FRACTAL AGGREGATION AND BREAKUP OF FINE PARTICLES

by

Bingru LI [a,b]***, Feifeng CAO*** [c*]***, Zhanhong WAN*** [d, e*]***,***
Zhigang FENG [b]***, and Honghao ZHENG*** [d]

[a] School of Mechanical Engineering, Hangzhou Dianzi University, Hangzhou, China
[b] Department of Mechanical Engineering, University of Texas, San Antonio, Tex., USA
[c] Department of Civil Engineering, Zhejiang University of Technology, Hangzhou, China
[d] Ocean College, Zhejiang University, Zhoushan, China
[e] State Key Laboratory of Satellite Ocean Environment Dynamics, Hangzhou, China

Original scientific paper
DOI: 10.2298/TSCI1603797L

Breakup may exert a controlling influence on particle size distributions and particles either are fractured or are eroded particle-by-particle through shear. The shear-induced breakage of fine particles in turbulent conditions is investigated using Taylor-expansion moment method. Their equations have been derived in continuous form in terms of the number density function with particle volume. It suitable for future implementation in computational fluid dynamics modeling.

Key words: *breakage, fractal aggregates, turbulent shear, computational fluid dynamics*

Introduction

Coagulation results in changes in the particle size distribution and a net loss of one particle per coagulation. Friedlander [1] presented the general dynamical equation or population balance equation for fine particles in flow system. The population balance equation can be written as in continuous form and for a homogeneous when the investigation is limited to aggregation and breakage:

$$\frac{\partial n(v;t)}{\partial t} = \underbrace{\frac{1}{2}\int_0^v \beta(v-\varepsilon,\varepsilon)n(v-\varepsilon;t)n(\varepsilon;t)\mathrm{d}\varepsilon}_{B^a(v;t)} - \underbrace{n(v;t)\int_0^\infty \beta(v,\varepsilon)n(\varepsilon;t)\mathrm{d}\varepsilon}_{D^a(v;t)} + \underbrace{\int_0^v a(\varepsilon)b(v|\varepsilon)n(\varepsilon;t)\mathrm{d}\varepsilon}_{B^b(v;t)} - \underbrace{a(v)n(v;t)}_{D^b(v;t)} \tag{1}$$

where $B^a(v;t)$ is increase of particle numbers in certain sizes, v, due to coagulation of smaller ones, ε, and $v-\varepsilon$, $D^a(v;t)$ loss of particles of size, v, by coagulation, $B^b(v;t)$ gain of, v, sized particles from breakup of larger ones and $D^b(v;t)$ loss of, v, sized particles by breakup to give smaller ones. The $n(v;t)$ is the number density function in terms of the particle volume. The $B(v,\varepsilon)$ is the aggregation kernel of two aggregates with volumes, v and ε. The $a(v)$ is the breakage kernel of an aggregate of mass, v.

* Corresponding author; e-mail: cff@zjut.edu.cn, wanzhanhong@zju.edu.cn

The $b(v|\varepsilon)$ is the fragment distribution function that defines the size distribution of the resulting aggregates.

The coagulation kernel depends on the mechanisms causing particles to approach each other and collide. There are several mechanisms that can induce relative movements among particles and, hence, lead to collisions. Breakup occurs to aggregates that cannot withstand external forcing, either from the surrounding flow or from collision with other aggregates. Breakup can be described by means of a breakup frequency, which relates to the shear and the collision with other aggregates.

Higashitani and Iimura [2] studied the breakup of 2-D and 3-D aggregates exposed to shear flow consisting of mono-distributed primary particles interacting by viscoelastic force. The results showed that the average number of primary particles in broken fragments is related to the intensity of the shear stresses. More relevant to the aggregation processes is two microscales, *i. e.*, Taylor microscale, λ, and Kolmogorov microscales, η_k. The Taylor microscale is representative of the energy transfer from large to small scales. The process of energy transfer (or energy cascade) is expressed by the energy dissipation rate per unit mass, ε. For large Reynolds numbers, the structure of turbulence tends to be approximately isotropic, and ε can be approximated:

$$\varepsilon \quad 15\frac{\upsilon u^{*2}}{\lambda^2} \tag{2}$$

where u^* is the isotropic fluctuating velocity scale and ν is the kinematic viscosity of the fluid. The numerical results showed that the average number of primary particles in broken fragments is related to the intensity of the shear stresses and the rate of turbulent shear is denoted within this context:

$$G \quad \sqrt{\frac{\varepsilon}{\upsilon}} \tag{3}$$

with $G \quad u^*/\lambda$ from eq. (1), *i. e.* neglecting the factor $(1/5)^{1/2}$.

The Taylor microscale is not the smallest length scale occurring in turbulence. At very small length scales, viscosity becomes effective in smoothing out velocity fluctuations, hence preventing the generation of infinitely small scales by dissipating small-scale energy into heat. The smallest scale of motion automatically adjusts itself to the value of the viscosity. Relating viscosity, ν, and energy dissipation rate, ε, we can write the Kolmogorov microscales of length, η_k, time, τ_k, and velocity, v_k:

$$\eta_k \quad \sqrt[4]{\frac{\nu^3}{\varepsilon}}, \quad \tau_k \quad \sqrt{\frac{\nu}{\varepsilon}}, \quad v_k \quad \sqrt[4]{\nu\varepsilon} \tag{4}$$

where η_k is the size of the smallest eddies. The small length scales are relevant for the forcing on the aggregates surface and for bringing aggregates distant form one another to collision. If aggregation is predominantly caused by turbulent mixing, the rate of turbulent shear, G, is also expressed as $G = \nu/\eta_k^2$, showing that active interaction between fluid and aggregates in aggregation phenomena occurs at small and very small (Taylor-to-Kolmogorov) scales, and such that $G = \tau_k^{-1}$ [3]. Turbulent mixing induces aggregation and, at the same time, subjects aggregates to higher shear stresses, hence causing breakup if $L \quad \eta_k$. For $L > \eta_k$ there may be aggregates surface erosion. However, experimental investigations and numerical simulations have hardly found aggregates with $L > \eta_k$ [2]. Thus, aggregate surface erosion is unlikely to occur and will be omitted in this paper.

Breakage and moment model

Some porosity aggregates are very sensitive to the electrochemical double layer forces. The primary particles are: rather stable against turbulence action, *i. e.* they do not break up. Low-to-mid values of shear rate (G =10 ~ 100 s^{-1}) aids aggregations, increasing collision among the particles and producing aggregates. In contrast, large aggregates are strongly affected by higher turbulent shear (G > 100 s^{-1}) because the internal bonds are weaker than those of the primary particles. This explains the sensitiveness of the modal aggregate size to G, as observed in several experimental campaigns and numerical simulations [4]. It is qualitatively suggested that the modal aggregate size reaches a maximum at a given G^* for limited residence time. The effect of G is however wider, affecting the full shape of the aggregate size distribution: very high and very small G are in general correlated to left-skewed aggregate size distributions, whereas $G \quad G^*$ result in right-skewed aggregate size distributions.

Aggregates with the same porosity are fragmented in the same fashion. Aggregates often have fractal-like structure, which means that their scales with size with an exponent known as fractal dimension, which in turn determines their density and mechanical properties. The mass-equivalent volume of an aggregate in section i, *i. e.* the volume of a fully coalesced aggregate that consists of x_{i} spherical monodisperse primary particles of diameter, d_{p}:

$$v_{\mathrm{i}} \quad x_{\mathrm{i}} v_{\mathrm{p}} \tag{5}$$

The collision diameter, $d_{\mathrm{c,i}}$, of an aggregate of section i is related to the number of primary particles x_{i} of diameter d_{p} in it, by:

$$d_{\mathrm{c,i}} \quad d_{\mathrm{p}} x_{\mathrm{i}}^{1/D_{\mathrm{F}}} \tag{6}$$

we can derivate next two equations:

$$\sqrt[3]{v_{\mathrm{c}}(v)} \quad \sqrt[3]{v_{\mathrm{p}}}[x(v)]^{1/D_{\mathrm{F}}}, \quad v \quad v_{\mathrm{p}} x(v) \tag{7}$$

The quantity necessary to characterize breakup is the breakup distribution function, that defines the size distribution of the resulting aggregates. Breakup of an aggregate produces a number of smaller aggregates. Theoretical and numerical works have been reported with experimental investigations on the effective aggregate breakup dynamics in literature. For instance, the hypothesis of Friedlander [1] considers a totally reversible process of breakup for which a aggregates resulting from aggregation of any two smaller aggregates would return the previous aggregates. Spicer and Pratsinis [5] have put forth three different hypotheses for the distribution function of the aggregates i resulting from aggregate j: binary, ternary, and Gaussian. The first one expresses that only two daughter aggregates are formed with identical mass, equal to one half of the mother's mass. Ternary breakup produces two aggregates with one quarter the mother's mass and one aggregate with one half the mother's mass. Finally, Gaussian breakup produces a full spectrum of aggregates with a Gaussian mass distribution. Also the numerical results from Higashitani and Iimura [2] mentioned previously suggest that a distribution function is more likely to occur in aggregation breakup. In general, some fragment distribution functions have been proposed although the theory for particle breakage is not well developed as the theory for particle aggregation. An extensive discussion of the problems related to them can be found in [6], where a generalized fragment distribution function is proposed. For uniform distribution, we can derived the equations:

$$b \quad \frac{v}{\varepsilon} \quad 2v_{\mathrm{p}}^{\frac{3}{D_{\mathrm{F}}}} \varepsilon^{\frac{3}{D_{\mathrm{F}}}}, \quad a(v) \quad k_{\mathrm{b}} \frac{\eta(\phi_{\mathrm{tot}})G}{\tau^*}^{q} v_{\mathrm{p}}^{\frac{1}{3}\ \frac{1}{D_{\mathrm{F}}}} v^{\frac{1}{D_{\mathrm{F}}}} \tag{8}$$

In order to use the Taylor expansion method of moments, applying the moment transformation to the general aggregation-breakage eq. (1), we can write two breakage items in terms of the moments of the number density function [7]:

$$M_k^{D^b} \quad \int_0^\infty D^b(v;t)v^k \mathrm{d}v, \quad M_k^{B^b} \quad \int_0^\infty v^k \mathrm{d}v \int_0^v a(\varepsilon)b(v|\varepsilon)n(\varepsilon;t)\mathrm{d}\varepsilon \qquad (9)$$

$k \quad 0$

$$M_0^{D^b} \quad \frac{1}{2}k_\mathrm{b} \quad {\frac{\eta(\phi_\mathrm{tot})G}{\tau^*}}^{q} \quad v_\mathrm{p}^{\frac{1}{3} \quad \frac{1}{D_\mathrm{F}}} \quad {\frac{M_1}{M_0}}^{\frac{1}{D_\mathrm{F}}} \quad \frac{1}{D_\mathrm{F}} \quad \frac{1}{D_\mathrm{F}^2} \quad \frac{M_0^2 M_2}{M_1^2} \quad 2 \quad \frac{1}{D_\mathrm{F}} \quad \frac{1}{D_\mathrm{F}^2} \quad M_0$$

$$M_0^{B^b} \quad 2k_\mathrm{b} \quad {\frac{\eta(\phi_\mathrm{tot})G}{\tau^*}}^{q} \quad v_\mathrm{p}^{\frac{1}{3} \quad \frac{2}{D_\mathrm{F}}} \quad {\frac{M_1}{M_0}}^{1 \quad \frac{2}{D_\mathrm{F}}} \quad \frac{1}{D_\mathrm{F}} \quad \frac{2}{D_\mathrm{F}^2} \quad \frac{M_0^2 M_2}{M_1^2} \quad 1 \quad \frac{1}{D_\mathrm{F}} \quad \frac{2}{D_\mathrm{F}^2} \quad M_0$$

$k \quad 2$

$$M_2^{D^b} \quad \frac{1}{2}k_\mathrm{b} \quad {\frac{\eta(\phi_\mathrm{tot})G}{\tau^*}}^{q} \quad v_\mathrm{p}^{\frac{1}{3} \quad \frac{1}{D_\mathrm{F}}} \quad {\frac{M_1}{M_0}}^{2 \quad \frac{1}{D_\mathrm{F}}} \quad 2 \quad \frac{3}{D_\mathrm{F}} \quad \frac{1}{D_\mathrm{F}^2} \quad \frac{M_0^2 M_2}{M_1^2} \quad \frac{3}{D_\mathrm{F}} \quad \frac{1}{D_\mathrm{F}^2} \quad M_0$$

$$M_2^{B^b} \quad \frac{2}{3}k_\mathrm{b} \quad {\frac{\eta(\phi_\mathrm{tot})G}{\tau^*}}^{q} \quad v_\mathrm{p}^{\frac{1}{3} \quad \frac{2}{D_\mathrm{F}}} \quad {\frac{M_1}{M_0}}^{1 \quad \frac{2}{D_\mathrm{F}}} \quad 3 \quad \frac{5}{D_\mathrm{F}} \quad \frac{2}{D_\mathrm{F}^2} \quad M_2 \quad 2 \quad \frac{5}{D_\mathrm{F}} \quad \frac{2}{D_\mathrm{F}^2} \quad \frac{M_1^2}{M_0}$$

Comments

Breakup can be usually divided into linear and non-linear. The former is induced by turbulent shear, while the latter is due to collision with other aggregates. The total breakup frequency can be computed as linear superposition of them. Furthermore, it increases with volumetric concentration as the number of collisions and rate of non-linear breakup increase. In addition, Kramer and Clark [8] have argued that collision among aggregates can play a role in the breakup process, but McGraw [9] suggests non-linear breakup to occur more frequently in three-body collision. In this paper shear breakup has been estimated to use formulated fragment distribution function. Taylor-expansion moments technique has been developed, which is indeed one of the most accurate and proposing for fluid and turbulent shear [3], Brownian motion [10], and differential sedimentation [11]. The hydrodynamic interaction of a three-body collision is schematized in [12] enabling to consider three-body collision as implicitly accounted for in case of time intervals larger than the breakup time scale. Recently, an investigation of fragmentation processes in particulate material has shown that a power-law distribution characterizes the resulting fragments but no such relation has been investigated in aggregates suspensions. The mentioned mechanisms may have an impact on the stochastic process of redistribution of particles with different sizes and on the restructuring process of aggregates structure [13]. Careful more distribution functions studies are warranted in order to make critical comparisons with the other model predictions and the proposed mechanism of aggregates breakup.

Acknowledgment

This work is supported by the National Natural Science Foundation of China (No. 11572283, 10902097), the National Natural Science Fund of Zhejiang Province (No.

Y6090257) and Open Fund of State Key Laboratory of Satellite Ocean Environment Dynamics (No. SOED0901).

References

[1] Friedlander, S. K., Smoke, Dust and Haze: Fundamentals of Aerosol Behavior, John Wiley and Sons, New York, USA, 2000
[2] Higashitani, K., Iimura, K., Two-Dimensional Simulation of the Breakup Process of Aggregates in Shear and Elongational Flow, *J. Colloid Interface Sci., 204* (1998), 2, pp. 320-327
[3] Cao, F., Wan, Z., *et al*., Numerical Modeling of Fine Particles Fractal Aggregate in Turbulent Flow, *Thermal Science, 19* (2015), 4, pp. 1191-1195
[4] Adler, P., Streamlines in and around Porous Particles, *J Colloid Interface Sci., 81* (1981), 2, pp. 531-535
[5] Spicer, P. T., Pratsinis, S. E., The Evolution of Floc Structure and Size Distribution During Shear-Induced Flocculation, *Water Res., 30* (1996), 5, pp. 1046-1056
[6] Serra, T., Casamitjana, X., Structure of the Aggregates During the Process of Aggregation and Breakup under Shear Flow, *J. Colloid Interface Sci., 206* (1998), 2, pp. 505-511
[7] Yu, M. Z., *et al.*, Numerical Simulation of Nanoparticle Synthesis in Diffusion Flame Reactor, *Powder Technol., 181* (2008), 1, pp. 9-20
[8] Kramer, T., Clark, M., Incorporation of Aggregate Breakup in the Simulation of Orthokinetic Coagulation, *J. Colloid Interface Sci., 216* (1999), 1, pp. 116-126
[9] McGraw, R., Description of Aerosol Dynamics by the Quadrature Method of Moments, *Aerosol Sci. Technol., 27* (1997), 2, pp. 255-267
[10] Wan, Z., *et al*., Method of Taylor Expansion Moment Incorporating Fractal Theories for Brownian Coagulation of Fine Particles, *Int. J. Nonlinear Sci. Numer. Simul., 13* (2012), 7, pp. 459-467
[11] Wan, Z., *et al*., Modeling of Aggregation Kinetics by a New Moment Method, *Appl. Math. Model., 39* (2015), 22, pp. 6915-6924
[12] Li, X., Zhang, J., Numerical Simulation and Experimental Verification of Particle Coagulation Dynamics for a Pulsed Input, *J. Colloid Interface Sci., 262* (2003), 1, pp. 149-161
[13] Zhou, K., Monte Carlo Simulation for Soot Dynamics, *Thermal Science, 16* (2012), 5, pp. 1391-1394

Paper submitted: December 13, 2015
Paper revised: January 4, 2016
Paper accepted: February 2, 2016