

Effects of Slow-Mixing on the Coagulation Performance of Polyaluminum Chloride (PACl)^{*}

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Abstract Conventional jar tests and on-line size monitoring were used to investigate the effects of slow-mixing intensity and duration on residual turbidity and floc size during charge neutralization coagulation and sweep flocculation with polyaluminum chloride. The compensatory effect of slow-mixing on coagulation performance following inadequate or excessive rapid-mixing was also examined. It is found that slow-mixing intensity has a more marked positive effect on charge neutralization coagulation than on sweep flocculation. The optimal root-mean-square velocity gradient, G , for slow-mixing is 15 s^{-1} for both coagulation mechanisms, and charge neutralization coagulation requires a longer slow-mixing duration. The optimal slow-mixing duration, based on residual turbidity, is longer than the time to form the largest mean flocs. The optimal product of G and mixing duration, GT , for slow-mixing during charge neutralization coagulation (13500) are higher than that during sweep flocculation (4500) and both are less than the range of values recommended by the American Water Works Association (24000-84000). The optimal GT value under various slow-mixing conditions increases with G . Appropriate extension of slow-mixing duration during charge neutralization coagulation can improve coagulation performance after an inadequate or excessive rapid-mixing duration, but during sweep flocculation, appropriate shortening of slow-mixing duration after an excessive rapid-mixing or appropriate extension of slow-mixing duration after an inadequate rapid-mixing is favorable.

Keywords slow-mixing, polyaluminum chloride, charge neutralization coagulation, sweep flocculation, GT value

1 INTRODUCTION

Coagulation is an important process that enhances separation in unit operations such as sedimentation, flotation and filtration, and is widely used for water and wastewater treatment. Many factors are involved in the coagulation process, including mixing conditions, coagulants, coagulation mechanisms and pH values.

There are generally two mixing regimes in the coagulation process, namely rapid-mixing and slow-mixing. The primary function of slow-mixing is to keep particles in suspension so that collisions between particles occur [1]. Furthermore, slow-mixing can provide a velocity gradient for collisions between particles of similar size that are larger than $1\text{ }\mu\text{m}$ [2, 3]. It is believed that eddies produced by mixing with similar size as particles can promote collisions between them and that larger eddies can only prevent particles from settling. High mixing intensities, characterized by the root-mean-square velocity gradient, G [4], will produce small eddies. Therefore, an appropriate increase in G is good for coagulation. However, eddies that are too small will induce floc breakup so it is very important to select proper G value for slow-mixing. Of course, slow-mixing duration is also important for coagulation and the GT value, the dimensionless product of G and

mixing duration, is usually recognized as an important design parameter for coagulation processes. The range of GT values recommended by the American Water Works Association (AWWA) is 24000-84000 and a typical slow-mixing duration is 20 min [5].

The size, strength and structure of flocs formed during coagulation are vital to subsequent separation units such as sedimentation, flotation and filtration. Many researchers [6-12] have found that, in addition to slow-mixing conditions, coagulants and coagulation mechanisms have significant effects on the size, strength and structure of flocs. This indicates that the effects of slow-mixing on coagulation are probably influenced by coagulants and coagulation mechanisms. Most researches and conclusions about slow-mixing during coagulation are based on single-molecule inorganic salt coagulants, such as aluminum sulfate and iron chloride [5, 13, 14]. However, there has been a lack of systematic research into the effects of slow-mixing on coagulation with inorganic polymer coagulants such as polyaluminum chloride (PACl) under different coagulation mechanisms. In fact, there are clear differences in molecular structure, formation conditions of hydrolyzed precipitates, morphology and properties of flocs between PACl and single-molecule inorganic salt coagulants [8, 15-18]. PACl has gradually replaced

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single-molecule inorganic salt coagulants and has been widely applied because it has the advantages of efficient coagulation rates, low temperature dependence and wide pH applicability range [17-21].

In this study, based on conventional jar tests, the effects of slow-mixing on coagulation with PACl are investigated by measuring the residual turbidity of a kaolin suspension after coagulation-settlement and monitoring the variation in floc size during charge neutralization coagulation and sweep flocculation. In addition, the compensatory effect of slow-mixing on inadequate or excessive rapid-mixing durations is studied.

2 MATERIALS AND METHODS

2.1 Experimental materials

The PACl (Nanning Chemical Industry Co., Ltd, Nanning, China) was in powdered form, with a basicity (OH/Al) of 1.35 and an Al_2O_3 content of 30%. The stock solution was obtained by dissolving PACl in deionized water, to an Al concentration of $2 \text{ mol}\cdot\text{L}^{-1}$. The fresh PACl solution for the tests had an Al concentration of $0.1 \text{ mol}\cdot\text{L}^{-1}$, obtained by diluting the stock solution one day before each batch of tests to avoid ageing and to maximize repeatability [22].

A stock solution of kaolin was prepared by dispersing 10 g kaolin in 5 L water, to give a concentration of $2 \text{ g}\cdot\text{L}^{-1}$. This could be used after rapid-mixing for 24 h. The experimental suspensions were obtained by diluting the stock solution to a concentration of $100 \text{ mg}\cdot\text{L}^{-1}$, with a turbidity of 68 NTU. The kaolin was chemically pure, with a particle size distribution of 0.275-39.8 μm and a mean particle size of 10.1 μm , produced by Beijing Xudong Chemical Plant, Beijing, China.

2.2 Experimental methods

2.2.1 Jar test

The conventional coagulation jar tests were

conducted with a JTY-4 mixer made by Beijing Daiyuan Measurement & Control Technology Development Center, Beijing, China. The coagulation vessel was a 1 L cylindrical container with an inner diameter of 105 mm. The mixing was provided by a $50\times 40 \text{ mm}$ ($H\times W$) flat rectangular blade, centrally located 15 mm above the base of the container. The rotation speed of the blade was adjustable.

The settling after coagulation took 10 min in all the tests. At the end of settling, a 10 mL supernatant sample was withdrawn with a syringe from 5 mm below the water level, to measure the residual turbidity.

According to a previous report [23], when the concentration of kaolin in simulated wastewater was $100 \text{ mg}\cdot\text{L}^{-1}$ (68 NTU), the optimal PACl dosage during charge neutralization coagulation and sweep flocculation were 2.16 and $54.0 \text{ mg}\cdot\text{L}^{-1}$ of Al, respectively, with corresponding residual turbidities of 0.395 and 0.540 NTU, respectively. In this work, all charge neutralization coagulation and sweep flocculation experiments were carried out under their respective doses in the previous experiments [23], with a final pH of 8.0 and a temperature of $(14.0\pm 1.0)^\circ\text{C}$.

2.2.2 Technology of on-line size monitoring

An on-line size monitor using laser light scattering (Mastersizer 2000, Malvern Co., UK) was applied to monitor floc size during coagulation. The mixer used in the test was the JTY-4 mixer described above. A peristaltic pump (BT00-300M, Baoding Longer Precision Pump Co., Ltd., Baoding, China) was used, with a silicone tube of internal diameter 4.8 mm.

A diagram of the on-line monitoring system is shown in Fig. 1. The kaolin suspension in the coagulation vessel was pumped into the sample cell of the laser particle size analyzer through the silicone tube for floc size measurement. The sample inlet was close to the inner wall of the coagulation vessel and located 34 mm below the water level. The outlet for the return sample from the sample cell was located just above the water level (to remove air bubbles) and was close

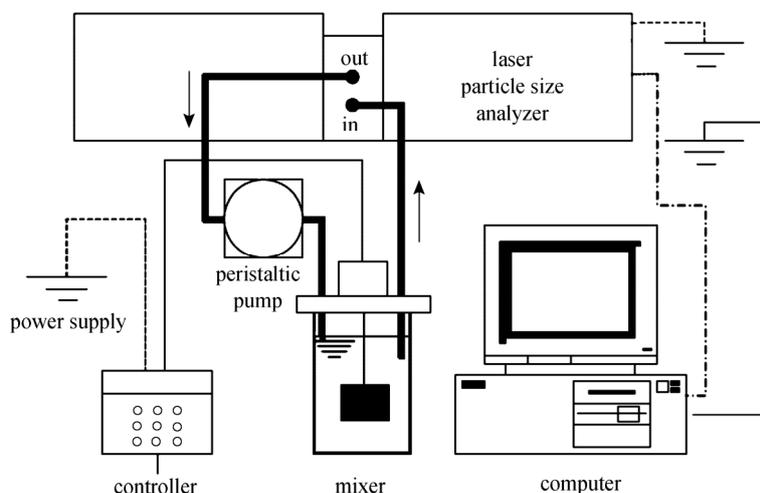


Figure 1 The on-line size monitor with laser light scattering

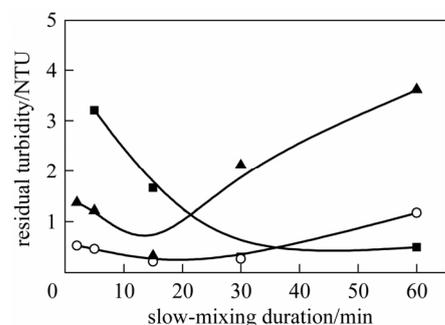
to the inner wall of the coagulation vessel. The sample inlet and the return sample outlet were located at opposite ends (radial direction) of the coagulation vessel. The sample flux was $34 \text{ ml}\cdot\text{min}^{-1}$. A higher flux would result in floc breakage [15], while a lower flux would result in floc settlement in the silicone tube and therefore cause a higher fluctuation of the particle size in the test result. The floc size was measured continuously, while the background measurement was carried out once before each set of experiments. The measurement duration was 30–50 s for the background measurement and 2 or 3 s for the sample measurement. The measurement interval was generally 10 s.

3 RESULTS AND DISCUSSION

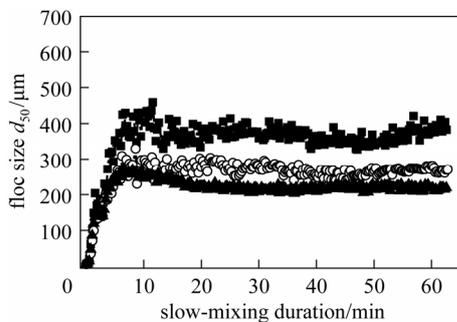
3.1 Effect of slow-mixing on coagulation with PACI

3.1.1 Effect of slow-mixing on charge neutralization coagulation

The effects of slow-mixing on the residual turbidity of charge neutralization coagulation with PACI were investigated at different slow-mixing intensities ($G=4, 15$ and 38 s^{-1}) and slow-mixing durations of 1–60 min (rapid-mixing: $G=244 \text{ s}^{-1}$, $t=3$ min). The results are shown in Fig. 2 (a). When the slow-mixing duration is within a certain range, such as $t<15$ min at $G=15$ or 38 s^{-1} , the residual turbidity decreases with slow-mixing duration. However, when the slow-mixing duration is longer, such as $t>15$ min at $G=15$ or 38 s^{-1} ,



(a) Variation in residual turbidity



(b) Variation in floc size

Figure 2 Effects of slow-mixing on PACI charge neutralization coagulation (temperature: $15.0 \text{ }^{\circ}\text{C}$; kaolin: $100 \text{ mg}\cdot\text{L}^{-1}$; PACI: $2.16 \text{ mg}\cdot\text{L}^{-1}$ of Al; rapid-mixing: $G=244 \text{ s}^{-1}$, $t=3$ min) slow-mixing intensity G/s^{-1} : 4; 15; 38

the residual turbidity increases with mixing duration, even at the G value as low as 4 s^{-1} . Furthermore, the increase in residual turbidity is more obvious when the mixing intensity is higher. It can also be seen that a higher slow-mixing intensity results in a shorter optimal slow-mixing duration. For example, the optimal slow-mixing duration is 15 min at $G=15 \text{ s}^{-1}$, yet it is 30 min at $G=4 \text{ s}^{-1}$. However, the GT value is not constant, as discussed in more detail in the following section.

Figure 2 (a) shows that the optimal slow-mixing intensity during charge neutralization coagulation with PACI is 15 s^{-1} and either higher or lower mixing intensity has a negative effect on flocculation. The variation in floc size during charge neutralization coagulation with PACI is shown in Fig. 2 (b). The floc size decreases with an increase in slow-mixing intensity, which is consistent with the results reported by others [6, 24, 25]. This means that a larger floc size does not imply better coagulation performance in the case of different slow-mixing intensities. By comparing Figs. 2 (a) and 2 (b), it also can be seen that the optimal slow-mixing duration is not simply the time to form the largest mean floc size but is longer, possibly because larger floc have a smaller fractional dimension and therefore a slower settling velocity [26–28].

3.1.2 Effect of slow-mixing on sweep flocculation

The effects of slow-mixing on the residual turbidity during sweep flocculation with PACI were also investigated under different slow-mixing intensities ($G=4, 15$ and 38 s^{-1}) and slow-mixing durations of 1–60 min (rapid-mixing: $G=166 \text{ s}^{-1}$, $t=3$ min). The results are shown in Fig. 3 (a). Similar to charge neutralization coagulation, for short slow-mixing durations, such as $t<10$ min at $G=4 \text{ s}^{-1}$, $t<5$ min at $G=15 \text{ s}^{-1}$ or $t<3$ min at $G=38 \text{ s}^{-1}$, the residual turbidity decreases with time. Extending the mixing duration increases the residual turbidity. The results also show that higher slow-mixing intensities cause more obvious increase in residual turbidity. At the same time, higher slow-mixing intensities reduce the optimal slow-mixing duration. For example, the optimal slow-mixing duration is 10 min at $G=4 \text{ s}^{-1}$ but it is 3 min at $G=38 \text{ s}^{-1}$. However, the GT value also varies, consistent with that during charge neutralization coagulation.

Figure 3 (a) shows that the optimal slow-mixing intensity for sweep flocculation with PACI is 15 s^{-1} . At this slow-mixing intensity, the floc size is close to that at 4 s^{-1} but larger than that at 38 s^{-1} [Fig. 3 (b)]. This is generally consistent with the experimental results during charge neutralization coagulation. Fig. 3 shows that the optimal slow-mixing duration is longer than the time for forming the largest mean floc size.

Based on the above experimental results, neither charge neutralization coagulation nor sweep flocculation could reach their best coagulation performance at overly low or high slow-mixing intensities. Therefore, we conclude that coagulation performance is affected by both slow-mixing duration and intensity. In addition, it can be seen that the optimal coagulation performance

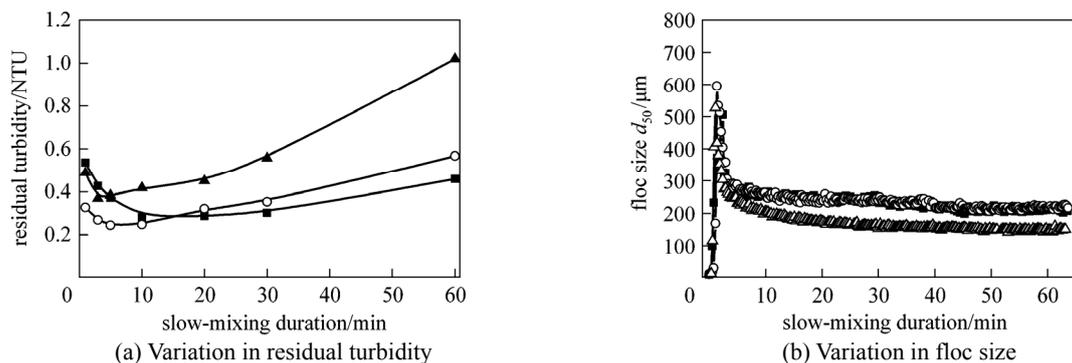


Figure 3 Effects of slow-mixing on sweep flocculation with PACI (temperature: 13.5 °C; kaolin: 100 mg·L⁻¹; PACI: 54.0 mg·L⁻¹ of Al; rapid-mixing: $G = 166 \text{ s}^{-1}$, $t = 3 \text{ min}$); slow-mixing intensity G/s^{-1} : 4; 15; 38

could be obtained only with the optimal slow-mixing duration and the optimal slow-mixing intensity. At a lower slow-mixing intensity, some flocs will settle to the bottom of the coagulation vessel, which would reduce the collision rate between flocs. On the other hand, at a higher slow-mixing intensity, the flocs will breakup, which has a negative effect on the formation of large, compact flocs. Therefore, if the slow-mixing intensity is not optimal, it is difficult to obtain the best coagulation performance, even when the coagulation time is optimal. Taking Fig. 2 (a) (charge neutralization coagulation) as an example, at the optimal slow-mixing intensity of 15 s^{-1} , the best coagulation performance is obtained at the optimal mixing duration of 15 min, with a residual turbidity of 0.214 NTU. At a lower mixing intensity of 4 s^{-1} , the best coagulation performance is obtained at an optimal mixing duration of 60 min, with a residual turbidity of 0.496 NTU. At a higher slow-mixing intensity of 38 s^{-1} , the best coagulation performance is obtained at an optimal mixing duration of 15 min, with a residual turbidity of 0.323 NTU. Fig. 3 shows the same behavior during sweep flocculation. This indicates that during slow-mixing, intensity is more important than mixing duration. It is also found that slow-mixing intensity has a greater positive effect on charge neutralization coagulation than on sweep flocculation with PACI. Furthermore, at the same slow-mixing intensity, charge neutralization coagulation requires a longer mixing duration than sweep flocculation. It is easy to understand this, based on Eq. (1).

Based on Figs. 2 (a) and 3 (a), the effect of G on the optimal GT value for slow-mixing can be obtained (Fig. 4). The optimal GT value increases with G . For charge neutralization coagulation, the optimal GT values at $G = 4, 15$ and 38 s^{-1} are 7200, 13500 and 34200, respectively. For sweep flocculation, the optimal GT values at $G = 4, 15$ and 38 s^{-1} are 2400, 4500 and 6840, respectively. All the GT values are smaller than the range recommended by the AWWA [5], except for the GT value at $G = 38 \text{ s}^{-1}$ during charge neutralization coagulation. This seems to imply that PACI requires a smaller GT value than conventional coagulants,

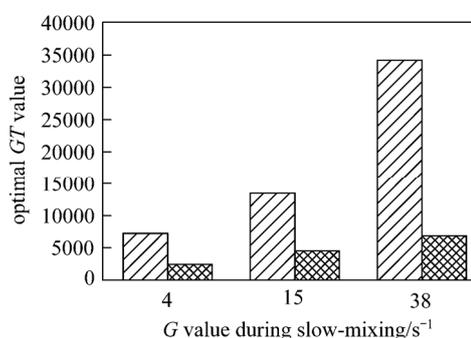


Figure 4 Effect of slow-mixing intensity on the optimal GT value during slow-mixing (temperature: $14.0 \pm 1.0 \text{ }^\circ\text{C}$; kaolin: 100 mg·L⁻¹; PACI: 2.16 mg·L⁻¹ of Al for charge neutralization coagulation and 54.0 mg·L⁻¹ of Al for sweep flocculation) (hatched bars: charge neutralization coagulation; cross-hatched bars: sweep flocculation)

such as aluminum sulfate. The optimal GT value during sweep flocculation is also smaller than that during charge neutralization coagulation for the same wastewater. This can be explained by Eq. (1), which is derived from the ortho-kinetic flocculation formula [29].

$$GT = \frac{\pi}{4\phi} \ln \left(\frac{N_0}{N_t} \right) \quad (1)$$

where N_t and N_0 are the floc number at flocculation time t and the primary particle number, respectively, and ϕ is the particle or floc volume fraction. According to Eq. (1), for a given wastewater, the GT value is constant when the same flocculation rate is obtained under the same flocculation conditions, except the G value and slow-mixing duration. However, this is not the case, because of the effect of floc breakup.

3.2 Compensatory effect of slow-mixing on rapid-mixing

3.2.1 Compensatory effect of slow-mixing during charge neutralization coagulation

The compensatory effect of slow-mixing is shown in Fig. 5 for charge neutralization coagulation

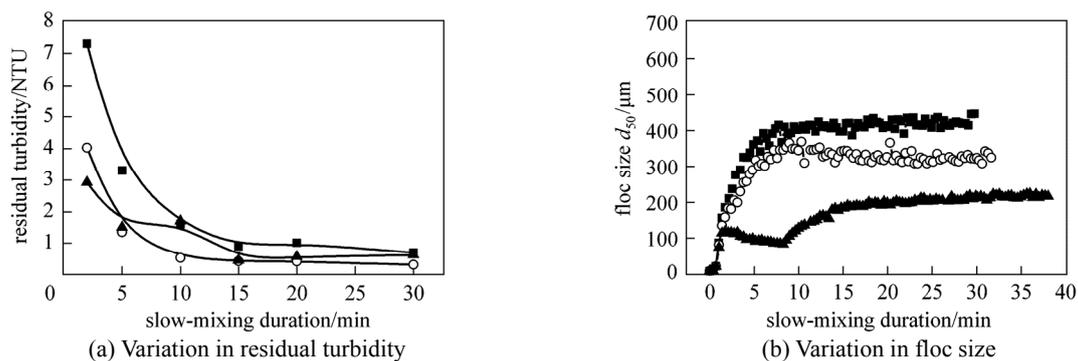


Figure 5 Compensatory effects of slow-mixing on charge neutralization coagulation with PACI (temperature: 15.0 °C; kaolin: 100 mg·L⁻¹; PACI: 2.16 mg·L⁻¹ of Al; slow-mixing intensity: $G = 15 \text{ s}^{-1}$, rapid-mixing intensity: $G = 244 \text{ s}^{-1}$)
 rapid-mixing duration/min: 1; 2; 8

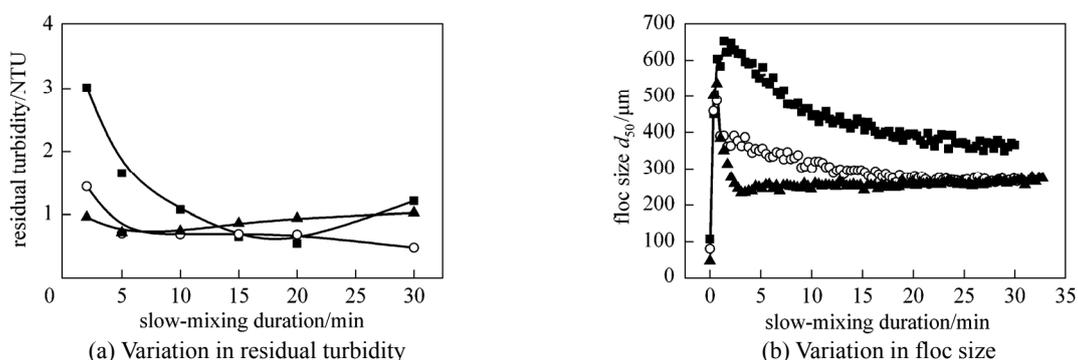


Figure 6 Compensatory effects of slow-mixing on sweep coagulation with PACI (temperature: 13.5 °C; kaolin: 100 mg·L⁻¹; PACI: 54.0 mg·L⁻¹ of Al; slow-mixing intensity: $G = 15 \text{ s}^{-1}$, rapid-mixing intensity: $G = 244 \text{ s}^{-1}$)
 rapid-mixing duration/s: 10; 50; 3 min

in which rapid-mixing is inadequate or excessive, compared to the optimal rapid-mixing time of 2 min at $G = 244 \text{ s}^{-1}$. At any given slow-mixing duration over 5 min, the coagulation performance obtained at the optimal rapid-mixing duration of 2 min ($G = 244 \text{ s}^{-1}$) is clearly superior to that at rapid-mixing durations of 1 or 8 min. After a rapid-mixing duration of 2 min, the residual turbidity of the kaolin suspension decreases to 0.53 NTU at a slow-mixing duration of 10 min, while the residual turbidity of the kaolin suspension with a slow-mixing duration of 20 min after rapid-mixing durations of 1 and 8 min decreases to 1.01 and 0.57 NTU, respectively. This indicates that for charge neutralization coagulation, prolonged slow-mixing could compensate for negative effects of inadequate or excessive rapid-mixing durations. However, the compensation is limited and does not attain the best performance under the optimal conditions for both rapid- and slow-mixing. Fig. 5 (b) shows that the flocs reach their largest size under the condition of 1 min rapid-mixing, while the residual turbidity is not the lowest. The reason for this is mainly the effect of the floc fractal dimension on settling rate [27, 28].

3.2.2 Compensatory effect of slow-mixing during sweep flocculation

Figure 6 shows the effect of slow-mixing when

rapid-mixing is inadequate or excessive during sweep flocculation, where the optimal rapid-mixing duration is 50 s at $G = 244 \text{ s}^{-1}$. This shows that the effects of rapid-mixing duration on residual turbidities are relatively minor.

When the slow-mixing duration was in the range 5-10 min, the coagulation performance at a rapid-mixing duration of 3 min is similar to that at rapid-mixing duration of 50 s, but when the slow-mixing duration is in the range 15-20 min, the coagulation performance at a rapid-mixing duration of 10 s is similar to that at rapid-mixing duration of 50 s. This indicates that, for sweep flocculation, when the rapid-mixing duration is too short, prolonging slow-mixing could compensate to some extent for the negative effects of insufficient rapid-mixing. However, when the rapid-mixing duration is too long, it appears that an appropriately shortened slow-mixing duration could attain superior coagulation performance. Overall, the coagulation performance at the optimal rapid-mixing duration is superior to those at inadequate or excessive rapid-mixing durations.

Figure 6 (b) shows the variation in flocculation size during sweep flocculation. As is the case in charge neutralization coagulation, and for the same reason, the flocs reach their largest sizes at a rapid-mixing duration of 1 min. Furthermore, it can be seen from Figs. 5

(a) and 6 (a) that the coagulation performance is improved with the increase of rapid-mixing duration when the slow-mixing duration is shorter than 5 min. This seems to indicate that prolonging rapid-mixing duration can improve the coagulation performance of PACl when slow-mixing is insufficient.

4 CONCLUSIONS

There is an optimal slow-mixing intensity and duration for coagulation with PACl. Higher or lower slow-mixing intensities and longer or shorter mixing durations produce inferior outcomes but intensity is more important than duration for coagulation with PACl. For a kaolin suspension of $100 \text{ mg}\cdot\text{L}^{-1}$, the optimal G value for slow-mixing is 15 s^{-1} .

The slow-mixing duration required for the optimal coagulation performance decreases with increasing slow-mixing intensity but prolonged mixing duration clearly adversely affects coagulation. The slow-mixing intensity has a more marked positive effect on charge neutralization coagulation than on sweep flocculation and charge neutralization coagulation requires a longer slow-mixing duration. The optimal slow-mixing duration based on residual turbidity is longer than that required for forming the largest mean floc size. For a kaolin suspension of $100 \text{ mg}\cdot\text{L}^{-1}$, the optimal slow-mixing durations at $G = 15 \text{ s}^{-1}$ are 15 min and 5 min for charge neutralization coagulation and sweep flocculation, respectively. The optimal GT value for slow-mixing in charge neutralization coagulation (13500) is larger than that in sweep flocculation (4500). It is also found that the optimal GT value increases with G .

Appropriate prolonging of the slow-mixing duration could improve coagulation performance in the case with inadequate or excessive rapid-mixing durations for charge neutralization coagulation. However, in the case of excessive rapid-mixing during sweep flocculation, shortening of the slow-mixing duration would be more favorable. The compensation by slow-mixing for inadequate or excessive rapid-mixing is, however, limited and the best coagulation performance could be obtained only under optimal mixing conditions.

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