# Simplified flocculation model for inorganic and polymer flocculants

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# (Short title: Simplified flocculation model)

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## ABSTRACT

Simulation of the flocculation process using both inorganic and polymer flocculants is very difficult because there are many factors which influence the process. In this study, a model which enables the visual understanding of qualitative trends of the flocculation system using both inorganic and polymer flocculants is proposed. It is a simplified one dimensional model which expresses flocculation under various additive manners of the flocculant. Various kinds of thought experiment were carried out by use of the model; the results demonstrate that the model can express many empirical qualitative trends of flocculation.

**Keywords:** Flocculation model, inorganic flocculant, polymer flocculant

## INTRODUCTION

Flocculation is a process in which individual particles of a suspension aggregate to form flocs. It is a common practice to use a polymer flocculant such as polyacrylamide concomitantly used with an inorganic flocculant such as alum. There are considerable researches conducted on flocculation process using inorganic and/or polymer flocculant. Theoretical development of particle collision model for the expression of the flocculation process spans a period of close to a 100 years (1-12), originated from the Smoluchowski equation using a population balance approach (1). Floc strength and breakage during the process had also been discussed by various researchers (10, 13, 14). However, a strict quantitative expression of the flocculation process seems to be far from complete; e.g. a flocculation by polymer flocculant is affected by many factors such as molecular weight of the polymer, its constituent monomer species and their combination, its concentration in a target colloidal system, and the affinity between polymer and colloidal particles (15-24). Although it is empirically known that the flocculation behavior using polymer flocculant depends on a slight difference of dispersion of polymer flocculant in a colloidal suspension

(25, 26), there is not any quantitative explanation for that. Hogg has assumed that polymer flocculant molecules randomly adsorb onto surfaces of colloidal particles and arrived at the conclusion that the occurrence of flocculation depends on the number of adsorption sites on a particle surface (27). The theoretical expression of flocculation process using both inorganic and polymer flocculant is even more difficult because there are many factors which influence the process; e.g. an adsorption rate of polymer flocculant molecules onto colloidal particles and its morphology on the particles strongly depends on ionic strength (i.e. concentration of inorganic flocculant) (17, 19, 20).

Sakohara et al. have developed a simplified one dimensional model which can express the effect of additive manner of a polymer flocculant solution on flocculation process (25). In this study, we propose an extension of Sakohara's model which enables us to understand the flocculation process using both inorganic and polymer flocculants. The model is one dimensional simplified model which can express qualitative trends of flocculation processes under various additive manners of flocculant solution. The reproducibility of the processes is also discussed by using the model.

## VISUALIZED ONE DIMENSIONAL FLOCCULATION MODEL

### Flocculation model for polymer flocculant

Following Sakohara's definition of legend symbols, we represent a polymer flocculant

molecule by a straight line which has only two active sites at both ends, and a suspended particle by a circle which also has only two active sites on its surface, as shown in Fig. 1. We simplify the bridging flocculation by polymer flocculant as shown in Fig. 2. Flocculation with polymer flocculant is assumed to occur in case of only one polymer flocculant existing between particles as indicated by the arrows in Fig. 2. On the other hand, the flocculation is assumed not to occur in case of more than one polymer flocculants existing between particles as indicated by the triangle in Fig. 2.

It is a well known fact that a colloidal suspension is sometimes stabilized by hydrophilic nature of polymer flocculant which excessively adheres to the particle surface (26). We express this situation in our model as shown in Fig. 3, where two polymer molecules exist between two particles and all active sites on particle surface are filled with polymer flocculant molecules. A lump of polymer flocculant and its excess adsorption on particles can be represented by Fig. 4; i.e. the former is represented by more than two free polymer molecules, while the latter is expressed by four molecules at both sides of a particle. Once the relative position of particles and flocculant molecules is assigned, it remains unchanged to represent the above unevenness of particle-flocculant mixture which depends on additive manner of flocculant.

**Flocculation model for inorganic flocculant**

Inorganic flocculant can disperse quickly just after its addition to a colloidal suspension. We express the existence of inorganic flocculant by a background color as shown in Fig. 5.

Flocculation with inorganic flocculant progresses much slower than with polymer flocculant. Slow flocculation by inorganic flocculant is simplified as shown in Fig. 6. Flocculation is assumed to occur in one place per $N$ spaces. In this study $N = 4$; i.e. flocculation by inorganic flocculant occurs where indicated by the arrows in Fig. 6.

**Factors that are not considered in the model**

For the purpose of simplification, the model does not consider the following events which are encountered in practice.

1. Breakages of flocs.
2. Changes in flocculation rate due to the growth of the flocs.
3. Changes in the compatibility among colloidal particles, polymer flocculant and inorganic flocculant due to their combination.
4. Changes in morphology and performance of polymer flocculant due to the existence of inorganic flocculant.
5. Changes in adsorption rate and adsorbed amounts of polymer flocculant due to the change of the flocculant morphology and its concentration.
6. Flocculation by polymer flocculant other than by the bridging effect.

Therefore, the model is restricted to the flocculation system where the polymer flocculant shows a stable performance regardless of the presence of inorganic flocculant.

## SIMULATION RESULTS USING THE ONE DIMENSIONAL MODEL

Figures 7 to 9 show visualized simulation results based on the simplified flocculation model explained above.

### Flocculation when using inorganic flocculant alone

Figure 7A represents the simulation result when using inorganic flocculant alone. The 1st to 13th block in the figure show the time step during which 1/4 of the spaces between particles disappear due to the combination of particles on two sides of the space. The space '1' disappeared in each step and we renumbered the spaces between particles at the following step, where the number of the left-end space was assigned the next one of the right-end space in the previous step, as shown in the figure. The figure represents that the flocculation by inorganic flocculant progresses slowly and steadily compared with polymer flocculant shown in the next section.

### Flocculation when using polymer flocculant alone

Figures 8B.1 to 8B.3 show the simulation results when using polymer flocculant alone. Here, we assumed that the flocculation by polymer flocculant is completed in one step. The

effect of dosage of polymer flocculant on the flocculation process was examined under good dispersion condition of flocculant molecules. In Fig. 8B.1, the flocculation was terminated at the 2nd step because of lack of sufficient flocculant molecules. The result of flocculation shown in Fig. 8B.2 was satisfactory because of adequate amount of flocculant, while in Fig. 8B.3 the suspension was stabilized due to an excessive dose of flocculant. The model can represent the existence of an optimum dosage of polymer flocculant.

Figures 8C.1 to 8C.3 are the results with different dispersion of polymer flocculant under the same polymer dosage. Numbers in the figures represent the degree of dispersion $S^2$ defined by the following equations. It implies the deviation from 'uniform conformation' of polymer molecules and particles.

$$S^2 = S_1^2 + S_2^2 \tag{1}$$

where $$S_1^2 = \frac{1}{N+1}\sum_{k=0}^{N}\left(p_k - \frac{P}{N+1}\right)^2 \tag{2}$$

and $$S_2^2 = \frac{1}{P+1}\sum_{k=0}^{P}\left(n_k - \frac{N}{P+1}\right)^2 \tag{3}$$

Here $N$ is the total number of particles, and $P$ is the total number of polymer molecules. $S_1^2$ denotes the deviation from the uniform conformation of polymer molecules with respect to $N$+1 positions, i.e. $N$-1 positions between $N$ particles and two ends, the left position of the particle at the left end and the right position of the particle at the right end. $S_2^2$ is the deviation from the uniform conformation of particles to the $P$+1 positions, i.e. $P$-1 positions between $P$

molecules of polymer flocculant and two ends, the left position of the molecule at the left end and the right position of the molecule at the right end. $p_k$ in Eq. (2) is the number of polymer molecules between $k$th and ($k$+1)th particles, and $n_k$ in Eq. (3) is the number of particles between $k$th and ($k$+1)th polymer molecules. The minimum $S^2$-value under fixed values of $N$ and $P$ implies the best dispersion under such condition. $S^2 < 2$ represents a good dispersion, while $S^2 > 2$ shows a bad dispersion in this study. Figures 8C.1 to 8C.3 show that the dispersion status affects the flocculation process. A total number of particles and flocs in colloidal suspension under a given dose of polymer flocculant depends on the dispersion status of the flocculant; i.e. an inhomogeneous distribution of polymer flocculant shown in Fig. 8C.1 results in no flocculation, while a homogeneous distribution shown in Fig. 8C.3 results in perfect flocculation.

Figures 8D.1 to 8D.5 show the results with extreme underdose and extreme overdose of polymer flocculant. In extreme under- or over-dosages, the degree of dispersion does not affect the results, as is evident from the comparison of Figs. 8D.1 and 8D.2 as well as the comparison of 8D.3 and 8D.4. Although the situation shown in Fig. 8D.5 results in the perfect flocculation, this is highly unlikely to happen.

Figures 9E.1 and 9E.2 are the results when adding polymer flocculant intermittently, i.e. adding a divided amount of flocculant in each step; the figures show that the degree of dispersion $S^2$ does not affect the results so much when adding intermittently. Adding only one

molecule in each step, the stabilization of particles due to over-dose would never arise as shown in Fig. 9E.3.

**Flocculation when using polymer flocculant in combination with inorganic flocculant**

Figures 10F.1 and 10F.2 are the results when adding inorganic flocculant first and then polymer flocculant. Comparison of Figs. 10F.1 and 7A shows that the addition of polymer flocculant decreases the number of steps until obtaining the maximum size of floc; i.e. the combination shortens the flocculation time.

In Fig. 10F.2, the same amount of polymer flocculant as in Fig. 8B.2, i.e. 11 molecules, was used, resulting in stabilization of the system in the 6th step. This is because the active sites of particles for polymer adsorption decreased due to the progress of flocculation by inorganic flocculant.

Figure 10G is the result when adding polymer firstly and then inorganic flocculant. The figure suggests that if the dose of polymer flocculant is appropriate, this mode of addition works well; i.e. in the latter half of the process, it proceeds slowly but stably by the inorganic flocculant.

Figure 10H shows the result when adding inorganic flocculant firstly and then polymer flocculant intermittently. Such mode of addition comprises all advantages shown in Figs. 9E.1, 10F.1 and 10G. That is, this mode of flocculant addition can save both operation time and

dosage of polymer flocculant.

## EXPERIMENTAL

A 0.4 μm polymethyl methacrylate particle (PMMA: MP-1000, Soken Chemical & Eng. Co.) was used as the model colloidal material. Aluminum sulfate (alum) was used as an inorganic flocculant. As a polymer flocculant, a cationic polymer flocculant (DIAFLOC KP204BS, Mitsubishi Rayon Co., Ltd., M.W. 13000 kDa, main component: dimethylaminoethyl acrylate) was used. 10 mL of PMMA suspension (2000 ppm) was introduced into a 25 mL test tube. This suspension was mixed with 10 – 15 mL of the flocculant solution by turning upside down using hand 30 times for 30 seconds. When using polymer flocculant, the test tube was inverted slowly 1 time for 3 min until the time of measurement to prevent settling of flocs, as shown in Fig. 11. Then, 3 mL of the mixture was taken to a cell of particle size analyzer (CAPA-300, Horiba, Ltd.) to measure the floc size distribution by the centrifugal method. When using an inorganic flocculant singly, 3 mL of the colloid/flocculant mixture was taken to a cell just after the mixing, followed by slowly inverting the cell for once for 3 min until the time of measurement as shown in Fig. 12. Each experimental run was repeated 5 times.

## RESULTS AND DISCUSSION

Figures 13 to 17 show the median diameter of suspension under various experimental

conditions, while in Figs. 18 to 22 their floc size distributions are shown. In Figs. 13 to 17, the upper and lower ends of error bars represent the maximum and minimum values, respectively. The keys in the figures show an arithmetic mean of each run.

Figures 13 and 18 represent the results when 10 mL of inorganic flocculant (2000 ppm) was added singly to an equal amount of suspension (2000 ppm PMMA); i.e. the concentration of inorganic flocculant in the colloid/flocculant mixture was 1000 ppm. The critical coagulation concentration (c.c.c) of the PMMA suspension was 79 ppm of alum in the mixture. It can be seen that flocculation by inorganic flocculant proceeds slowly and steadily, as expected in Fig. 7A.

Figures 14 and 19 are the results when 10 mL suspension (2000 ppm) was diluted firstly by 8 mL of water and then mixed with 2 mL of polymer flocculant (100 ppm solution of KP204BS). The flocculation by polymer flocculant was very fast; the floc size distribution has not changed after 30 s mixing time as shown in the figures. This supports the assumption used in our model; i.e. the flocculation by polymer flocculant is completed in one step as shown in all simulation results except Fig. 7A.

Figures 15 and 20 show the results where 10 mL suspension was firstly mixed with water, and then with a polymer flocculant. The amounts of water and flocculant are shown in Table 1.

It is a well known fact that the median diameter of floc formed by polymer flocculant

varies depending on the dosage of flocculant, showing the existence of the optimum dosage where the maximum value of median diameter is obtained. We also have confirmed this empirical knowledge as shown in Fig.15. In the figure, the reproducibility of data was poor near the optimum dosage, while it was satisfactory under other dosages. This result is in accord with the simulation results shown in Figs. 8C.1 to 8C.3 and 8D.1 to 8D.4; i.e. Figs. 8C.1 to 8C.3 represent that the results depend strongly on the degree of dispersion $S^2$ at the imaginary optimum dosage (9 polymers for 10 particles), while Figs. 8D.1 to 8D.4 show that in extreme underdose or extreme overdose of polymer flocculant, the results are affected not so much by the degree of dispersion $S^2$.

Figures 16 and 21 show the results when 10 mL of suspension (2000 ppm) was mixed with 5 mL of 100 ppm polymer flocculant under various mixing methods. These figures contain the results of intermittent addition of polymer solution of the same amount. It can be seen from Fig. 16 that the intermittent addition is the most effective method for obtaining the largest floc; Fig. 21 shows that the intermittent addition works well in removing small size flocs in suspension. The effectiveness of the intermittent addition can also be expressed clearly by the simulation results shown in Figs. 9E.1 to 9E.3.

Figures 17 and 22 show the results of flocculation when 10 mL of 2000 ppm suspension was mixed with 10 mL of 2000 ppm inorganic flocculant solution firstly and then mixed with a polymer flocculant solution. The amount of dosage of polymer flocculant was shown in

Table 2. The optimum dosage of polymer flocculant was 0.005 mg/mg-solid as shown in Fig. 17. This value is less than that (0.01 mg/mg-solid) in Fig. 15 where adding polymer flocculant alone. This difference was expected earlier in comparison of Figs. 8B.2 and 10F.1.

As mentioned above, the simulation model proposed in this paper can explain qualitatively flocculation processes under various conditions. Number of primary particles or polymer molecules in the simulation was restricted up to several tens in this study for visual comprehensibility. The model seems to be suitable for finding fundamental aspects of the flocculation process by computer simulation. Further research will be conducted along this line.

## CONCLUSIONS

A simplified one dimensional model for flocculation using both inorganic and polymer flocculants has been proposed.

The model can express the following empirical knowledge.

1. Flocculation with inorganic flocculant is reproducible.
2. Reproducibility of flocculation with polymer flocculant depends on the dosage, additive manner and mixing intensity.
3. Flocculation with polymer flocculant progresses faster than with inorganic flocculant.
4. It would be better to disperse polymer flocculant well for better turbidity removal.
5. The intermittent addition of polymer flocculant gives better turbidity removal.

6. For the flocculation process using both inorganic flocculant and polymer flocculant, an optimum amount of polymer flocculant decreases as flocculation with inorganic flocculant progresses.

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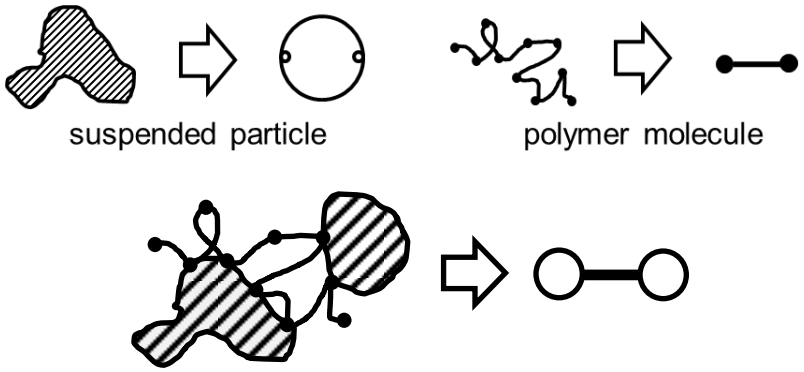


FIG. 1. Legend symbols of suspension particle and polymer flocculant and modeling of bridging flocculation.

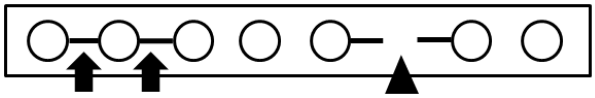

FIG. 2. Modeling of flocculation by polymer flocculant. Bridging occurs where it is indicated by the arrows, and does not occur where it is indicated by the triangle.

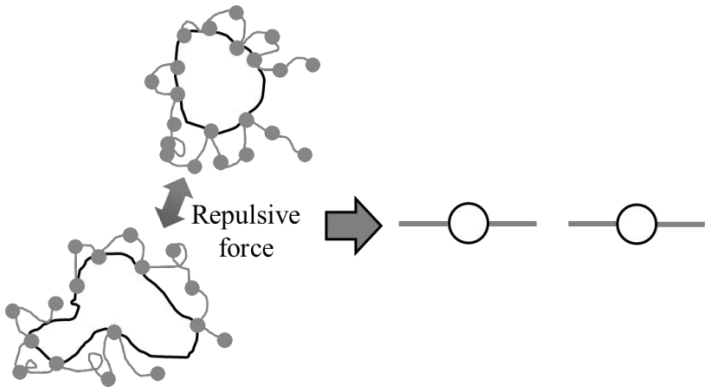


FIG. 3. Modeling of stabilization of colloidal particles with an excess of polymer flocculant.

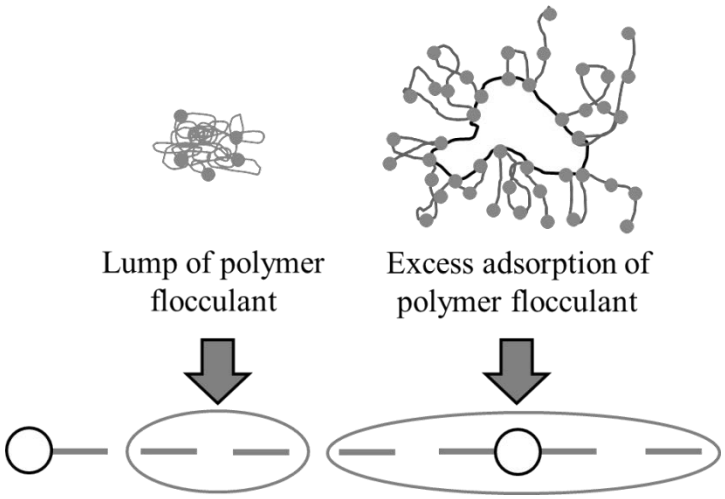


FIG. 4. Modeling of lump and excess adsorption of polymer flocculant.

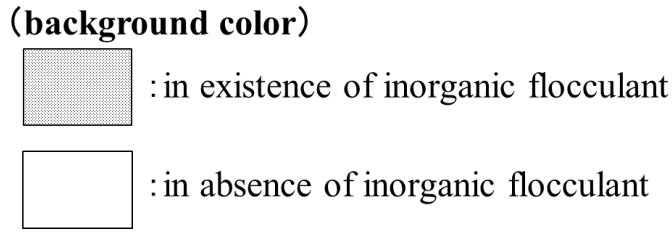


FIG. 5. Legend symbols of inorganic flocculant.

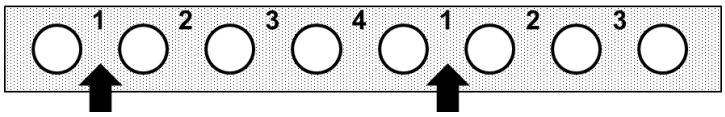


FIG. 6. Modeling of flocculation by inorganic flocculant. Flocculation occurs where it is indicated by arrows.

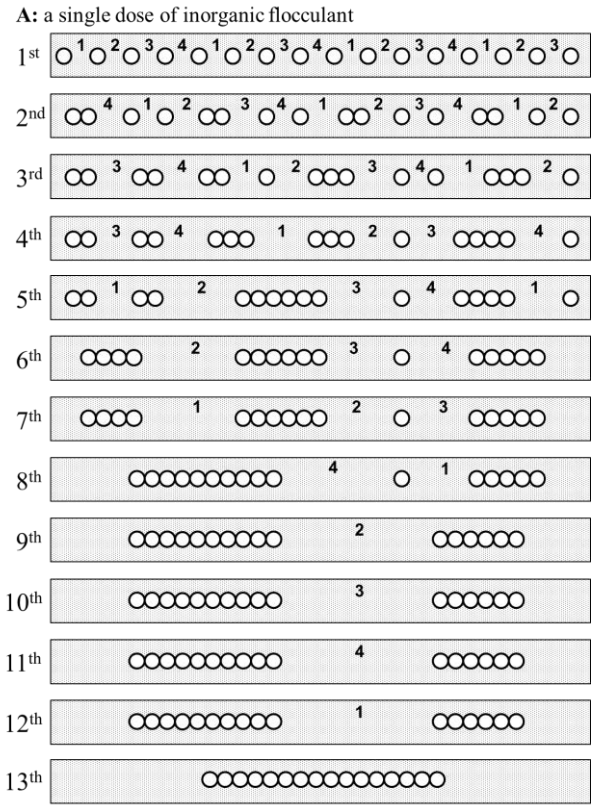


FIG. 7. Results of visualized simulation based on the model when using inorganic flocculant alone.

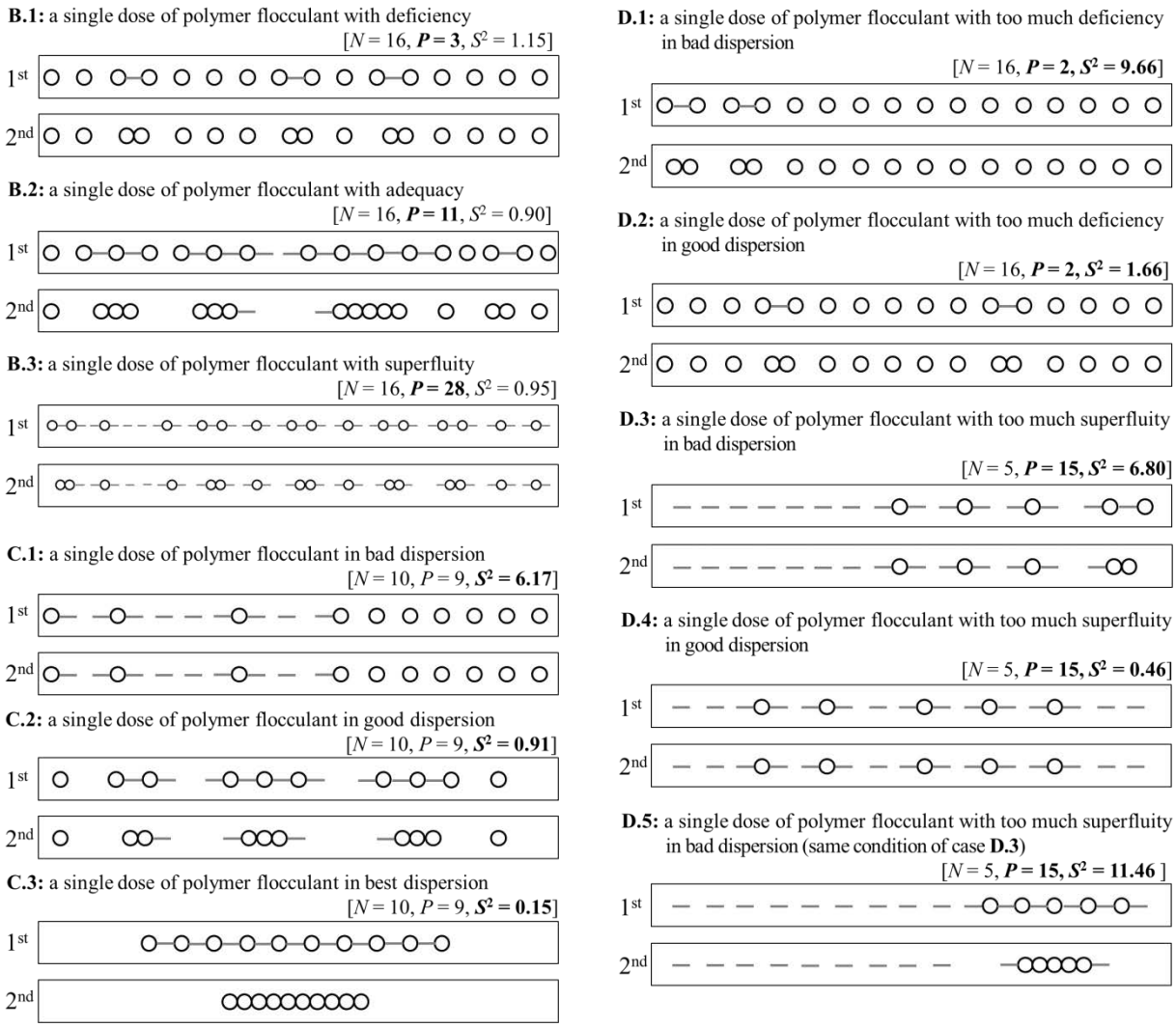


FIG. 8. Results of visualized simulation based on the model when using polymer flocculant alone. The 2nd step represents the final state of flocculation.

FIG. 9. Results of visualized simulation based on the model when adding polymer flocculant intermittently ($P$ = 1 or 2 per step). $N$ represents the total number of free primary particles and flocs, and $S^2$ was evaluated at each step.

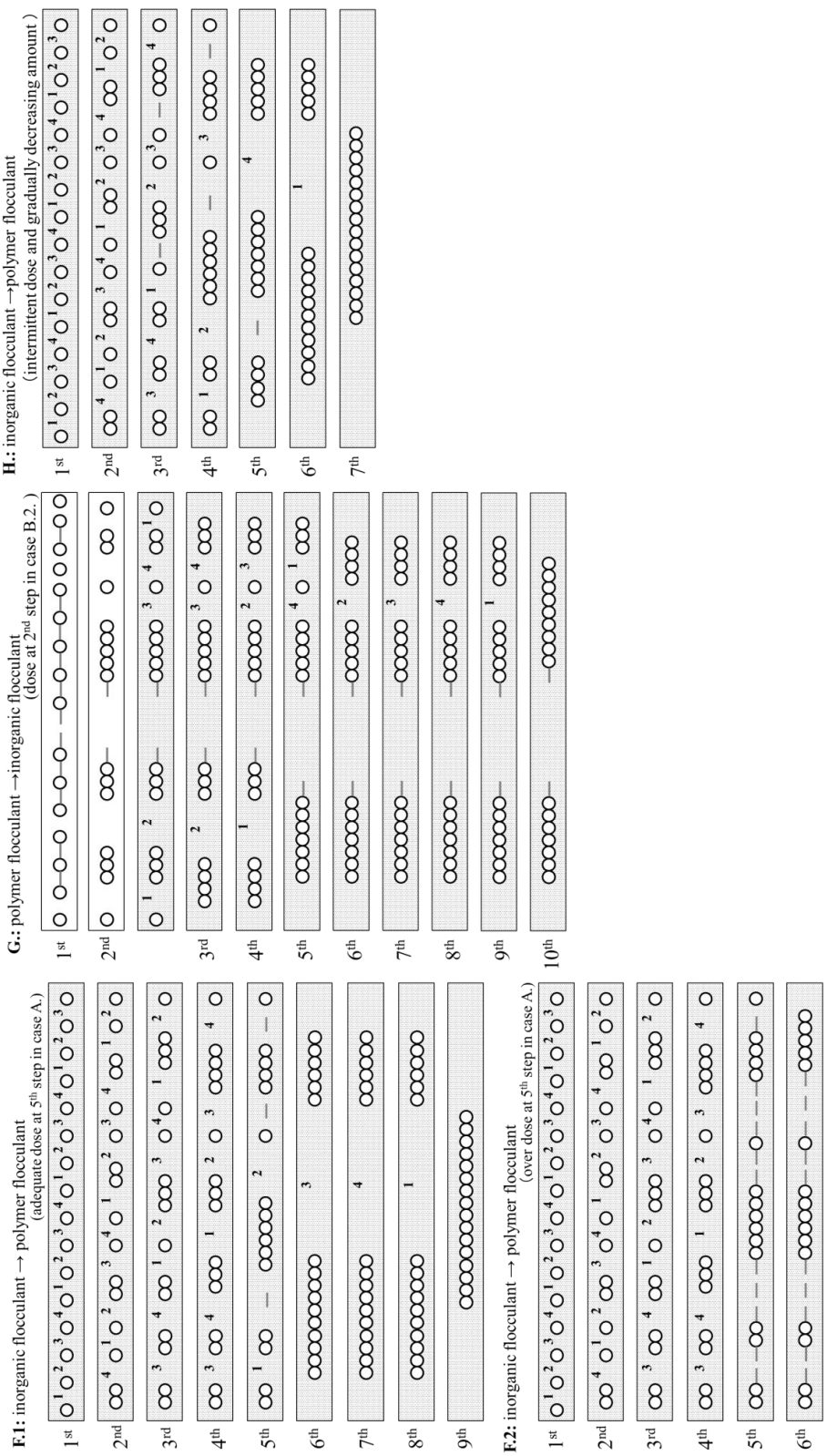


FIG. 10. Results of visualized simulation based on the model when using polymer flocculant in combination with inorganic flocculant. In case G, polymer flocculant was added first and then inorganic flocculant added. The other cases were conducted in reverse order.

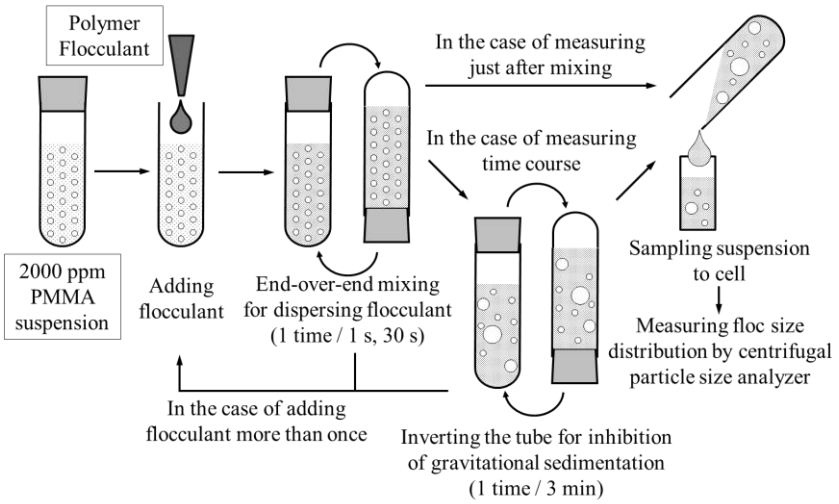


FIG. 11. Measuring method of floc size distribution of suspensions when using polymer flocculant alone or using both inorganic and polymer flocculants.

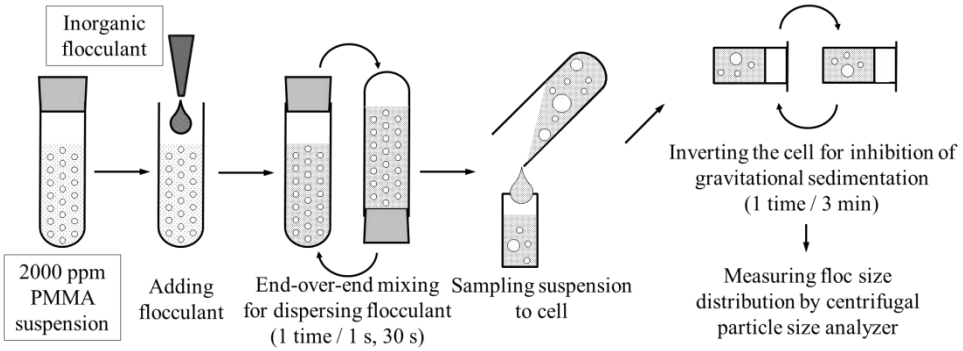


FIG. 12. Measuring method of floc size distribution of suspensions when using inorganic flocculant alone.

TABLE 1 Dosages of water and polymer solution of experimental runs shown in Figs. 15 and 20.

| Run No | Water | Polymer solution | | Mass ratio of polymer /suspended particle [mg/mg] |
|---|---|---|---|---|
| | Dosage [ml] | Concentration [ppm] | Dosage [ml] | |
| 1~5 | 9 | 10 | 0.0005 | 0.0005 |
| 6~10 | 8 | 10 | 0.001 | 0.001 |
| 11~15 | 5 | 10 | 0.0025 | 0.0025 |
| 16~20 | 9 | 100 | 0.005 | 0.005 |
| 21~25 | 8 | 100 | 0.01 | 0.01 |
| 26~30 | 5 | 100 | 0.025 | 0.025 |
| 31~35 | 9 | 1000 | 0.05 | 0.05 |
| 35~40 | 8 | 1000 | 0.1 | 0.1 |

TABLE 2 Dosages of polymer solution of experimental runs shown in Figs. 17 and 22.

| Run No | Polymer solution | | Mass ratio of polymer /suspended particle [mg/mg] |
|---|---|---|---|
| | Concentration [ppm] | Dosage [ml] | |
| 1~5 | 10 | 2 | 0.001 |
| 6~10 | 10 | 5 | 0.0025 |
| 11~15 | 100 | 1 | 0.005 |
| 16~20 | 100 | 2 | 0.01 |
| 21~25 | 100 | 5 | 0.025 |

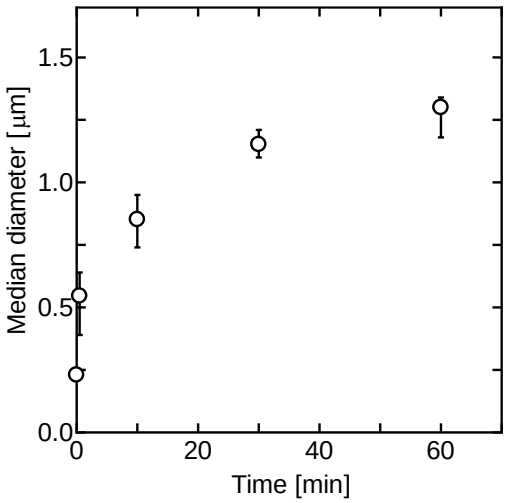


FIG. 13. Time course of the median diameter of flocs in suspension after the dose of inorganic flocculant solution alone.

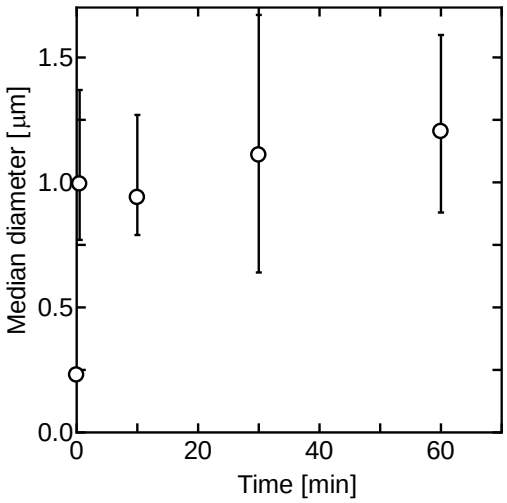


FIG. 14. Time course of the median diameter of flocs in suspension after the dose of polymer flocculant solution alone.

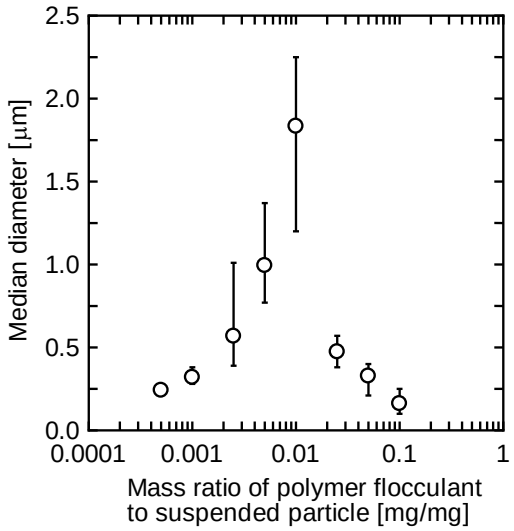


FIG. 15. Effect of polymer flocculant dosage on the median diameter of flocs at just after end-over-end mixing.

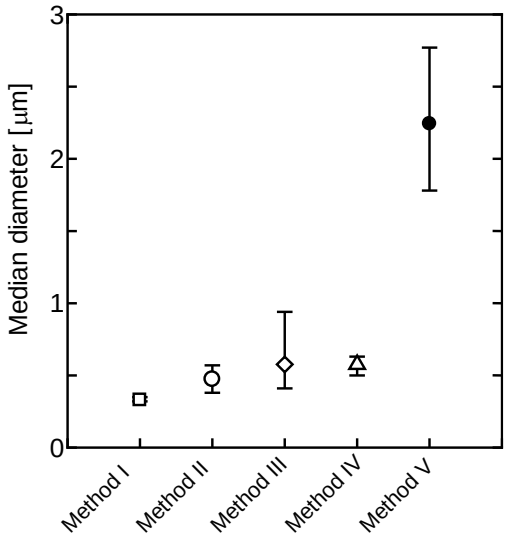


FIG. 16. Effect of addition manner and mixing method of polymer flocculant on the median diameter of flocs.

I: One-time dose, no mixing

II: One-time dose, end-over-end mixing

III: One-time dose, test tube shaking apparatus

IV: One-time dose, ultrasonic bath

V: 5-times stepwise dose, end-over-end mixing

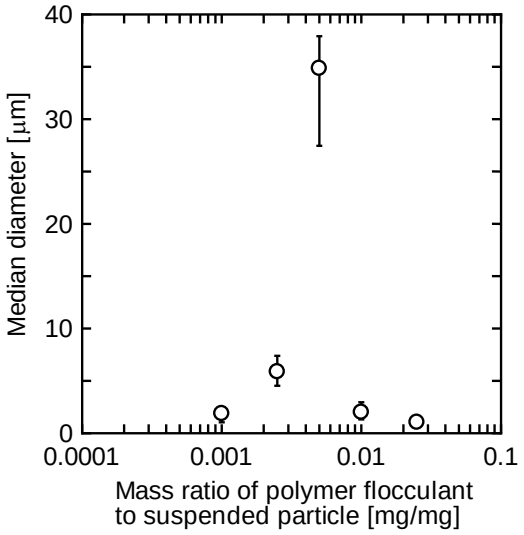


FIG. 17. Effect of polymer flocculant dosage on the median diameter of flocs at just after end-over-end mixing for polymer flocculant which was added 10 min after inorganic flocculant addition.

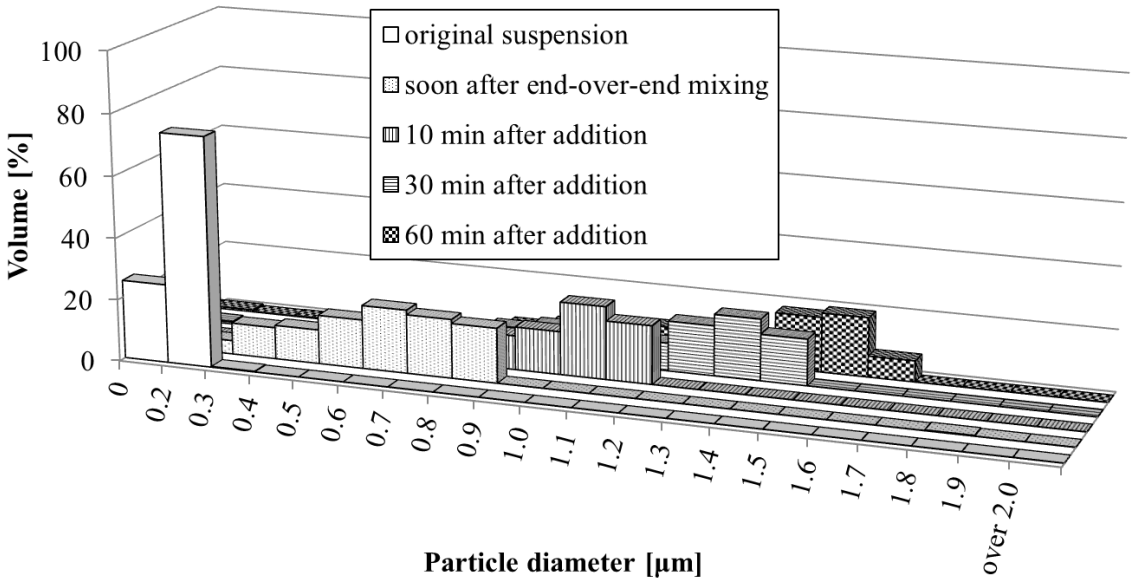


FIG. 18. Time courses of floc size distribution when singly using inorganic flocculant alone.

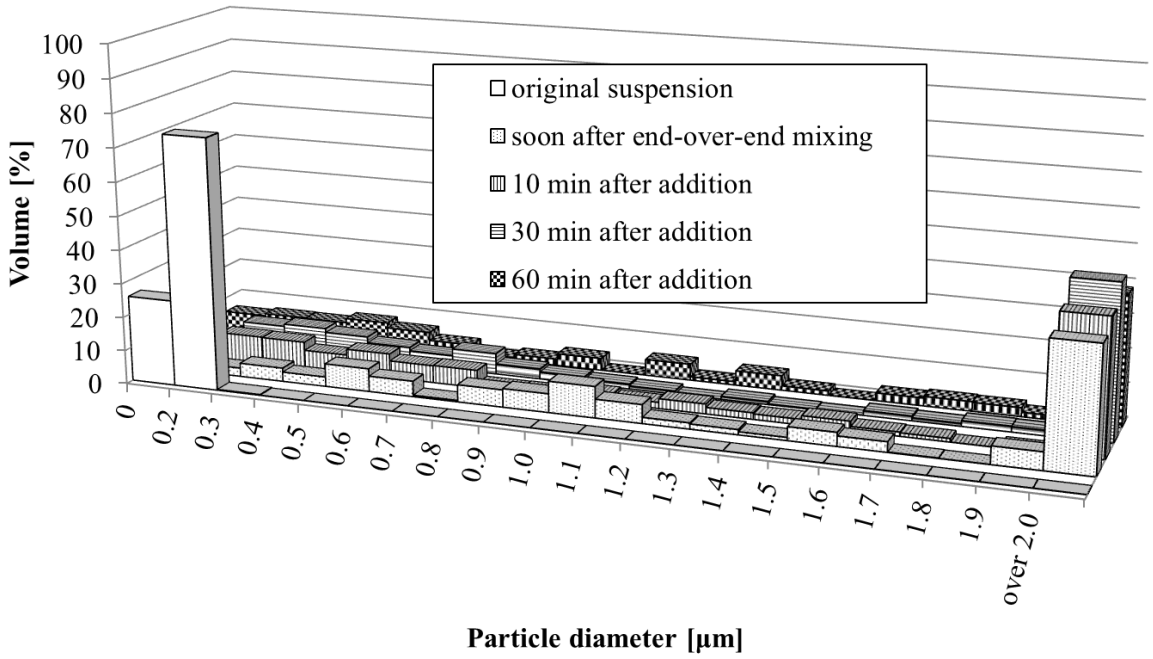


FIG. 19. Time courses of floc size distribution adding polymer flocculant alone.

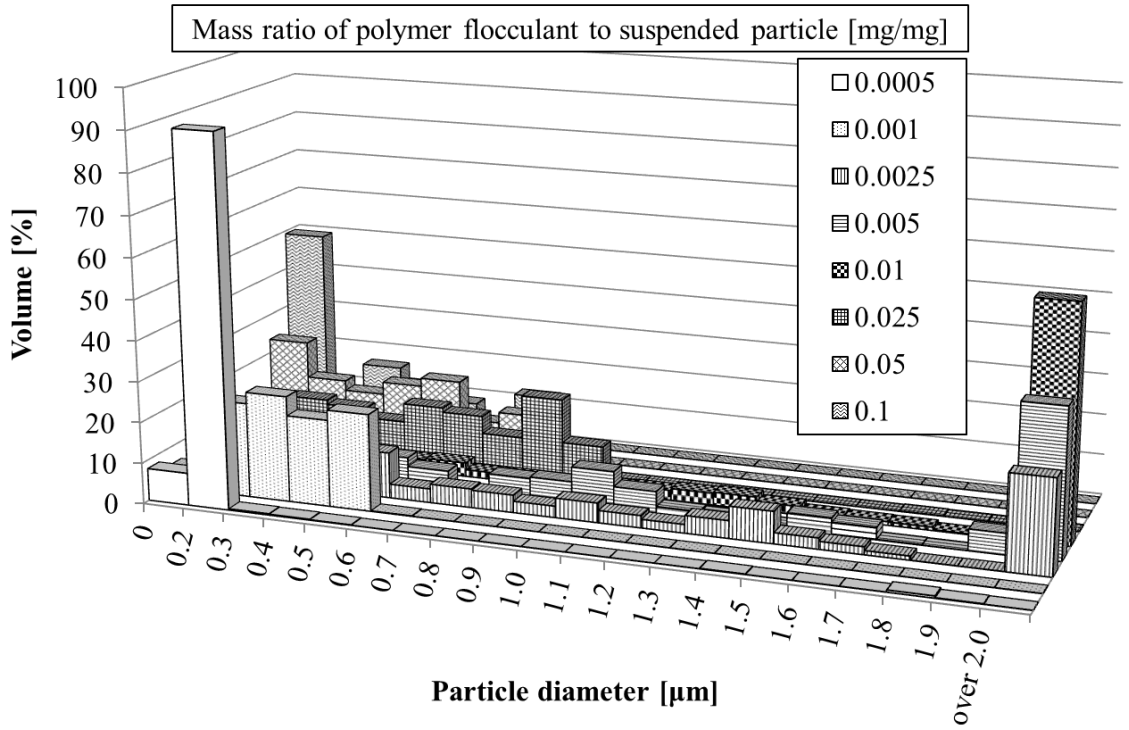


FIG. 20. Effect of polymer flocculant dosage on floc size distribution soon after end-over-end mixing.

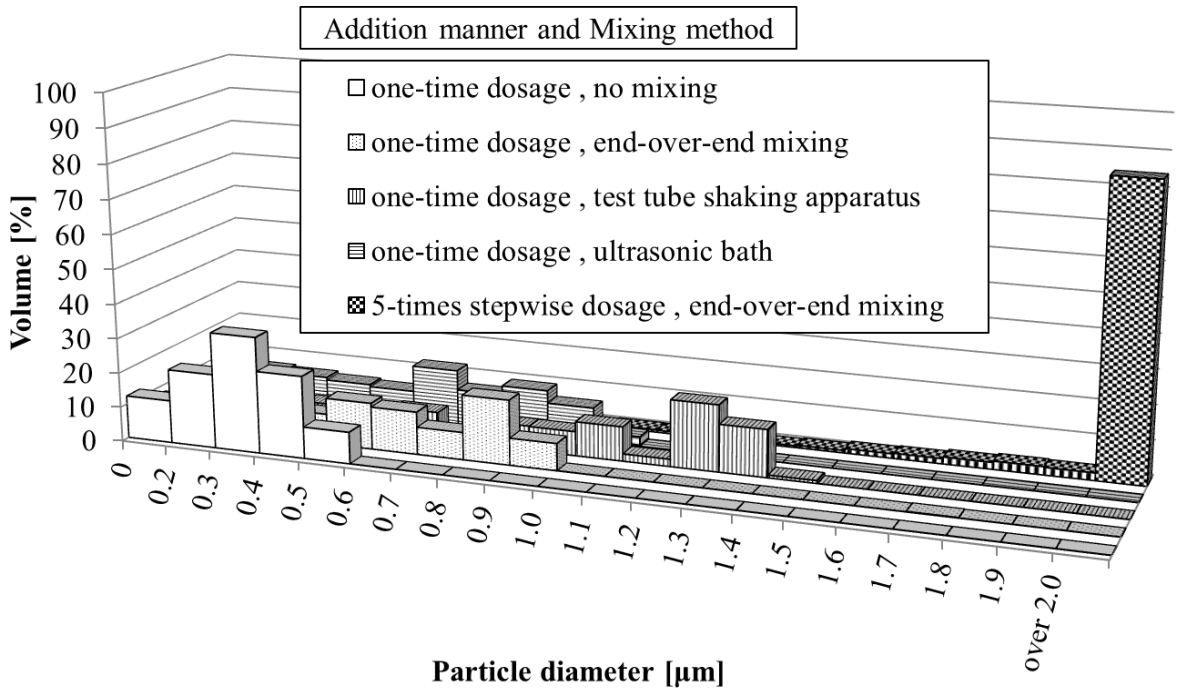


FIG. 21. Effect of addition manner and mixing method of polymer flocculant solution on floc size distribution soon after mixing.

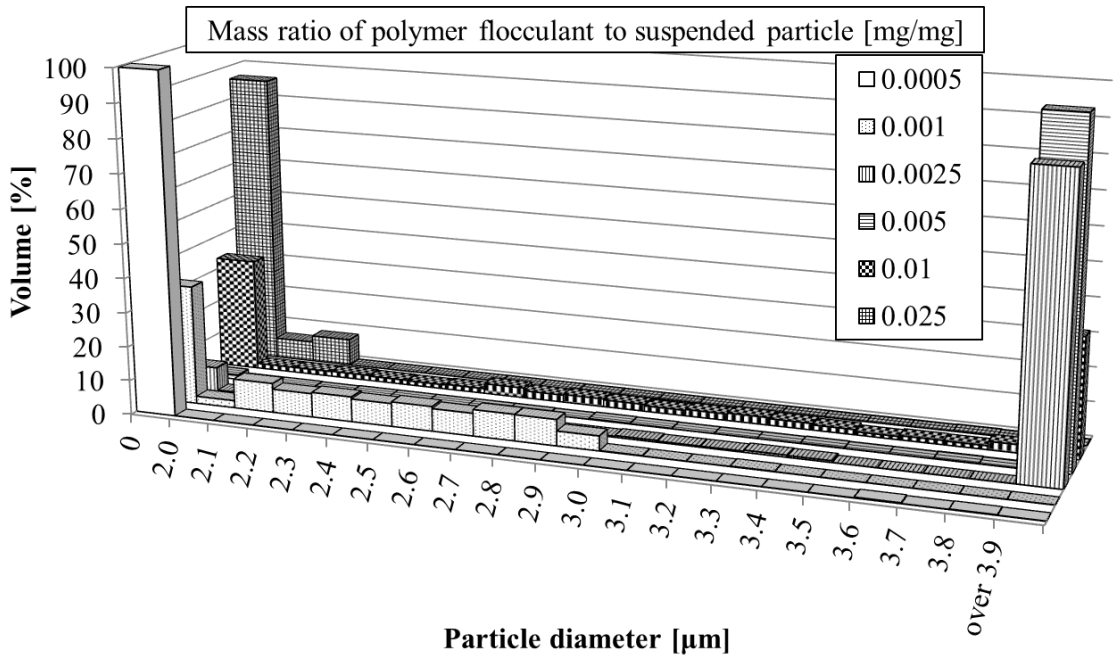


FIG. 22. Effect of polymer flocculant dosage on floc size distribution adding polymer flocculant 10 min after inorganic flocculant dose.