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Author(s)	Tambo, Norihito; Takakuwa, Tetsuo; Funamizu, Naoyuki
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A Mathematical Model of Turbulent Flocculation Process

Norihito TAMBO* Tetsuo TAKAKUWA* Naoyuki FUNAMIZU*

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Abstract

First, a dimensionless rate equation including a floc disintegration term was developed to describe the entire process of turbulent flocculation. Next, in order to reduce the number of variables, a binary grouping method of particle size distribution was proposed. By these methods, it became possible to compare an experimental result of flocculation process with a numerical solution. As a result it was found that the floc concentration, and the intensity and period of slow agitation had an effect on both of the growth rate and the disintegration rate of flocs.

1. Introduction

The time variation of floc size distribution in the flocculation process is described by simultaneous differential equations of s variables. Here s denotes the number of primary particles included in the maximum size floc. The value of s usually reaches a high order of 10^3 to 10^6 . Thus, it is very difficult to solve these equations even when a high speed computer is used. A solution to the problem is to reduce the number of variables by introducing a grouping of flocs. The authors propose a binary grouping method of floc size distribution. By the method, for example, a 10^6 -fold particle is classified as a floc of Group 20 because 10^6 is nearly equal to 2^{20} in the binary system. Thus, simultaneous differential equations of 10^6 variables can be simplified to those of 20 variables. The purpose of this paper is to show the details of grouping method with a satisfactory material balance and to depict the time variation of floc size distribution.

2. Formulation of Turbulent Flocculation Model

A floc is an aggregate of a number of primary particles. A floc containing i primary particles ($i=1, 2, \dots, s$) is defined as an i -fold floc. For turbulent flocculation controlled by a viscous subrange transport rate, the number of collision-agglomeration between i and j -fold flocs per unit time and in a unit volume, RC_{ij} , is written as follows^v.

$$RC_{ij} = 12\pi\alpha\beta\sqrt{\frac{\epsilon_0}{\mu}}\left(\frac{d_i}{2} + \frac{d_j}{2}\right)^3 n_i n_j \quad (1)$$

where π is the circular constant, α a collision-agglomeration coefficient, β a constant,

*) Department of Sanitary Engineering, Faculty of Engineering, Hokkaido University.

ε_0 the effective energy dissipation rate, μ the viscosity of water, d the floc diameter, n the number of flocs in a unit volume, and the subscripts i and j denote an i - and a j -fold floc, respectively. Based on Eq. (1), the growth rate of i -fold floc, dn_i/dt , is described as Eq. (2) for a flocculating system where the maximum floc is s -fold²⁾.

$$\frac{dn_i}{dt} = \frac{3}{2} \pi \alpha \beta \sqrt{\frac{\varepsilon_0}{\mu}} \left\{ \frac{1}{2} \sum_{j=1}^{i-1} (d_j + d_{i-j})^3 n_j n_{i-j} - n_i \sum_{j=1}^{s-i} (d_i + d_j)^3 n_j \right\} \quad (2)$$

The density of a floc decreases with size growth because of embracing the interstitial water. The relationship between the effective density ρ_e and the diameter d is observed³⁾ to be

$$\rho_e = a \cdot d^{-k} \quad (3)$$

where a and k are constants dependent on floc characteristics. From Eq. (3) and the definition of i -fold floc, the diameter of i -fold floc, d_i , can be written as

$$d_i = d_1 \cdot (i \cdot \rho_{e,1} / \rho_{e,i})^{1/3} = d_1 \cdot i^{1/(3-k)} = d_1 \cdot i^f \quad (4)$$

where $\rho_{e,1}$ and d_1 are the effective density and diameter of a primary particle, respectively and $f=1/(3-k)$. Substitution of Eq. (4) into Eq. (2) gives

$$\frac{dn_i}{dt} = \frac{3}{2} \pi \alpha \beta d_1^3 \sqrt{\frac{\varepsilon_0}{\mu}} \left[\frac{1}{2} \sum_{j=1}^{i-1} \{j^f + (i-j)^f\}^3 n_i n_{i-j} - n_i \sum_{j=1}^{s-i} (i^f + j^f)^3 n_j \right] \quad (5)$$

At a final stage of the flocculation process, floc size distribution reaches a definite equilibrium state. This kind of self-preserving final distribution is realized when a disintegration term is introduced. If it is assumed that floc disintegration is brought about by the dynamic pressure difference $\Delta p \propto (\varepsilon_0/\mu)d^2$ of turbulent eddies in the viscous subrange, the number of disintegration of i -fold floc per unit time and in a unit volume, RD_i , may be expressed as follows.

$$RD_i = \frac{c\varepsilon_0}{\mu} \left(\frac{d_i^2 - d_1^2}{d_s^2 - d_1^2} \right) = \frac{c\varepsilon_0}{\mu} \left(\frac{i^{2f} - 1}{s^{2f} - 1} \right) = \frac{c\varepsilon_0}{\mu h} (i^{2f} - 1) \quad (6)$$

where c is a coefficient relating to floc disintegration and $h=s^{2f}-1$. In addition to the above assumption, if it is assumed that a floc particle is disintegrated into two same size flocs, the basic flocculation equation is written in a dimensionless form as follows.

$$\begin{aligned} \frac{dN_i}{dm} = & \frac{1}{2} \sum_{j=1}^{i-1} \{j^f + (i-j)^f\}^3 N_i N_{i-j} - N_i \sum_{j=1}^{s-i} (i^f + j^f)^3 N_j \\ & + \frac{cG'}{bn_0 d_1^3 h} \left[2 \{ (2i)^{2f} - 1 \} N_{2i} + \{ (2i-1)^{2f} - 1 \} N_{2i-1} \right. \\ & \left. + \{ (2i+1)^{2f} - 1 \} N_{2i+1} - (i^{2f} - 1) N_i \right] \quad (i=1, 2, \dots, s) \end{aligned} \quad (7)$$

where $m=bG'd_1^3 n_0 t$, $b=(3/2)\pi\alpha\beta$, $G'=\sqrt{\varepsilon_0/\mu}$, $N_i=n_i/n_0$ and n_0 is the total number of primary particles existing in the system. In Eq. (7), the first two terms on the right hand side are the variations due to collision-agglomeration and the third term is the variation due to disintegration.

3. Binary Grouping of Floc Size Distribution

By binary notation, 2^{K-1} to (2^K-1) -fold particles are classified as Group K . Thus, for a flocculation system where the maximum agglomeration number s is less than 2^s , the binary notation is described by S groups of floc as shown in Table 1. When the binary notation is used, the collision-agglomeration terms of Eq. (7) are represented by the following three types of reaction.

TABLE 1. Composition of Floc Groups

Group K	Particles consisting of Group K (i -fold particles)	Number of particles consisting of Group K	Number of primary particles in Group K (L_K)
1	1	1	1
2	2, 3	2	2.5
3	4, 5, 6, 7	4	5.5
4	8, 9, 10, 11, ..., 15	8	11.5
5	16, 17, 18, ..., 31	16	23.5
$\vdots$	$\vdots$	$\vdots$	$\vdots$
$S-1$	$2^{S-2}, 2^{S-2}+1, \dots, 2^{S-1}-1$	2^{S-2}	$(2^{S-1}+2^{S-2}-1)/2$
S	$2^{S-1}, 2^{S-1}+1, \dots, s$	$s+1-2^{S-1}$	$(2^{S-1}+s)/2$

$$(1) \quad (\text{Group } I) \times (\text{Group } K) \rightarrow [\text{Group } K] + [\text{Group } K+1] \quad (K=2, 3, \dots, S-1; I=1, 2, \dots, K-1)$$

$$(2) \quad (\text{Group } I) \times (\text{Group } K) \rightarrow [\text{Group } K] \quad (K=S; I=1, 2, \dots, S-1)$$

$$(3) \quad (\text{Group } K) \times (\text{Group } K) \rightarrow [\text{Group } K+1] \quad (K=1, 2, \dots, S-1)$$

For the disintegration term, the reaction is written as follows.

$$(4) \quad (\text{Group } K) \rightarrow [\text{Group } K-1] + [\text{Group } K] \quad (K=2, 3, \dots, S)$$

Thus, the rate of floc number variation in Group K can be shown as follows.

$$\begin{aligned} \frac{dN_K}{dm} = & \sum_{I=1}^{K-1} A_{I,K}^{(1)} N_I N_K + \sum_{I=1}^{K-2} B_{I,K-1}^{(1)} N_I N_{K-1} - \sum_{I=1}^{K-1} B Y_{I,K}^{(1)} N_I N_K - \sum_{I=K+1}^{S-1} C Y_{I,K}^{(1)} N_I N_K \\ & (K=2 \sim S-1) \quad (K=3 \sim S) \quad (K=2 \sim S-1) \quad (K=1 \sim S-2) \\ & - \sum_{I=K+1}^{S-1} D Y_{I,K}^{(1)} N_I N_K + \sum_{I=1}^{K-1} A_{I,K}^{(2)} N_I N_K - B Y_{S,K}^{(2)} N_S N_K + A_{K-1,K-1}^{(3)} N_{K-1} N_{K-1} \\ & (K=1 \sim S-2) \quad (K=S) \quad (K=1 \sim S-1) \quad (K=2 \sim S) \\ & - A Y_{K,K}^{(3)} N_K N_K + D_{K+1}^{(4)} N_{K+1} + E_K^{(4)} N_K \\ & (K=1 \sim S-1) (K=1 \sim S-1) (K=2 \sim S) \end{aligned} \quad (8)$$

Here, the superscripts (1) to (4) correspond to the reactions presented above. While the general forms of coefficients in Eq. (8) are detailed in the appendix, an example with respect to the reactions (1) and (4) will be shown in the following.

If we assume $I=2$ and $K=3$, and a uniform size distribution in each group, the reaction (1) is described as in Table 2. Among flocs of Group $I=2$, $(2 \times 2 + 3)/8/2.5$ of them is transferred by the reaction to Group $K=3$ and the remaining

TABLE 2. Collision-agglomeration between Groups 2 and 3

	4	5	6	7	(Group K)
(Group I)	2	6	7	8	9
	3	7	8	9	10

TABLE 3. Disintegration of Group 3

4	5	6	7	(Group K)
2	2	3	3	
2	3	3	4	[Group K-1]+[Group K]

$(2 \times 2 + 3 \times 3)/8/2.5$ is transferred to Group $K+1=4$. The same is seen for Group $K=3$ as $13/8/5.5$ and $31/8/5.5$ are transferred to Group $K=3$ and $K+1=4$, respectively. The representative collision diameter is the sum of $7/3$ and $13/3$ -fold particle diameters. At the same time, by these reactions Groups $K=3$ and $K+1=4$ increase their contents by $20/8/5.5$ and $44/8/11.5$, respectively. The sum of $13/5$ and $31/5$ -fold particle diameters is the representative collision diameter in this case. On the other hand, the reaction (4) for Group $K=3$ is exemplified as in Table 3. When one floc is disintegrated into two particles, the number of Group $K-1=2$ and $K=3$ are increased by $(2 \times 3 + 3 \times 4)/4/2.5$ and $4/4/5.5$, respectively.

Thus, if the functional forms as $F(p, q) = (p^f + q^f)^3$ and $H(u) = (cG'/bn_0d_1^3h) \cdot (u^{2f} - 1)$ are used for the collision-agglomeration term and the disintegration term of Eq. (7), respectively, then a set of rate equations for the two reactions exemplified above is written as follows.

$$\begin{aligned}
 \frac{dN_2}{dm} &= -\frac{7}{8 \times 2.5} F\left(\frac{7}{3}, \frac{13}{3}\right) N_2 N_3 - \frac{13}{8 \times 2.5} F\left(\frac{13}{5}, \frac{31}{5}\right) N_2 N_3 \\
 &\quad + \frac{9}{2 \times 2.5} H(5.5) \\
 \frac{dN_3}{dm} &= \left(\frac{20}{8 \times 5.5} - \frac{13}{8 \times 5.5}\right) F\left(\frac{7}{3}, \frac{13}{3}\right) N_2 N_3 - \frac{31}{8 \times 5.5} F\left(\frac{13}{5}, \frac{31}{5}\right) \\
 &\quad + \left(\frac{1}{5.5} - 1\right) H(5.5) \\
 \frac{dN_4}{dm} &= \frac{44}{8 \times 11.5} F\left(\frac{13}{5}, \frac{31}{5}\right) N_2 N_3
 \end{aligned}$$

The summation of these equations shows that the material balance, i. e.,

$$\sum_{K=1}^s L_K N_K = 1 \quad \text{or} \quad \sum_{K=1}^s L_K \frac{dN_K}{dm} = 0 \quad (9)$$

holds. Here, L_K is the number of primary particles included in the floc of Group K .

4. Computation and Experiment

Two constants b and c which are implicitly included in Eq. (8), are evaluated by comparing simulation results obtained under steady and unsteady conditions with experimental measurements.

Under the steady condition, the relation as

$$dN_K/dm = 0 \quad (K=1, 2, \dots, S) \quad (10)$$

should hold. Thus, the number of flocs in Group K can be calculated from the simultaneous quadratic equations which are obtained by setting the left hand side of Eq. (8) to be zero. These equations can be solved for various values of c/b by using the Newton linearizing method. Here, it should be noted that one of these equations is not independent from the others and must be replaced by Eq. (9). As the value of c/b , the solution of the minimum error relative to the steady state experimental result is used.

When the value of c/b is substituted into Eq. (8), this equation is solved by use of the Runge-Kutta-Gill method. The computational result and experimental result will coincide when dimensionless flocculation time, m , is expanded by an appropriate factor of b . Thus, the value of b is obtainable from the relation as $b = m/(G'd_1^3n_0t)$ where the value of G' , d_1 and n_0 are known. In the next step, the value of c is obtained as $c = (c/b) \times b$.

A batch flocculation experiment by using kaolinite-aluminum floc was carried out under the conditions shown in Table 4. The floc diameters were measured at various time intervals by close-up photography. The minimum diameter was defined as 2.87×10^{-3} cm in this experiment. Two constants a and k with respect to floc density were measured from the settling test and were 2.83×10^{-4} and 1.3, respectively.

TABLE 4. Summary of Experimental Conditions and Computational Results

Run	Kaolinite dosage (mg/l)	Period of rapid agitation (min)	G' (sec ⁻¹)	n_0 (10 ³ cm ⁻³)	d_s (cm)	s	c/b	b	c
1	40	1	2.6	3.50	0.155	881	0.0638	25.2	1.61
2	40	1	3.8	3.50	0.125	611	0.0293	17.5	0.513
3	40	5	2.6	3.50	0.160	930	0.0592	19.6	1.16
4	40	5	3.8	3.50	0.110	492	0.0270	13.1	0.355
5	60	1	2.6	5.25	0.175	1083	0.0841	4.10	0.344
6	60	1	3.8	5.25	0.150	834	0.0335	3.86	0.129
7	60	5	2.6	5.25	0.135	697	0.0654	5.57	0.364
8	60	5	3.8	5.25	0.105	455	0.0321	3.29	0.106

* Aluminum to turbidity ratio is 0.04.

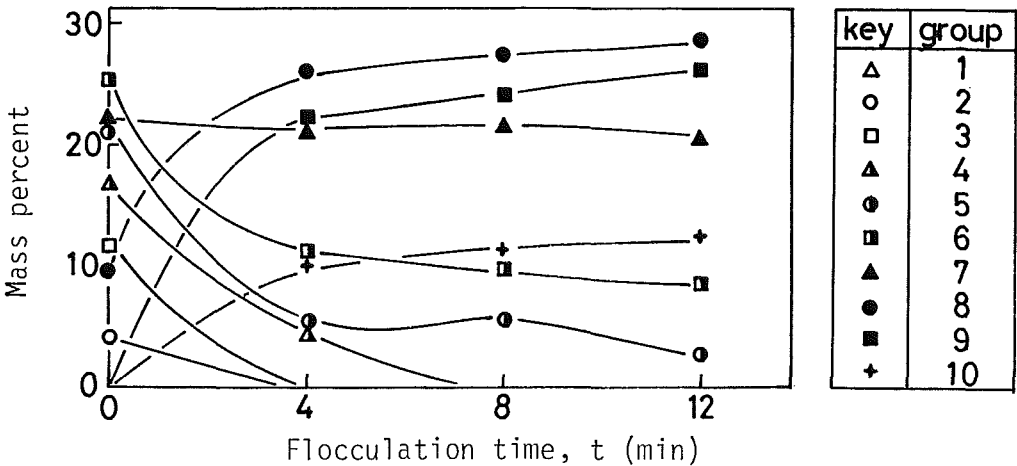


Fig. 1. Time variation of mass percent of flocs (Experimental run 3)

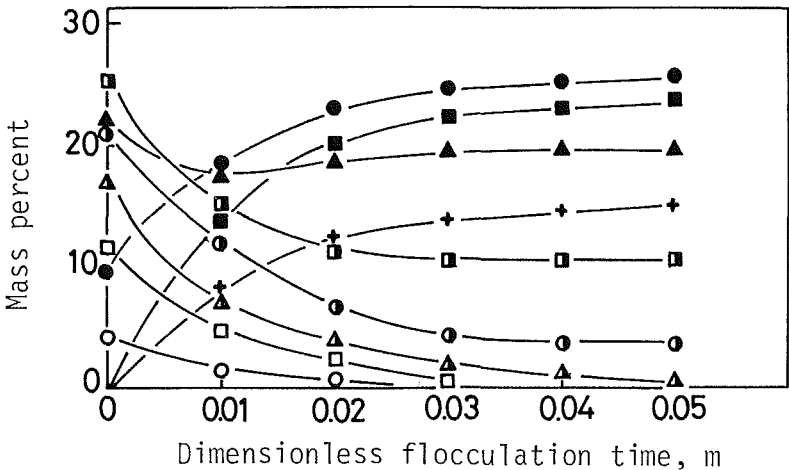


Fig. 2. Computational result (Run 3)

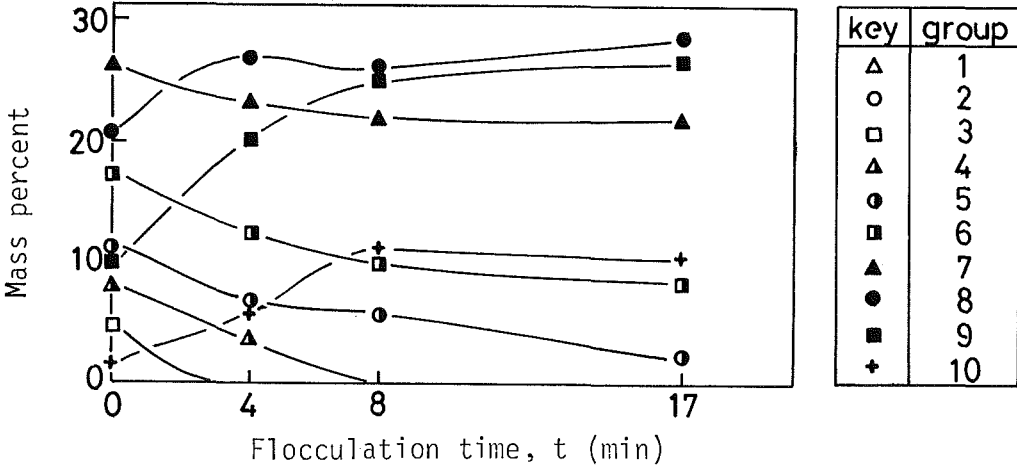


Fig. 3. Time variation of mass percent of flocs (Experimental run 6)

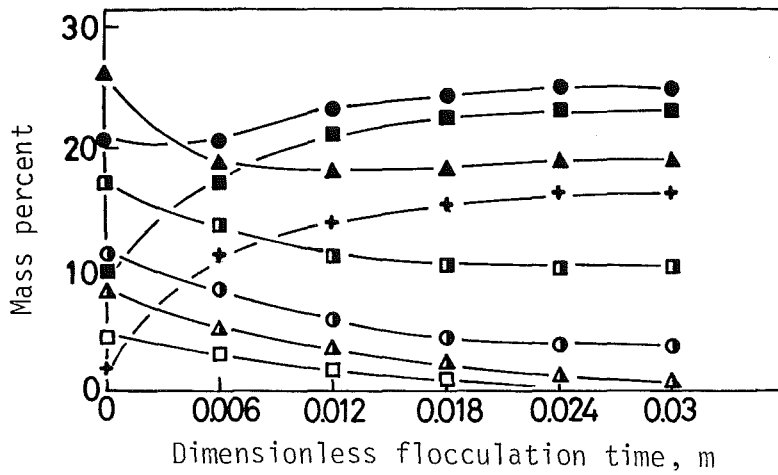


Fig. 4. Computational result (Run 6)

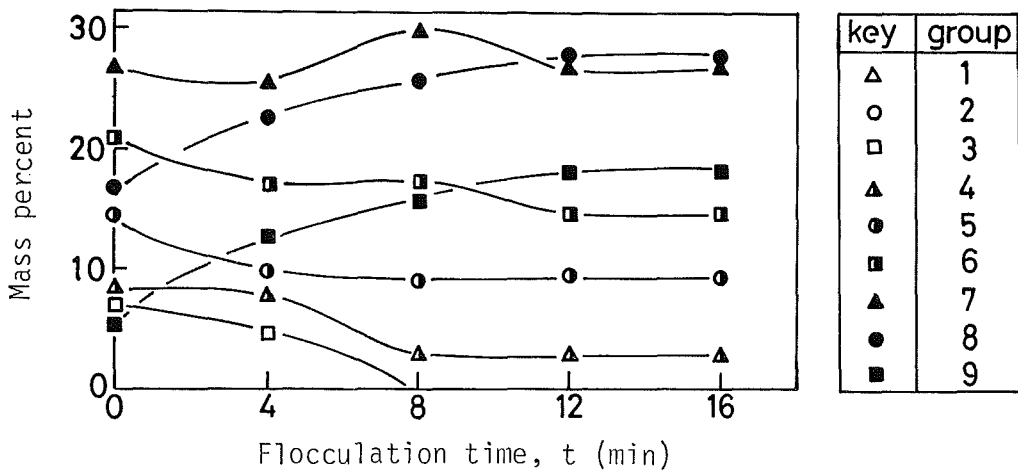


Fig. 5. Time variation of mass percent of flocs (Experimental run 8)

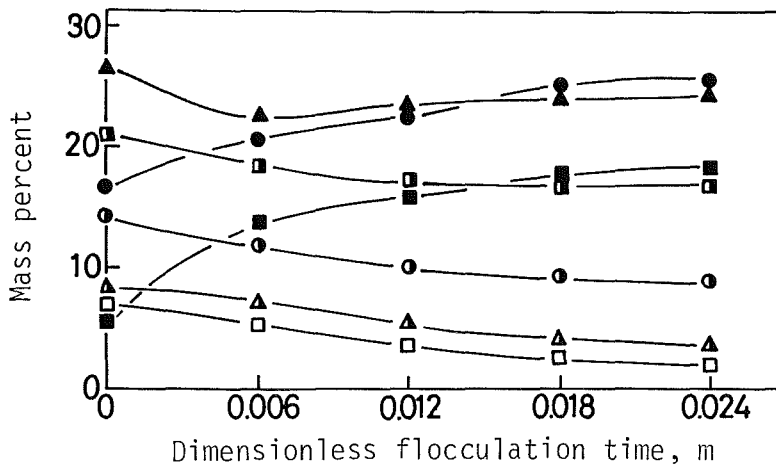


Fig. 6. Computational result (Run 8)

The results of three experimental runs are shown in Figs. 1, 3 and 5. The computational results for the three cases are depicted in Figs. 2, 4 and 6, respectively. It seems from these figures that the computational results are in close agreement with the experimental results, relative to the problems involved in conducting these experiments. The values of b and c were obtained for eight runs as in Table 4. These data show that the higher the kaolinite concentration is, the more intensive the slow agitation becomes and likewise the longer the period of rapid agitation is, the larger the values of b and c are.

5. Conclusion

In the past, the entire process of turbulent flocculation could not be analyzed numerically due to the difficulty of solving a large number of differential equations. The difficulty was dissolved by introducing the binary grouping method of floc size distribution.

The main results of the present study may be summarized as follows.

- 1) The final steady state of floc number distribution can be attained by introducing a disintegration term of flocs to the conventional rate equation of collision-agglomeration flocculation.
- 2) The binary grouping method has been precisely shown with a satisfactory material balance.
- 3) The turbulent flocculation process is assumed to be characterized by the two coefficients b and c . The former has an effect on floc growth rate and the latter on floc disintegration rate.
- 4) Two coefficients mentioned above are independently obtainable by running a comparison of a computational solution with the experimental result under a steady and unsteady state.
- 5) The experimental and computational results are in a fairly good agreement and show that the values of b and c are affected by the floc concentration, the intensity of slow agitation and the period of rapid agitation.

Acknowledgement

The authors are grateful to Mr. E. Matsuo for his assistance in the experiment. The calculations were conducted by the FACOM 230-75 computer at the Hokkaido University Computing Center.

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Appendix: General Form of the Coefficients in Eq. (8)

(I) If the coefficients $A_{I,K}^{(1)}$ to $A_{K-1,K-1}^{(3)}$ in Eq. (8) are denoted together as

$$\frac{T}{z \cdot L_K} \left\{ \left(\frac{x}{r} \right)^I + \left(\frac{y}{r} \right)^I \right\}^3$$

where L_K is obtainable from Table 1, each value is calculated as follows.

(1) For $A_{I,K}^{(1)}$ ($K=2, 3, \dots, S-1$; $I=1, 2, \dots, K-1$):

a) in the case where $2^K \leq s - 2^I + 1$

$$z = 2^{K+I-2} \equiv z_1, \quad T = x = 2^{I-2} (9 \cdot 2^{K+I-2} - 3 \cdot 2^{K-1} - 7 \cdot 2^{2I-1} + 9 \cdot 2^{I-1} - 1) / 3 \equiv T_1, \\ y = 2^{I-2} (9 \cdot 2^{2K-2} - 9 \cdot 2^{K+I-1} + 3 \cdot 2^{K-1} + 7 \cdot 2^{2I-2} - 1) / 3 \equiv y_1, \quad r = 2^{I-2} (2^K - 3 \cdot 2^{I-1} + 1) \equiv r_1.$$

b) in the case where $s - 2^{I-1} \geq 2^K \geq s - 2^I + 2$

$$z = 2^{I-2} (2^K - 2^{I-1} + 1) + (s - 2^K - 2^{I-1} + 1) (3 \cdot 2^{I-1} + 2^K - s - 2) / 2 \equiv z_2, \quad T = x = T_1, \quad y = y_1, \\ r = r_1.$$

c) in the case where $2^K \geq s - 2^{I-1} + 1$

$$z = 2^{I-2} (2s - 2^K - 3 \cdot 2^{I-1} + 3) \equiv z_3, \quad T = x = T_1, \quad y = y_1, \quad r = r_1.$$

(2) For $B_{I,K-1}^{(1)}$ ($K=3, 4, \dots, S$; $I=1, 2, \dots, K-2$):

a) in the case where $2^{K-1} \leq s - 2^I + 1$

$$z = 2^{K+I-3}, \quad T = 2^{I-1} (9 \cdot 2^{K+I-3} - 3 \cdot 2^{K-2} + 7 \cdot 2^{2I-3} - 9 \cdot 2^{I-2} + 1) / 3, \\ x = 2^{I-2} (7 \cdot 2^{2I-1} - 9 \cdot 2^{I-1} + 1) / 3, \quad y = 2^{I-2} (9 \cdot 2^{K+I-2} - 3 \cdot 2^{K-1} - 7 \cdot 2^{2I-2} + 1) / 3, \\ r = 2^{I-2} (3 \cdot 2^{I-1} - 1).$$

b) in the case where $s - 2^{I-1} \geq 2^{K-1} \geq s - 2^I + 2$

$$z = 2^{I-2} (2^{K-1} - 2^{I-1} + 1) + P_1 (2^{K-1} + 3 \cdot 2^{I-1} - s - 2) / 2, \\ T = 2^{2I-3} (2^K + 2^{I-1} - 1) + P_1 \{ 3 \cdot 2^{I-1} (2^{K-1} + 2^{I-1} - 1) - (P_1 + 1) (2^{K-2} - 2^{I-1} + s) \} / 3, \\ x = 2^{I-1} (7 \cdot 2^{2I-1} - 9 \cdot 2^{I-1} + 1) / 3 - (2^{K-1} + 2^I - s - 2) (2^{K-1} + 2^I - s - 1) (s - 2^{K-1} + 2^{I+1}) / 6, \\ y = 2^{I-2} (9 \cdot 2^{K+I-2} - 3 \cdot 2^{K-1} - 7 \cdot 2^{2I-2} + 1) / 3 \\ - (2^{K-1} + 2^I - s - 2) (2^{K-1} + 2^I - s - 1) (2^K - 2^I + s) / 6, \\ r = 2^{2I-2} + P_1 (3 \cdot 2^{I-1} + 2^{K-1} - s - 2) / 2, \quad \text{where } P_1 = s - 2^{K-1} - 2^{I-1} + 1.$$

c) in the case where $2^{K-1} \geq s - 2^{I-1} + 1$

$$z = 2^{I-2} (2s - 2^{K-1} - 3 \cdot 2^{I-1} + 3), \quad T = P_2 (s + 2^{K-1}), \quad x = P_2 (3 \cdot 2^{I-1} - 1), \\ y = P_2 (2^{K-1} - 3 \cdot 2^{I-1} + s + 1), \quad r = 2P_2, \quad \text{where } P_2 = 2^{I-2} (s - 2^{K-1} + 1).$$

(3) For $BY_{I,K}^{(1)}$ ($K=2, 3, \dots, S-1$; $I=1, 2, \dots, K-1$):

a) in the case where $2^K \leq s - 2^I + 1$

$$z = z_1, \quad T = y = 2^{I-2} (9 \cdot 2^{K+I} - 3 \cdot 2^K - 7 \cdot 2^{2I-2} + 1) / 3, \\ x = 2^{I-2} (7 \cdot 2^{2I-1} - 9 \cdot 2^{I-1} + 1) / 3, \quad r = 2^{I-2} (3 \cdot 2^{I-1} - 1).$$

b) in the case where $s - 2^{I-1} \geq 2^K \geq s - 2^I + 2$

$$z = z_2, \quad T = y = 2^{I-2} (9 \cdot 2^{K+I-1} - 3 \cdot 2^K - 7 \cdot 2^{2I-2} + 1) / 3 - P_3 (2^{K+1} - 2^I + s) / 6, \\ x = 2^{I-2} (7 \cdot 2^{2I-1} - 9 \cdot 2^{I-1} + 1) / 3 - P_3 (s - 2^K + 2^{I+1}) / 6, \\ r = 2^{2I-2} + (s - 2^K - 2^{I-1} + 1) (2^K + 3 \cdot 2^{I-1} - s - 2) / 2, \\ \text{where } P_3 = (2^K + 2^I - s - 2) (2^K + 2^I - s - 1).$$

c) in the case where $2^K \geq s - 2^{I-1} + 1$

$$z = z_3, \quad T = y = P_4 (2^K - 3 \cdot 2^{I-1} + s + 1), \quad x = P_4 (3 \cdot 2^{I-1} - 1), \quad r = 2P_4, \\ \text{where } P_4 = 2^{I-2} (s - 2^K + 1).$$

(4) For $CY_{I,K}^{(1)}$ ($K=1, 2, \dots, S-2$; $I=K+1, K+2, \dots, S-1$):

a) in the case where $2^I \leq s-2^K+1$

$$z=2^{K+I-2} \equiv z_4, \quad T=y=2^{K-2}(9 \cdot 2^{K-1}-7 \cdot 2^{2K-1}+9 \cdot 2^{K+I-2}-3 \cdot 2^{I-1}-1)/3 \equiv T_2,$$

$$x=2^{K-2}(7 \cdot 2^{2K-2}-9 \cdot 2^{K+I-1}+9 \cdot 2^{2I-2}+3 \cdot 2^{I-1}-1)/3 \equiv x_1, \quad r=2^{K-2}(2^I-3 \cdot 2^{K-1}+1) \equiv r_2.$$

b) in the case where $s-2^{K-1} \geq 2^I \geq s-2^K+2$

$$z=2^{K-2}(2^I-2^{K-1}+1)+(s-2^I-2^{K-1}+1)(3 \cdot 2^{K-1}+2^I-s-2)/2 \equiv z_6,$$

$$T=y=T_2, \quad x=x_1, \quad r=r_2.$$

c) in the case where $2^I \geq s-2^{K-1}+1$

$$z=2^{K-2}(2s-3 \cdot 2^{K-1}-2^I+3) \equiv z_6, \quad T=y=T_2, \quad x=x_1, \quad r=r_2.$$

(5) For $DY_{I,K}^{(1)}$ ($K=1, 2, \dots, S-2$; $I=K+1, K+2, \dots, S-1$):

a) in the case where $2^I \leq s-2^K+1$

$$z=z_4, \quad T=y=2^{K-2}(7 \cdot 2^{2K-1}-9 \cdot 2^{K-1}+1)/3, \quad x=2^{K-2}(9 \cdot 2^{K+I-1}-7 \cdot 2^{2K-2}-3 \cdot 2^I+1)/3, \\ r=2^{K-2}(3 \cdot 2^{K-1}-1).$$

b) in the case where $s-2^{K-1} \geq 2^I \geq s-2^K+2$

$$z=z_5, \quad T=y=2^{K-2}(7 \cdot 2^{2K-1}-9 \cdot 2^{K-1}+1)/3-P_3(s+2^{K+1}-2^I)/6,$$

$$x=2^{K-2}(9 \cdot 2^{K+I-2}-3 \cdot 2^I-7 \cdot 2^{2K-2}+1)/3-P_3(s-2^K+2^{I+1})/6,$$

$$r=2^{2K-2}+(s-2^{K-1}-2^I+1)(3 \cdot 2^{K-1}+2^I-s-2)/2.$$

c) in the case where $2^I \geq s-2^{K-1}+1$

$$z=z_6, \quad T=y=P_5(3 \cdot 2^{K-1}-1), \quad x=P_5(s-3 \cdot 2^{K-1}+2^I+1),$$

$$r=2P_5, \quad \text{where } P_5=2^{K-2}(s-2^I+1).$$

(6) For $A_{I,K}^{(2)}$ ($K=S$; $I=1, 2, \dots, K-1$):

a) in the case where $2^I \leq s-2^{K-1}$

$$z=r=2^{I-2}(2s-2^K-3 \cdot 2^{I-1}+3),$$

$$T=x=2^{I-2}\left\{3 \cdot 2^{K-1}-9 \cdot 2^{K+I-2}-7 \cdot 2^{2I-1}+9 \cdot 2^{I-1}(s+2)-3s-4\right\}/3,$$

$$y=2^{I-2}\left\{3(s-2^{K-1}-2^{I-1}+1)(s+2^{K-1}-2^{I-1})-(2^{I-1}-1)(3s-2^{I+1}+2)\right\}/3.$$

b) in the case where $s-2^{K-1}+1 \leq 2^I \leq 2s-2^K+1$

$$z=r=P_6/2, \quad T=x=P_6(s-2^{K-1}+2^I)/6, \quad y=P_6(s+2^K-2^{I-1})/6,$$

$$\text{where } P_6=(s-2^{K-1}-2^{I-1}+1)(s-2^{K-1}-2^{I-1}+2).$$

c) in the case where $2^I \geq 2s-2^K+2$

$$T=x=y=0.$$

(7) For $BY_{S,K}^{(2)}$ ($K=1, 2, \dots, S-1$):

a) in the case where $2^K \leq s-2^{S-1}$

$$z=r=2^{K-2}(2s-2^S-3 \cdot 2^{K-1}+3),$$

$$T=y=2^{K-2}\left\{3 \cdot 2^{S-1}-9 \cdot 2^{K+S-2}-7 \cdot 2^{2K-1}+9 \cdot 2^{K-1}(s+2)-3s-4\right\}/3,$$

$$x=2^{K-2}\left\{3(s-2^{S-1}-2^{K-1}+1)(s+2^{S-1}-2^{K-1})-(2^{K-1}-1)(3s-2^{K+1}+2)\right\}/3.$$

b) in the case where $s-2^{S-1}+1 \leq 2^K \leq 2s-2^S+1$

$$z=r=P_7/2, \quad T=y=P_7(s-2^{S-1}+2^K)/6, \quad x=P_7(s+2^S-2^{K-1})/6,$$

$$\text{where } P_7=(s-2^{S-1}-2^{K-1}+1)(s-2^{S-1}-2^{K-1}+2).$$

c) in the case where $2^K \geq 2s-2^S+2$

$$T=x=y=0.$$

(8) For $A_{K-1,K-1}^{(3)}$ ($K=2, 3, \dots, S$):

a) in the case where $3 \cdot 2^K \leq 3(s+1)$

$$z=2r=2^{2K-3}, \quad T=2x=2y=2^{2K-4}(3 \cdot 2^{K-2}-1).$$

b) in the case where $3s+4 \leq 3 \cdot 2^K \leq 4s$

$$z=2r=(s-3 \cdot 2^{K-2}+1)(5 \cdot 2^{K-2}-s-2)+2^{K-2}(2^{K-2}+1),$$

$$T=2x=2y=2^{2K-4}(3 \cdot 2^{K-2}+1)-(s+2^{K-1})(2^K-s-1)(2^K-s-1)(2^K-s-2)/3.$$

c) in the case where $3 \cdot 2^K \geq 4s+1$

$$z=2r=P_8, \quad T=2x=2y=P_8(s+2^{K-2})/3, \quad \text{where } P_8=(s-2^{K-1}+1)(s-2^{K-1}+2).$$

(9) For $AY_{K,K}^{(3)}$ ($K=1, 2, \dots, S-1$):

a) in the case where $3 \cdot 2^{K+1} \leq 3(s+1)$

$$z=r=2^{2K-2}, \quad T=x=y=2^{2K-3}(3 \cdot 2^{K-1}-1).$$

b) in the case where $3s+4 \leq 3 \cdot 2^{K-1} \leq 4s$

$$z=r=2^{K-2}(2^{K-1}+1)+(s-3 \cdot 2^{K-1}+1)(5 \cdot 2^{K-1}-s-2)/2,$$

$$T=x=y=2^{2K-3}(3 \cdot 2^{K-1}-1)-(2^{K+1}-s-1)(2^{K+1}-s-2)(s+2^K)/6.$$

c) in the case where $3 \cdot 2^{K+1} \geq 4s+1$

$$z=r=P_9/2, \quad T=x=y=P_9(s+2^{K-1})/6, \quad \text{where } P_9=(s-2^K+1)(s-2^K+2).$$

(II) If the coefficients $D_{K+1}^{(4)}$ and $E_K^{(4)}$ in Eq. (8) are denoted together as

$$\frac{cG'}{bn_0 d_1^3 h} \cdot \frac{v}{L_K} (L_l^{2f}-1),$$

the values of v and l are calculated as follows.

(1) For $D_{K+1}^{(4)}$ ($K=1, 2, \dots, S-1$):

a) in the case where $1 \leq K \leq S-2$

$$v=3(2^K-1)/2, \quad l=K+1.$$

b) in the case where $K \geq S-1$

$$v=(2^K+s)/2, \quad l=K+1.$$

(2) For $E_K^{(4)}$ ($K=2, 3, \dots, S$):

a) in the case where $2 \leq K \leq S-1$

$$v=1-L_K, \quad l=K.$$

b) in the case where $K=S$ and $s \neq 2^K-1$

$$v=-L_K, \quad l=K.$$

c) in the case where $K=S$ and $s=2^K-1$

$$v=(1-L_K), \quad l=K.$$