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Fractal nature of flocs and compact floc formation

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Abstract: Random flocs formed under conventional operation condition are of fractal nature. There is a simple relation of $D_f = 3 - K_p$ between D_f, the fractal dimension and K_p, the floc density index in the floc density function $\rho_e \infty d_p^{Kp}$. Through a stepwise agglomeration model, the process of floc growth was discussed. It was shown that at each agglomeration step, additional void water was entrapped in the floc and thus affected floc density and structure. By analyzing the parameters of the agglomeration model, it was further proved that a floc formed in this manner is of fractal nature with its fractal dimension depending on the void ratio ϵ and agglomeration number m. A decrease in ϵ or increase in m may result in an increase in D_f, which implies a transition of floc from loose structure to compact. Mechanical syneresis and one-by-one attachment are two pathways for compact floc formation - the former can be realized by prolonged mechanical agitation and the later by the fluidized pellet bed operation. Experimental results show that spherical pellets of high density can be obtained by both methods. However, the pellets formed by mechanical syneresis still show a tendency of decrease in their density with increase in particle size, and the fractal dimension of such kind of pellets is about 2.40 - 2.47. In contrast, the pellets formed through one-by-one attachment show almost identical density regardless of the particle size, and the fractal dimension can be considered to be near 3. The optimum condition for fluidized pellet bed operation was also discussed and comparison was made on the density of compact flocs with that of random flocs formed by conventional operation. The fluidized pellet bed method is proved to be the most effective way to achieve floc compaction.

Key words: *morphology*; *fractal*; *floc density*; *syneresis*; *one-by-one attachment*; *fluidized pellet bed*

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1 Introduction

The relationship between floc structure and floc density has long been a subject attacting scientific concern in the field of coagulation and flocculation studies. In 1963 Vold Carried out the first significant work and proposed the 'ballistic aggregation model' in which the floc formation process was simulated by successive addition of primary particles to a growing aggregate in a random manner[1]. Sutherland made some comments upon this model, and pointed out that this model does not physically represent the manner in which real flocs form[2]. Later, he proposed a modified 'cluster aggregation model' by taking into account the process of collisions among clusters with various sizes to form larger aggregates[3]. In addition to Vold and Sutherland's theoretical work, Tambo, Lanvanker, Francois and other researchers conducted numerous experimental studies on the relation between floc density and floc size[4-6]. It is widely recognized that the effective density (buoyant density) of floc ρ_e

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decreases with floc diameter d_p in an inversely proportional relation as $\rho_e \propto d_p^{K_p}$, where Kp is the exponential coefficient. Because particle growth in a real flocculator is a process of random collision and aggregation, as a floc grows, the ratio of void space among the component primary particles increases. This results in a decrease of floc density. Yusa, Higashitani et al[7, 8] studied the method to alter such kind of property of the random flocs, and proposed a technique for pellet-like particle formation by using organic polymer flocculant and exerting mechanical agitation in concentrated suspension. After prolonged agitation under the polymer bridging, the void water is dispersed from the already formed random flocs and dense pellets form eventually. The mechanism of this process is described by Yusa as 'mechanical syneresis'.[7] Following a similar principle but different operation method, Tambo and Wang carried out a series of studies on compact floc formation by a fluidized pellet bed operation[9-13]. Under a well-controlled condition, primary particles are continuously introduced into the fluidized bed and attached onto the surface of the grown particles. This method is found to be successful in the treatment of high turbidity surface water such as that from the Yellow River in China. The mechanism of this process is described as 'one-by-one attachment'.[9] These studies have brought about improvement of the conventional flocculation process and showed the possible pathways to promote the formation of compact flocs with large size, high density, great settling velocity and low moisture contents.

In early 1980s Mandelbrot introduced the basic idea of fractal geometry[14], Which provided a completely new theoretical approach to the study on the structure of flocs. In a fractal geometrical system, the structure of an object can be characterized by its fractal dimension which, in the case of particle aggregation, indicates the degree of the occupation of the embedding space by the particles composting the aggregates. A number of studies demonstrated that the fractal dimension would be affected by shearing force and the coagulant dose during flocculation[15, 16]. However, there are still questions on how the fractal nature of flocs affects their density and other properties, and how to control operation condition to alter their fractal nature. Pelleting flocculation operation has shown its advantage over conventional operation in promoting compact floc formation, but theoretical study is still needed to bridge floc morphology with flocculation operation modes. In this paper, a theoretical analysis is first preformed to understand the relationship between floc morphology and floc density. Then pathways to alter the fractal nature of flocs and to realize compact floc formation are proposed. The characteristic density-size relations of flocs formed by different operation methods and with different suspensions are revealed on the basis of experimental results.

2 Fractal nature of flocs

2.1 Fractal dimension and floc density coefficient

According to the concept of fractal nature, the mass of an object M and its representative length L have the following relation

$$M \propto L^{D_f} \tag{1}$$

Apparently in the Euclidean geometric system, i.e. for non-fractal object, $D_f = 3$, but in the non-Euclidean system, i.e. for fractal object, $D_f < 3$, where D_f is called the fractal dimension.

Conventionally, we are used to the Euclidean geometric system and like to write Eqn 1 in the following way

$$M = \rho V = \alpha \rho L^3 \tag{2}$$

where ρ = density, V = volume ($V = \alpha L^3$), α = geometric factor (e.g. for spherical object, $\alpha = 4\pi/3$).

Considering a floc, its representative length is the floc diameter d_p, and its density follows the Tambo-Watanabe floc density function[4]

$$\rho = \beta d^{-K_p} \tag{3}$$

where, β = coefficient, K_p = floc density index.

Substituting Eqn 3 into Eqn 2, and considering $L = d_p$, we get

$$M = \alpha\beta d_p^{3-K_p} \propto d_p^{3-K_p} \tag{4}$$

Comparing Eqn 4 with Eqn 1, we get

$$D_f = 3 - K_p \tag{5}$$

Eqn 5 shows that as long as $K_p > 0$, $D_f < 3$ always holds. From the concept of fractal nature, we understand that an ordinary floc is a typical fractal object.

2.2 Evaluation of fractal dimension

The fractal dimension of a floc can be evaluated by measuring its projected area and perimeter as has been done by Aratani et al. who revealed that under ordinary flocculation condition the fractal dimension of floc has the value of $D_f = 1.2 - 1.8$ and D_f increases with the increase of agitation intensity[17]. The ballistic aggregation model proposed by Vold results in a model floc which consists of a central core and an outer region like a group of projection tentacles' as shown in Fig. 1[1]. Lanvankar et al. used this model and calculated the density-size relation of ferric hydroxide flocs as[5]

$$\rho \propto A_p^{-0.338} \propto d_p^{-0.676} \tag{6}$$

where A_p = projected area of the floc.

According to Eqn 3 and Eqn 5, the fractal dimension of the model floc can be calculated as D_f = 2.324. As is shown in Fig. 2, Sutherland's cluster aggregation model floc[3] is apparently looser in structure. The floc density index is evaluated as K_p = 0.9 - 1.0 and the fractal dimension is D_f = 2.0 - 2.1. Tambo and Watanabe's experiments on aluminum hydroxide flocs revealed a relationship as shown in Fig. 3 and Eqn 3. The value of the floc density index K_p was found to increase with ALT ratio (aluminum concentration dosed/suspended solid concentration). Under ordinary coagulation conditions, K_p = 1.0 - 1.4.

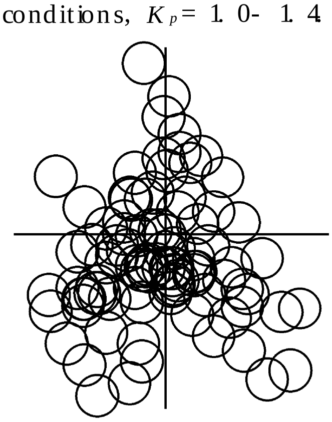

Fig. 1 Vold model floc

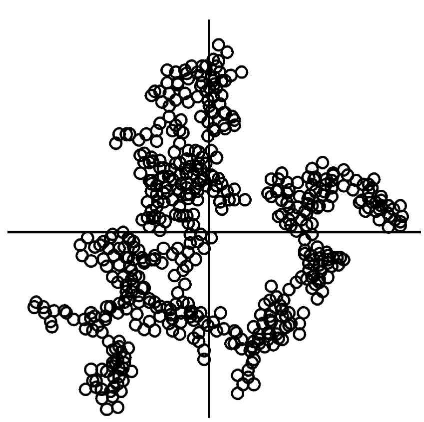

Fig. 2 Sutherland model floc

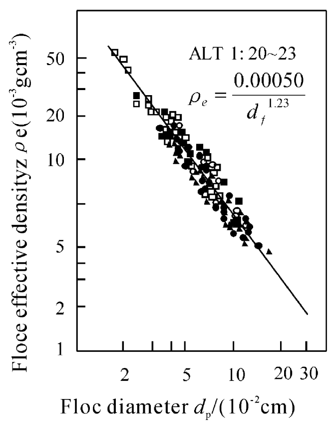

Fig. 3 Tambo floc density function

3 Simplified model of multilevel floc

3.1 Multilevel floc model

According to Sutherland's cluster aggregation model, the process of floc formation can be considered as a process in which primary particles agglomerate with each other to form small clusters, and then the small clusters agglomerate step by step to form larger ones. Supposing the first step of particle agglomeration is the combination of primary particles to form so-called first-level aggregates (Fig. 4), the effective density of the aggregate ρ_1 can be calculated by the effective density of the primary particle ρ_0 and the void ratio of the aggregate ϵ_1.

$$\rho_1 = \rho_0 (1 - \epsilon_1) \quad (7)$$

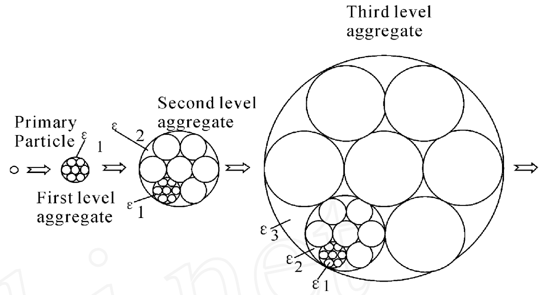

Fig. 4 Stepwise growth model of floc

Then if the second step of agglomeration is considered to be a combination of the first level aggregates to form so-called second-level aggregates, the effective density of the second-level aggregate ρ_2 can be calculated in a similar way:

$$\rho_2 = \rho_1 (1 - \rho_2) \quad (8)$$

where ϵ_2 is the ratio of void water among the first-level aggregates in a second-level aggregate. A combination of Eqns 8 and 7 yields

$$\rho_2 = \rho_0 (1 - \rho_1)(1 - \rho_2) \quad (9)$$

Further, if agglomeration progresses in a stepwise way to the nth level, the density of a nth level aggregate becomes

$$\rho_n = \rho_{n-1}(1 - \rho_n) = \rho_0 (1 - \rho_1)(1 - \rho_2) \cdots (1 - \rho_n) \quad (10)$$

where, the parameters with subscripts $n-1$ and n are those connecting with the $(n-1)$th and the nth steps of agglomeration, respectively.

Assuming that the void ratios at all these steps have the same value, i.e.

$$\epsilon_1 = \epsilon_2 = \cdots = \epsilon_n = \epsilon \quad (11)$$

then, Eqn 10 becomes

$$\rho_n = \rho_0 (1 - \epsilon)^n \quad (12)$$

At each of the abovementioned steps, if the agglomeration number of the lower level aggregates into a newly formed aggregate is m, then the relationship between the diameter of the nth aggregate d_n and that of the primary particle d_0 is derived as

$$d_n = d_0 [m/(1 - \epsilon)]^{n/3} \quad (13)$$

where n = agglomeration level.

Substituting Eqn 12 into Eqn 13 to eliminate the agglomeration level n and after rearrangement, we get

$$\rho_n = \rho_0 (d_0/d_n)^{-3\ln(1-\epsilon)/\ln[m/(1-\epsilon)]} \quad (14)$$

Denoting $\beta = \rho_0 d_0^{-3\ln(1-\epsilon)/\ln[m/(1-\epsilon)]}$, $K_p = -3\ln(1-\epsilon)/\ln[m/(1-\epsilon)]$, Eqn 14 can be rewritten as

$$\rho_n = \beta d_n^{-K_p} \quad (15)$$

Apparently Eqn 15 has the same form as Eqn 3. Because $1 - \epsilon < 1$, K_p is always positive. Therefore, the simplified multilevel model floc follows the same size-density relationship as ordinary flocs, and the floc density index K_p relates to the agglomeration number m and void ratio ϵ but irrelevant to the agglomeration level n.

3.2 Fractal dimension of the model floc

In the proposed multilevel model, because agglomeration proceeds in the same manner at each step and the resultant cluster at each step becomes an elementary particle for the next step, the model floc holds the property of self-similarity. Self-similarity is the basic property of a fractal object[14]. Therefore, the model floc is considered to be typically fractal. Following the relationship between fractal dimension D_f and floc density index K_p as shown in Eqn 5, the fractal dimension of the proposed multilevel model floc can be calculated as

$$D_f = 3 + 3\ln(1-\epsilon)/\ln[m/(1-\epsilon)] \qquad (16)$$

With any value of ϵ ($0 < \epsilon < 1$), $\ln(1-\epsilon) < 0$ and $\ln[m/(1-\epsilon)] > 0$. Therefore D_f is always less than 3. This accords with the concept of fractal nature.

In order to investigate how D_f varies with the two parameters m and ϵ, we took different values of m and drew the D_f - ϵ curves in Fig. 5 following Eqn 16. It is seen from Fig. 5 that there is a tendency of decrease in D_f value with increase in void ratio ϵ, and the smaller the m value, the more dominant this tendency is. In addition, D_f increases as the agglomeration number m increases. The limit void ratio is about 0.4 when spherical particles of identical diameter are piled up[18]. Therefore with a given m value, D_f reaches the maximum at $\epsilon = 0.4$. As shown in Fig. 5, even with a small m value (e.g. $m = 5$), the value of D_f is still as large as 2.24 at $\epsilon = 0.4$.

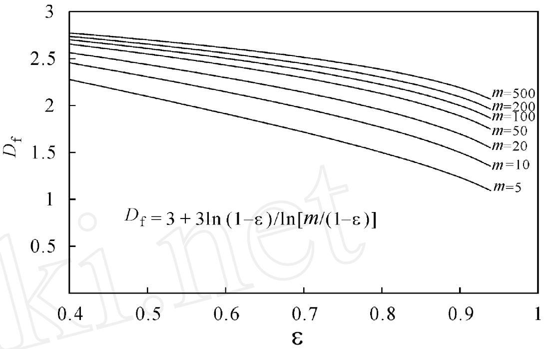

Fig. 5　Vaaiation of fractal dimension D_f with ϵ and m

From the above discussion on the simplified multilevel model floc, we understand that to decrease void ratio ϵ and/or increase the agglomeration number m at each step can result in an increase in the fractal dimension D_f. The increase of D_f value implies a transition of the flocs from loose structure to compact structure. Therefore, to decrease ϵ and to increase m are two possible pathways to make compact flocs. The former is called mechnical syneresis[7, 8], and the latter is called one-by-one attachment[9].

4　Experiment and results

4.1　Experiment of compact floc formation by mechanical syneresis

4.1.1　Methods of experiment

Concentrated suspension was prepared using kaolinite clay (ASP 170 manufactured by Engelhard Company, USA, mean size 0.55μm) to tap water. The clay concentration was adjusted to 3 g/L at pH = 7.0. A conventional jar test was conducted as following: (1) adding polyaluminum chloride (PAC) into the suspension and mixing rapidly at 300 rpm for 3 min; (2) at the end of rapid mixing, adding non-ionic polyacrylamide (Accofloc N - 100PWG, MW = 1.6×10) into the suspension and changing the mixing speed into 60 rpm to start prolonged agitation; (3) after given period of time, stopping agitation to let particles settle and then taking photographs using a telephoto lens for particle shape observation; (4) picking single particle gently using a pipette and then dropping it slowly into a quiescent settling tube for recording settling velocity and particle diameter using a single-frame, multiple flash technique; (5) after transforming the recorded data (single-fame, multiple-image photographs) through a digitizer to the computer, calculating the density of single particles one by one from the projected area diameter and terminal velocity using Stokes or Allen equation according to Reynolds' number of the particle. The dosages of PAC and polymer were controlled at ALT = 0.002 and PT = 0.001 (PT = polymer concentration dosed/suspended solid concentration). The agitation speed of 60 rpm at the second stage is higher than that for ordinary flocculation to meet the requirement of mechanical syneresis.

4.1.2　Experiment results

The aggregates were found to undergo compaction to become pellet-like. Until 30 min, only bulky random flocs formed but particles became denser and denser between 30- 60 min and gradually became pellet-like. At 120 min they were already spherical in appearance. Still longer agitation seemed to have no apparent effect. Fig. 6 is a comparison of the particle appearance at 30, 60 and 120 min.

The density of particles after each period of agitation was measured. Fig. 7 shows their density-size relations. During the prolonged agitation, particle density increases with time but shows no change after 120 min. This coincides with the observation result and indicates that prolonged agitation can bring about particle compaction. The effective density of a particle with about 0.1 cm in dianeter can reach a value larger than 0.1g/cm^3. On the other hand, in spite of the particle appearance, there still exists a linear relationship in the log log plot as $\rho_e \propto d_p^{-K_p}$. Calculating the density-size relation using the data of 120-360 min, the floc density index was evaluated as K_p = 0.53-0.60. Following

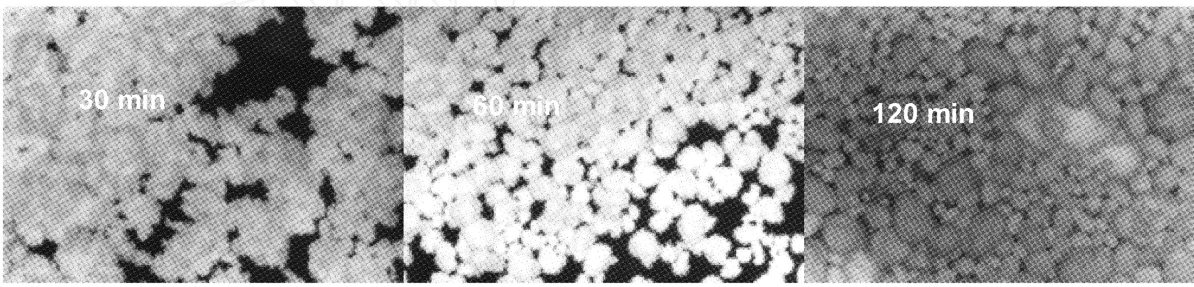

Fig. 6 Variation of the appearance of aggregates in jar-test

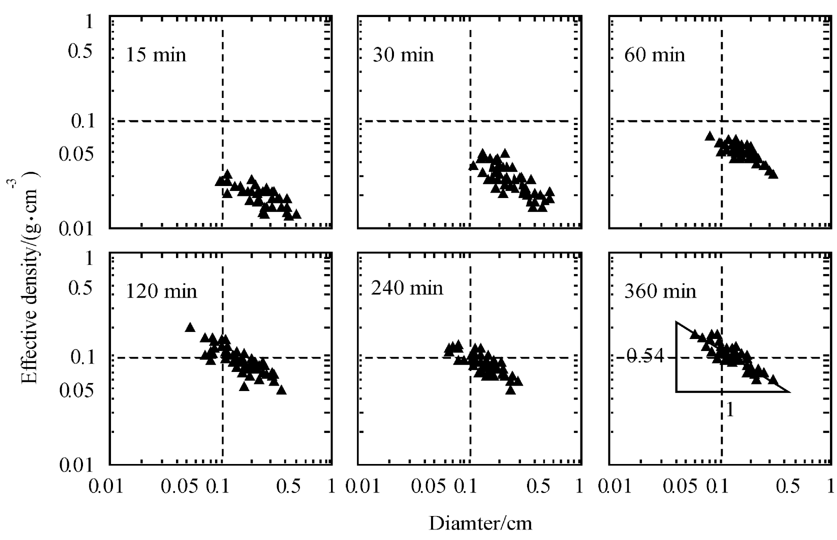

Fig. 7 ρ_e-d_p relation after various duration of agitation

the relation of D_f = 3- K_p, the fractal dimension of flocs formed under this operation condition can be calculated as D_f = 2.40-2.47 which is much higher than that of ordinary flocs and even higher than that derived from Vold's ballistic aggregation model.

4.2 Experiment of compact floc formation by one-by-one attachment

The one-by-one attachment mode is realized by fluidized pellet bed operation. Fig. 8 shows the flow chart of the experiment system for the treatment of high-concentration suspensions. Raw water was prepared using kaolinite clay in the same way as described above for the former experiment. PAC and polymer were also utilized. The operation proceeded in two stages. First, raw water was coagulated by adding PAC to the suspension in the flash mixer. The dosage of PAC was at a level just to bring about destabilization of the clay particles but not let them grow larger. After a detention period about 2 min in the flash mixer and pH adjusting, the suspension is led to the bottom of the fluidized pellet bed. At the bottom inlet of the fluidized bed, polymer flocculent was added to the destabilized suspension. The up-flow rate was controlled at 18 m/h. Moderate agitation was exerted in the fluidized bed at a G value about 30 s^{-1}. After a starting period, a fluidized grown pellet layer with very high concentration was formed and reached a steady state. Under this condition, as primary particles enter the bed, they dispersed quickly to the surroundings of the grown pellets and attached onto their surface. With the great difference between the concentrations of these two kinds of particles with much different sizes, the opportunity of contacts among primary particles is much less than that between primary particles and the grown pellets. With the great shearing force in the bed, random collision and agglomeration are restricted and one-by-one attachment becomes the dominant mode of particle agglomeration. As pellets grow, the fluidized layer is self-renewed continuously and the surplus particles overflow into the separation column.

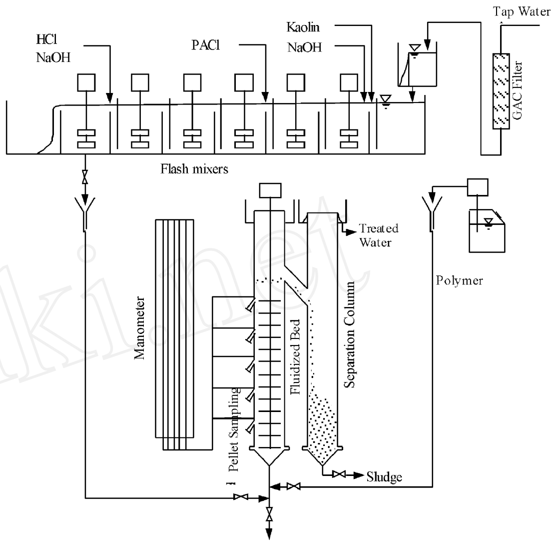

Fig. 8 Fluidized pellet bed separation system

Experiments were conducted with kaolinite clay suspensions of 0.3, 1.0, 3.0 and 10.0 g/L in turbidity at up-flow rate of 18 m/h. The optimum dosages of PAC and polymer for the treatment of these suspensions are ALT = 0.006, 0.003, 0.002, 0.001 and PT = 0.003, 0.0015, 0.001, 0.0005, respectively. Fig. 9 shows the appearance of pellets in the fluidized bed when raw water turbidity was 3.0 g/L. It is clear that particles formed through such kind of operation are dense and spherical. Fig. 10 shows the size-density relation of pellets. It was seen that (1) in each case the size of pellets were distributed in a comparatively narrow range, and as raw water turbidity increased, the effective density of pellets increased as well (the average ρ_e were calculated as 0.112, 0.139, 0.170, 0.292 g/cm^3 respectively in the 4 cases); (2) in each case the pellets held almost equally effective density regardless of their sizes, and the characteristic of random flocs, i.e. a linear decrease in effective density with particle size did not appear in the log-log plot at all; (3) comparing Fig. 10 with Fig. 7, although spherical and pellet-like particles have formed by both mechanical syneresis and one-by-one attachment, particles generated in the fluidized pellet

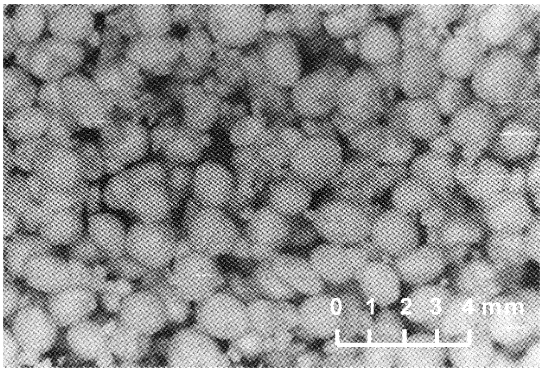

Fig. 9 Particles in the fluidized pelled bed

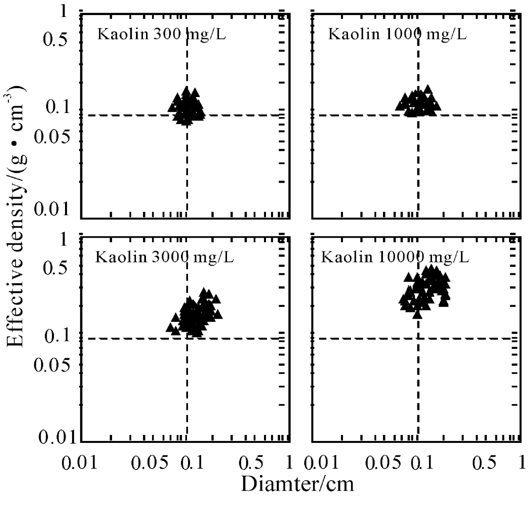

Fig. 10 ρ_e- d_p relation at various raw water turbidity bed show much larger density. The fractal dimension of the pellets thus formed cannot be evaluated from the ρ_e- d_p relation. By comparing the data shown in each graph of Fig. 10 more carefully, it is noticeable that larger particles seem to have larger density. The reason is still unknown but at least we can draw a conclusion that pellets formed in the fluidized bed are completely different from the conventional random flocs. The fractal dimension of such particles is thought to be very close to 3.0.

4.3 Control of the fluidized pellet bed process

Rational control of chemical dosing is the prerequisite condition for the fluidized pellet bed operation. Table 1 shows the results of experiment using 3.0 g/L kaolinite clay suspension with various dosages of PAC and polymer. Overdosing of PAC results in a decrease of pellet density or makes pelleting impossible, and in the contrary, overdosing of polymer brings about formation of large clumps or spoil fluidization condition. The optimum chemical dosages for 3.0 g/L suspension are recommended as ALT = 0.002 and PT = ALT/2.

Tab. 1 Condition of pelleting and results of treatment at different coagulant dosages

(raw water turbidity: 3.0 g/L)

ALT ratio	0.001	0.002	0.003	0.004	0.002			
PT ratio	0.001				0.0005	0.001	0.0015	0.002
Fluidized state	△	○	○	×	×	○	○	△
ρ_e of pellets/g·cm^{-3}	0.20	0.17	0.11	-	-	0.17	0.21	0.25
Moisture content of pellet sludge/%	73.5	75.6	81.6	-	-	75.6	75.0	72.9
Supernatant concentration/mg·L^{-1}	20.0	2.0	1.0	-	-	2.0	1.0	80.0

○: well pelleting; △: formation of large clumps; ×: impossible for pelleting

When metal salts such as alum or PAC are used as coagulant, zeta potential is a very useful parameter for determining the optimum dosage[10]. Clay particles usually has a zeta potential of -25 to -30 mV. For conventional coagulation operation, a zeta potential of -10mV is often taken as the minimum requisite degree of destabilization. For batch coagulation operation such as jartest in a laboratory, the minimum degree of destabilization is lower and a zeta potential of -13 to -15mV is thought to be optimum. For the fluidized pellet bed operation, the optimum dosage of PAC is much lower than the former two cases. The corresponding zeta potential is about -20mV. Fig. 11 shows the relation between optimum ALT and raw water turbidity with zeta potential ζ as referential parameter. As raw water turbidity increases, the required specific dosage, i.e. ALT ratio decreases.

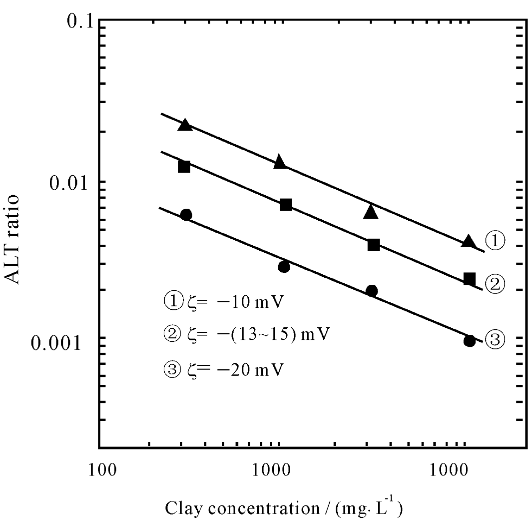

Fig. 11 Relation of raw water turbidity, ALT ratio and ζ potential

5 Discussion

According to the proposed multilevel floc model, floc density can be expressed as

$$\rho_e = \rho_0 (1-\rho_1)(1-\rho_2)\cdots(1-\rho_n) \tag{17}$$

or

$$\rho_e = \rho_0 (1-\rho)^n \quad (18)$$

if the void ratio introduced to the growing floc is the same at each agglomeration step.

From Eqn 17 or Eqn 18, we understand that the introduction of new void ratio to the floc at each step is the reason for the decrease of floc density with increasing floc size. By mechanical syneresis, higher level void water can be dispersed from a grown random floc and restructuring of primary particles within the floc can proceed to certain extent. As a result, the density and fractal dimension of the floc can be increased. However, from the experiment results mentioned in the previous sections, we noticed that there is still a tendency of decrease in floc density with increase in floc size. The reason for this may be attributed to incomplete restructuring. There may still exists higher level void water within the floc. Therefore, the fractal dimension D_f, although as high as 2.40 to 2.47, is still less than 3.

In the up-flow fluidized pellet bed operation, because particle growth proceeds through one-by-one attachment of primary particles onto the surface of a grown particle, almost no higher level void water is introduced into the particle during its growth, hence the floc density can be expressed as

$$\rho_e = \rho_0 (1-\rho) \quad (19)$$

Under such circumstances, floc density only depends on the density of primary particle ρ_0 and void ratio among primary particles ϵ. This makes it possible for particles to become identical in density regardless particle size. By analyzing size distribution and density of primary particles under different raw water turbidity and PAC dosage, and comparing with the density of pellets formed in the fluidized bed, the void ratio of the pellets was evaluated as 0.47 to 0.58[9]. The limit void ratio for piling identical spherical particles is about 0.4[18]. With irregular shaped particles, the limit void ratio should be greater. Therefore, for the pellets generated, $\epsilon = 0.47 - 0.58$ is thought to be the first level void ratio but not include the influence of higher level void water. In this case, the fractal dimension is close to 3. The $\rho_e - d_p$ relation shown in Fig. 10 provides evidence to this assumption.

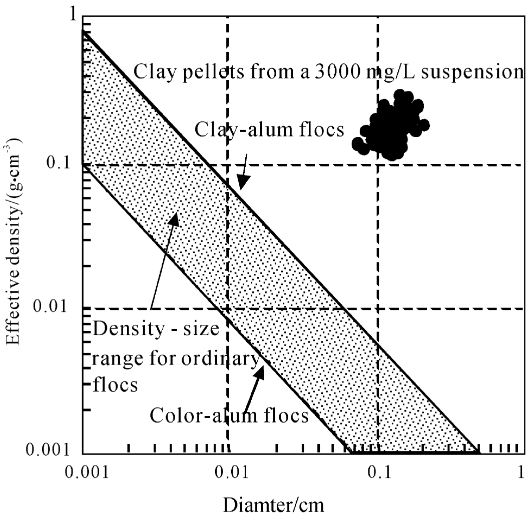

Fig. 12 Density-size relation of pellets in comparison with conventional flocw

Fig. 12 summarizes the density-size relation of clay pellets (3.0 g/L clay suspension) formed by the fluidized bed operation in comparison with that of ordinary flocs. The advantages of fluidized pellet bed operation over conventional flocculation is very clear——much larger particle size and higher density, and above all, remarkable change of the fractal nature of flocs.

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絮凝体的分形特征和致密型絮凝体形成

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摘 要: 传统混凝条件下形成的随机型絮凝体具有分形特征, 其分形维数D_f与其密度函数$\rho_S \propto d_p^{-K_p}$的指数$K_p$之间具有$D_f = 3 - K_p$的关系. 通过建立分步成长絮凝体模型, 讨论了在絮凝过程中逐次导入颗粒间的空隙率对絮凝体密度和构造的影响. 模型参数分析的结果进一步证明了分步成长的絮凝体是一个典型的分形, 其分形维数取决于空隙率ϵ和颗粒结合个数m. 降低ϵ或提高m均有利于提高D_f, 使絮凝体由松散型向致密型过渡. 脱水收缩和逐一附着模式是达到这一目的的两种操作模式, 前者可以通过延长机械搅拌时间来实现, 而后者通过造粒流化床实现. 实验结果表明两种方式均能提高形成的球状颗粒的密度, 但是前者所形成的团粒依然具有颗粒密度随粒径增大而降低的特点, 其分形维数为2.40～2.47; 而逐一附着模式所形成的团粒密度基本上与粒径无关, 其分形维数接近于3. 通过讨论造粒流化床操作条件, 并将试验得到的致密型絮凝体密度和常规絮凝体密度进行比较, 说明该方法实现逐一附着型絮凝体操作是促使絮凝体致密化的有效途径.

关键词: 形态学; 分形; 絮凝体密度; 脱水收缩; 逐一附着; 造粒流化床

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