SIMULATION AND VERIFICATION OF PARTICLE COAGULATION DYNAMICS FOR A PULSED INPUT

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ABSTRACT

The existing method was improved to simulate the coagulation process. In this method, Smoluchowski discrete equation was directly solved using the fourth order Runge-Kutta method. Through incorporating five different collision results of two colliding particles in different or same sections into calculation process, the coagulation process was simulated exactly. This process avoids the complex conversion process and additional assumptions as the previous works. Mass balance was strictly realized during the whole process. Changes due to different fractal dimension D, shear rate G and different coagulation kernels were demonstrated. Finally, experimental study was used to check the simulation result.

KEYWORDS

Numerical simulation; coagulation; particle size distribution; volume weighted mean size; mass balance

INTRODUCTION

Coagulation and flocculation, that transform many small and slowly settling particles into larger and faster settling aggregates, play an important role in the transport and removal of particulate pollutants and biomass in natural waters and engineered treatment systems. As coagulation proceeds, the sizes of particles constantly change in water. The rate of coagulation depends on the intensity of fluid mixing and other collision mechanisms, particle concentration and size distribution, solution and particle chemistry, structure of the aggregates formed and many other factors. Therefore, adequate description and mathematical modelling of the complex coagulation process remain one of the most fundamental and challenging tasks in the field of environmental engineering.

The kinetic description for changes in particle size distribution induced by coagulation has been established based on Smoluchowski’s equation, i.e.,

\[
\frac{dn_k}{dt} = \frac{1}{2} \sum_{i+j=k} \alpha \beta(i,j)n_i n_j - \sum_{i=1}^{\infty} \alpha \beta(i,k)n_i n_k
\]  

(1)

where the subscripts i, j and k refer to discrete particle size classes; \(n_i\), \(n_j\) and \(n_k\) are the number concentrations of the particle in size classes i, j and k, respectively, the kernel of interaction, \(\beta\), represents the collision frequency function between the particles indicated and \(\alpha\) is the collision efficiency. It has been realised that the two important assumptions in the Smoluchowski approach, the rectilinear collision model and coalesced solid spheres of particle aggregates, are not realistic. Recent research indicates that aggregates formed by coagulation are fractals which have much larger sizes than the coalesced spheres (Jiang and Logan, 1991). In addition, curvilinear collision models have been advanced that count for hydrodynamic and short-range forces between approaching particles (Han and Lawler, 1992). Therefore, to improve the prediction for the rate of particle coagulation and flocculation, the new curvilinear approaches and fractal geometry should be included in modelling the coagulation dynamics.
THEORY AND APPROACH

Numerical Simulation

Sectional Method and the Characteristic Size of Each Size Section. In the present study, the sectional method (Gelbard et al., 1979; Burd and Jackson, 1997) was used to solve the general kinetic equation (1) and thus to carry out numerical simulation. The whole particle size range was divided into a number of contiguous sections in terms of the mass of particles (42 sections in this paper). The upper bound of each section was certain times of its lower bound, i.e., \(m_k = pm_{k-1}\). The coefficient \(p\), which can be varied, was chosen as 2 in this paper.

In each section, the particles are assumed to follow a power-law size distribution function. The average mass of the particles within the section can be calculated by \(\overline{m}_k = 1.38m_k\), which was used as the characteristic size (mass) for computing the collision frequency function of particles involved.

Fractal Aggregates. Aggregates formed by coagulation are highly porous and fractal (Jiang and Logan, 1991). The relationship between the mass and the size of a fractal aggregate can be written as \(m \propto l^D\), where \(l\) is the actual length of the aggregate and \(D\) is the fractal dimension. A fractal aggregate with \(D < 3\) has a larger size than its solid equivalent diameter of the coalesced sphere. The actual length of an aggregate may be determined from its mass (Li and Logan, 1995) by

\[
l = bm^{1/D}
\]

where \(b\) is a constant. In this simulation study, all particles in the entire size range were assumed to have the same fractal dimension and the internal permeation through the aggregates were negligible.

For a pulsed input of particles, the number of particles decreases and the average size of all particles increase due to the formation of particle aggregates. The volume-weighted mean size, \(\bar{\bar{I}}_{av}\), defined as

\[
\bar{\bar{I}}_{av} = \frac{\sum_{k=1}^{\infty} n_k V_k \bar{I}_k}{\sum_{k=1}^{\infty} n_k V_k} = \frac{\sum_{k=1}^{\infty} n_k V_k \bar{I}_k}{\sum_{k=1}^{\infty} n_k V_k}
\]

was used as the representative size for all particles and aggregates, where \(V_k\) is the volume of the particles with the characteristic size of \(\bar{I}_k\) in the \(k\)th section. During the simulation, the total mass of particles in the entire size range was strictly conserved.

Rectilinear Coagulation Kernels. Rectilinear model considers that particles move in straight lines until collisions occur between them. The collision frequency functions for the three collision mechanisms are as follows:

\[
\beta_{Br}(i,j) = \frac{2kT}{3\mu} \left( \frac{1}{l_i} + \frac{1}{l_j} \right) (l_i + l_j)
\]

Brownian motion: \(\beta_{Br}(i,j)\)
Fluid shear:

\[ \beta_{sh}(i, j) = \frac{G}{6} (l_i + l_j)^3 \]  

Differential sedimentation:

\[ \beta_{ds}(i, j) = \frac{\pi}{4} (l_i + l_j)^3 \left| U_i - U_j \right| \]  

where \( k \) is Boltzmann’s constant, \( T \) is the absolute temperature of the fluid, \( \mu \) the fluid viscosity, \( G \) the velocity gradient of the fluid, \( U \) is the settling velocity of an aggregate, which can be calculated by Stokes’ law.

The total collision frequency function is the summation of these three independent inter-particle collision mechanisms, i.e.,

\[ \beta(i, j) = \beta_{br}(i, j) + \beta_{sh}(i, j) + \beta_{ds}(i, j) \]  

Curvilinear Collision Kernels. Curvilinear collision models have been proposed, which include the hydrodynamic effects and short-range forces between colliding particles, to overcome the limitation of the rectilinear method. There are not analytical solutions available for the curvilinear models. Han and Lawler (1992) have recently proposed a comprehensive approach for calculating the curvilinear collision frequency functions from the rectilinear model using

\[ \beta(i, j) = e_{br}\beta_{br}(i, j) + e_{sh}\beta_{sh}(i, j) + e_{ds}\beta_{ds}(i, j) \]  

where \( e_{br} \), \( e_{sh} \) and \( e_{ds} \) are the correction factors for the collision mechanisms indicated. The numerical solutions for approximating the correction factors in the graphical form can be found from the work of Han and Lawler (1992).

Experimental Study

Coagulation experiment was conducted with standard red-dyed latex microspheres 2.8 \( \mu \)m in diameter (Polysciences, Inc.) using a Jar-test device (Phipps & Bird). The standard particles were placed in a 600 mL beaker filled with 3.5\% NaCl solution. Particles were kept suspended by a flat paddle rotated at a rate of 15 rpm. Alum (\( \text{Al}_2(\text{SO}_4)_3\cdot18\text{H}_2\text{O} \)) was added as a flocculent at a concentration 10 mg/L, while 0.1 M NaHCO\(_3\) was used to maintain the solution pH between 7 and 8.

During the initial 2 hours of the coagulation experiment, samples were gently withdrawn from the particle solution at various time intervals using counting cells (GRATICULES, London) each having an effective volume of 1 mL. The particles and particle aggregates in each sample were counted in terms of a series of particle sizes under a microscope (BX60, Olympus), and the size distribution of the particles was therefore obtained.

RESULTS AND DISCUSSION

Simulated Evolution of Particle Size Distribution

Particle coagulation dynamics were well simulated as demonstrated by the evolution in particle size distribution (Figure 1). As coagulation took place, small particles joined together to form larger aggregates. The mass-size distributions were unimodal in shape with a size section corresponding to the peak mass concentration. Driven by coagulation and flocculation, the position of the peak moved with time from the smaller size to the larger size. Both the rectilinear model and curvilinear model gave the similar trend in the change of particle size distributions. However, coagulation predicted by the rectilinear model could occur at a rate nearly an order of magnitude faster than that predicted by the curvilinear model. When the rectilinear model was applied, it took 600 sec for the peak size to grow from 1 \( \mu \)m of the primary particles to about 20 \( \mu \)m, while nearly 5000 sec were needed to see the same growth in peak size if the curvilinear model was used. It has been indicated that the rectilinear model overpredicts inter-particle collisions, but the curvilinear model may underestimate the collision rates for permeable fractal aggregates (Li and Logan, 1997).
Affecting Factors for Coagulation Kinetics

Fractal Dimension. The fractal dimension of particle aggregates seriously affected the rate of coagulation. The kinetics of aggregate formation increased remarkably as the fractal dimension decreased (Figure 2). When the rectilinear model was assumed, it spent around 900 sec for the mean particle size to increase from 1 µm to 60 µm if all particles and aggregates were Euclidean objects with D=3.0. This time could be shortened to 320 sec for the aggregates with D=2.5 and to 120 sec for those with D=2.0. Under the same coagulation conditions with the curvilinear model applied, 9200 sec were needed for D=3.0, 2050 sec for D=2.5 and 600 sec for D=2.0. The prediction based on the curvilinear model is generally more sensitive to the magnitude of fractal dimension than that from the rectilinear model.

![Figure 2: Change of volume weighted mean size with time for different fractal dimensions using the rectilinear model and curvilinear model.](image)

Figure 2: Change of mass distribution (mass conc. in each section) with time using the rectilinear model and curvilinear model. D=3.0, α=0.1, G=15 /s, initial conc.=5x10^3 g/mL.
Shear Rate. The coagulation rate increased with the degree of fluid shear (Figure 3). During the very initial stage of rectilinear coagulation, the time required for the mean size to increase from 1 μm to 20 μm was 250 sec at a higher shear rate of G=50 /s in comparison with a time of 1650 sec at G=5 /s. With the curvilinear collision model, the time required to achieve the same extent of coagulation would be reduced from 6000 sec to 2300 sec as the shear rate increased from 5 /s to 50 /s. The significance of shear rate in coagulation of small particles was apparently greater for the rectilinear collision kernels than the curvilinear kernels as suggested by Han and Lawler (1992). For larger particles with the mean size beyond 20 μm, both the rectilinear and curvilinear models had the similar degree of sensitivity to the change in shear rate.

Comparison Between Numerical Simulation and Experimental Study

The prediction of simulation compared reasonably well with the experimental results in the evolution of particle size distributions (Figure 4). For a pulsed input, the change in size distribution obtained from model simulation and experimental measurement followed the similar trend of development. A fractal dimension of D=2.2 had to be assumed for the particle aggregates formed, suggesting the fractal nature of the aggregates. The similarity between the predicted and measured particle size distributions in a coagulation system indicated that the modelling approach established in this study can be applied to describe the dynamic process of particle coagulation and flocculation.

CONCLUSIONS

An improved sectional modelling technique, which includes a new curvilinear collision model and the fractal geometry of aggregates, has been developed to carry out numerical simulation for the complex coagulation process. The prediction of model simulation compared well with the results of the previous research and the experimental results. Different effects on the coagulation rate of different factors: fractal dimension D and shear rate G were demonstrated. These analyses are very useful to the real water supply and wastewater treatment works. In water supply and wastewater works, coagulation rate can be controlled through changing some operation conditions.
ACKNOWLEDGEMENT

This research was supported by HKU7327/98E from the Research Grants Council of Hong Kong SAR Government, China.

REFERENCES


Figure 4: Evolution of measured aggregate mass distribution vs. predicted by modelling simulation. D=2.2, G=15 /s.