$L_i \propto n, \quad i = 1, 2, 3 \qquad (2)$$

Considering that the mean space distance and collision possibility among all particles will be increased linearly with primary particle number, these assumptions may be easily understandable. In fact, these assumptions can be verified strictly in a longer space, the interested readers may touch authors privately.

In addition, the characteristic size of various aggregates can be related with the number of

component particles within an aggregate and the characteristic size of primary particle in following expression:

$$L_i = \sqrt[i]{n(i)} L_0, \quad i = 1, 2, 3 \qquad (3)$$

and then the by-pass area of single particle and different aggregates noted as (i=1,2,3 respectively) can be written as:

$$S_i = n(i)\frac{\pi L_0^{\ 2}}{4} = \frac{\pi}{4}\left(\sqrt[i]{n(i)}L_0\right)^i L_0^{\ 2-i} = \frac{\pi}{4}(L_i)^i L_0^{\ 2-i} \propto (L_i)^i \propto n^i, \quad i = 0,1,2,3 \qquad (4)$$

The important is that the symbol i in Eq. (3) and (4) is not only a subscript but also the Topological dimension of the single particles, the linear, planar and volumetric aggregates separately.

The resistance of single particles and various aggregates to by-pass of liquid medium, de noted as R_i $(i = 0, 1, 2, 3$ respectively) should be directly proportional to the corresponding by-pass areas:

$$R_i \propto S_i \propto (L_i)^i \propto n^i, \qquad i = 0,1,2,3 \qquad (5)$$

The total resistance of the left single particles and various aggregates after coagulation or flocculation is that (see Eqs. 1 and 5)

$$\sum R_i \propto \sum S_i = n_i S_i \propto n_i (L_i)^i \propto n^{i+1}, \ i = 0,1,2,3 \quad (6)$$

The contribution of single particles and aggregates to the total viscosity of the suspended system is essentially proportional to the resistance:

$$\eta_i \propto \sum R_i \propto n^{i+1}, \qquad i = 0, 1, 2, 3 \quad (7)$$

Noticing that

$$n \propto X \quad (8)$$

where is defined as the volume ratio concentration, i.e. the ratio of all solid particles' volume to liquid volume, Eq. 7 can be rewritten as an equation:

$$\eta_i = \eta^{(i)} X^{i+1}, \qquad i = 0, 1, 2, 3 \quad (9)$$

with $\eta^{(i)}$the coefficients and named here as special viscosity of the single particles and various aggregates.

Then the total apparent viscosity of the suspended system is equal to the sum of the contributions of liquid medium, single particles, linear, planar and volumetric aggregates:

$$\eta^{(i)} \quad (10)$$

where η_m is the liquid medium viscosity. Taking account into the fractal characteristics of the various aggregates, the Topological dimensions of various aggregates should be substituted by the fractal dimensions and Eq.10 becomes:

$$\eta = \eta_m + \sum_{i=0}^{3} \eta^{(i)} X^{i+1} \qquad (11)$$

with D_f $(i = 1,2,3)$ the fractal dimension of linear, planar and volumetric aggregates respectively. The Eq.11 presents a new approach to the measurement of fractal dimensions of aggregates, especially to the determination of the fractal dimensions of aggregates with different morphologies by surveying the viscosity of suspended system. The fractal dimension obtained in this way is defined as viscous fractal dimension by authors to be distinguished from other fractal dimensions.

4. Experimental results

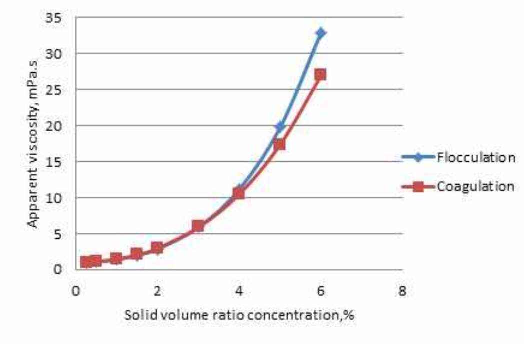


Fig.3 Experimental Results

Two sets of experiments including coagulation and flocculation are arranged to verify the model as shown by Eq.11, and an improved Brookfield DV-III+ coaxial cylinder rheometer is used to measure the apparent viscosity of the suspended solid liquid systems after coagulation and flocculation.

The experimental conditions are listed in Table 1, the measured results are drawn in Fig.3 and some parameters in Eq.11 summarized in Table 2. It can be seen that Eq.11 is certainly a useful model in both chemical and particuological fields and that with the model some important information of the aggregate morphology can be obtained by viscosity measurements. More detailed experiments are in processing and will be reported lately.

5. Conclusions

The fractal dimension of aggregates resulted from coagulation or flocculation is an essential parameter describing quantitatively the complex morphology of coagulates or flocs. The rheological parameters of a solid-liquid stably suspended system should imply some information of the aggregate structure, and the model derived in this paper is to uncover the relation

Table 1. Experimental conditions

	Coagulation	Flocculation
Primary particles	oxide iron powders, average diameter 4.2μm	
Solution pH	6.9	
Solution temperature	20℃	
Shear rate, s^{-1}	110.7	
Flocculants	/	polyacrylamide, with molecular weight 3 million
Flocculants' dose	/	400g/t

Table 2. Model Parameters

	coagulation	flocculation
η_m, mPa·s	1.00	1.00
$\eta^{(0)}$, mPa·s	2.51	2.50
$\eta^{(1)}$, mPa·s	3809	3698
$\eta^{(3)}$, mPa·s	27467	26895
$\eta^{(4)}$, mPa·s	220320	210875
D_{f1}	1.00	0.92
D_{f2}	1.89	1.82
D_{f2}	2.88	2.76

of the apparent viscosity and the fractal dimensions of various aggregates.

The model as shown in Eq.11 presents a new concept, the viscous fractal dimension, and makes it possible that the fractal dimensions of aggregates with various morphologies be distinguished and measured indirectly. Of course, it is necessary to point out that the model can not be directly used to predict the apparent viscosity of a solid-liquid system before the fractal dimensions are determined. In other words, the importance of the model is to determine the fractal dimensions by measuring the viscosity, not to predict the viscosity itself.

6. Nomenclatures

n,	total number of primary particle
i,	subscript with value of 0,1,2 and 3 representing single particles, linear, planar and volumetric aggregates respectively and equaling to the corresponding topological dimensions of the particle and aggregates
$n_i, i=0,1,2,3$,	number of single particles, linear, planar and volumetric aggregates respectively
$n(i), i=0,1,2,3$,	average number of primary particles in a single particle, a linear aggregate, a planar aggregate and a volumetric aggregate respectively
L_o,	characteristic size of primary particle
$L_i, i=1,2,3$,	average characteristic size of linear, planar and volumetric aggregates respectively
S_0,	by-pass area of a single particle
$S_i, i=1,2,3$,	average by-pass area of linear, planar and volumetric aggregates respectively
R_0,	resistance to by-pass of a single particle
$R_i, i=1,2,3$,	average resistance to by-pass of linear planar and volumetric aggregates respectively
X,	volumetric ratio concentration of a suspended system, i.e. the ratio of volume of total primary particles to that of liquid medium
η_m,	viscosity of liquid medium
$\eta_i, i=0,1,2,3$,	viscosity resulted from single particles, linear planar and volumetric aggregates respectively
η,	apparent viscosity of a suspended system
$D_{fi}, i=1,2,3$,	viscous fractal dimensions of linear, planar and volumetric aggregates respectively
$\eta^{(i)}, i=0,1,2,3$,	special viscosity of single particles, linear, planar and volumetric aggregatesrespectively, i.e. the values of η_i as $X=1$

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