

## Coagulation of high turbidity water: the effects of rapid mixing

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### ABSTRACT

High turbidity water during the rainy season has long been a challenge to the waterworks in Taiwan. Coagulation is the critical process for solving this problem; however, the dispersion of coagulants in the concentrated solid phase is very difficult. In this study, the effects of rapid mixing on the PACl coagulation of high turbidity water were investigated in terms of turbidity and particle count. Operational parameters of coagulant dosage, coagulant concentration and mixing intensity in the rapid-mixing step exhibited great impacts on the residual turbidity and particle count of the simulated system. Aggregation degree, as evaluated by the Photometric Dispersion Analyser (PDA), was influenced by the intensity of rapid mixing. Poor rapid mixing failed to induce aggregation to effectively coagulate the small and hard to settle particles.

**Key words** | aggregation, coagulation, flocculation, particle count, turbidity, water treatment

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### INTRODUCTION

In Taiwan, river water is the main source of fresh water. Many river waters contain high concentrations of suspended solids because of over-development in agriculture and industry. The Feng-Yuan water treatment plant, located in mid-Taiwan, is one example. In dry seasons, approximately 70% of the year, the turbidity of the raw water is maintained below 20 NTU. However, the turbidity can rise from 20 NTU to hundreds or even thousands of NTU when heavy rainfall occurs. This fluctuation in water turbidity during the rainy season has long been a challenge to the traditional purification process in the Feng-Yuan water facility. The situation has worsened because of serious soil erosion in the watershed after the 921 earthquake, the major earthquake that occurred in Taiwan on 21 September 1999.

Coagulation is the most widely used physicochemical process for particle removal in water treatment facilities. Coagulants are added to transform dispersed particles into aggregates for subsequent removal by sedimentation or filtration. Coagulation can be broken down into three sequential steps: formation of active coagulant species, particle destabilization and collision of destabilized

particles. The instant coagulants such as alum and ferric chloride are added to water and a series of hydrolysis reactions takes place; these generate active coagulant species including the monomer, polymer and precipitate within an extremely short period of time (Amirtharajah & Mills 1982). It is necessary to transfer the active coagulant species onto the turbid particles to achieve effective coagulation. Many researchers suggested that extremely fast alum dispersion by rapid mixing is crucial (Amirtharajah & Mills 1982; Kawamura 2000). The dispersion of coagulants in the concentrated solid phase is very difficult. In a study of highly turbid water treatment and sludge conditioning, researchers concluded that rapid mixing is critical for performance (Langer and Klute 1993; Tseng *et al.* 1998). They found that the rates of polymer adsorption and floc formation are much higher in increased particle contact, and the problem of local overdosing was more profound than in the dilute suspension.

PACl is the coagulant commonly used in water treatment facilities in Taiwan. The preparation of PACl is essentially a conditional and a controlled hydrolysis of an aluminium chloride solution. Studies (Yao 1987;

Van Benschoten & Edzwald 1990) on the use of PACl indicate that the polymeric species is the most active species, consisting mostly of  $Al_{13}$  and other ionic complexes containing 6 to 13 aluminium atoms. As indicated by the study (Van Benschoten & Edzwald 1990), the Al polymers were stable within the time scales typically found in water treatments. Therefore, PACl is more effective than other easily hydrolysed coagulants, such as alum (Pouillot & Suty 1992; Kadeing *et al.* 1992). It has been suggested that PACl coagulation can be attributed to charge neutralization and bridging, similar to polymer coagulation (Yao 1987). Since the adsorption of poly-electrolyte is not reversible, insufficient dispersion of PACl species in highly turbid water will hamper local overdosing in the mixing tank.

Different rapid mixing resulting in various levels of aggregation have been reported by many researchers (Lettermann *et al.* 1973; Mhaisalkar *et al.* 1991). An optical instrument, Photometric Dispersion Analyser (PDA), was employed to monitor the dynamics of particle aggregation in this study. Gregory was the first researcher to introduce PDA in monitoring the state of aggregation of flocculation (Gregory 1985). The optical technique was applied to describe the dynamics of flocculation (Ching *et al.* 1994). Recently, our laboratory has proved that the ratio output of PDA can accurately reflect the state of aggregation, which offers a useful index for determining the optimal coagulant dosage (Huang & Chen 1996; Kan & Huang 1998). In this study, the object was to investigate the roles of some operational parameters of rapid mixing including coagulant dosage, coagulant concentration, dosing site, mixer type and mixing intensity when raw water with high turbidity is treated. Batch trials were performed to evaluate the effects of various rapid-mixing conditions in terms of turbidity and particle count. The effects of rapid-mixing intensity on the flocculation were investigated by PDA measurement.

## MATERIALS AND METHODS

A synthetic stock suspension of clay was prepared by dispersing reagent-grade clay (Merck, Reagent-grade) in

the reverse osmosis (R.O.) water. Test suspensions were prepared by diluting the clay to  $250 \text{ mg l}^{-1}$ , for which the turbidity was measured as 180 NTU. The conductivity of the water sample was adjusted to  $250 \mu\text{S}$  with  $\text{NaClO}_4$  solution (Merck, Reagent-grade) and the alkalinity was adjusted to  $100 \text{ mg l}^{-1}$  as  $\text{CaCO}_3$  by adding  $\text{NaHCO}_3$  (Merck, Reagent-grade). The particle size distribution of the synthetic water was analysed by a particle size analyser (PS-230, Coulter). The mean particle size was determined as  $4.5 \mu\text{m}$ .

The PACl was produced by Showa Chemicals Incorporation (Reagent-grade, Japan). The ligand ratio,  $r$ , of the stock PACl solution was equal to 2 and the concentration was 10% by weight as  $\text{Al}_2\text{O}_3$ . The PACl coagulant working solution was freshly prepared from the stock solution for each test. It was prepared by dissolving to  $1,000 \text{ mg l}^{-1}$  as Al with deionized (D.I.) water. All the dosages reported in this study are in  $\text{mg l}^{-1}$  as Al.

Standard jar tests were conducted at a controlled temperature of  $25^\circ\text{C}$ . The mixing device was a square acrylic vessel ( $L \times W \times H$ ,  $11.5 \text{ cm} \times 11.5 \text{ cm} \times 21 \text{ cm}$ ) which is often referred to as a gator jar. The mixing was provided by a  $76 \text{ mm} \times 25 \text{ mm}$  flat rectangular blade centrally located in the vessel. The blade was driven by a thin spindle via a motor of adjustable speed from 10 to 300 rpm (PB-700 Jar tester, Phipps & Bird Inc.). We adopted 20-minute slow mixing at 30 rpm ( $G = 25 \text{ s}^{-1}$ ) for each batch test. The velocity gradient was adopted directly from the study by Cornwell and Bishop (1983), in which the same mixer and vessel were used. The PACl was injected above the liquid surface, which was similar to the operation practised in Taiwanese water works.

Various rapid-mixing parameters were evaluated in this study. The experimental conditions were the same as the jar tests mentioned above except for the tested parameter. The zeta-potential (ZP) of the particles was measured by the Zeta-meter (Zeta-Meter System 3.0, ZETA METER Inc.) After settling for 5, 10 and 20 minutes, the residual turbidity and particle number of the supernatant were measured by turbidimeter (Hach Ratio/XR) and particle counters, respectively. Particle numbers were measured in the size range  $0.2\text{--}15 \mu\text{m}$  by two particle counters. One of the particle counters (WGS267, Met-One Inc., USA) is based on light blockage

theory, which detects the particles ranging from 2 to 15  $\mu\text{m}$ . The other counter (KS28B, Rion Inc., Japan) is based on light scattering theory, and measures particles from 0.2 to 2  $\mu\text{m}$ .

An optical technique, PDA monitoring, was applied to detect the dynamics of the flocculation. This technique, which has been well described by Gregory (1985), utilizes the measurement of fluctuations in the intensities of light transmitted through a moving suspension. The signals, reflecting the scattering condition of the aggregates flowing through the detector, were recorded by a detector and their root mean square (rms) calculated ( $V_{\text{rms}}$ ). From the  $V_{\text{rms}}$  and the mean transmitted light intensity (dc), the state of aggregation in the suspension was quantified using the ratio of  $V_{\text{rms}}$  to dc, as shown in Equation (1),

$$\text{Ratio} = \frac{V_{\text{rms}}}{\text{dc}} = \left( \frac{NL}{A} \right)^{\frac{1}{2}} C \quad (1)$$

where  $N$  is the number concentration,  $L$  is the length of the optical path,  $A$  is the effective cross-sectional area of the light beam, and  $C$  is the cross-section of the scattering light.

The PDA monitoring system comprised a mixing device, tube, recycling pump and PDA as indicated by Gregory & Rossi (2000). A 0.3-m Tygon tube with 3-mm i.d. conveyed the suspension from the jar vessel to the flow-through detector of the PDA within 7 s. The flow of the suspension was maintained by a peristaltic pump located downstream of the PDA, and the suspension was recycled back to the mixing vessel. The ratio output from the PDA detector was continuously monitored every 10 s. To set up the system, the distilled water was first run through the PDA, and the DC output was adjusted to 10 V. The ratio output was then set at 0.5 by conveying the sample through the detector.

## RESULTS AND DISCUSSION

### Effect of coagulant dosage

Jar tests with several PACl dosages, ranging from 0 to 10  $\text{mg l}^{-1}$  as Al, were performed on synthetic water to

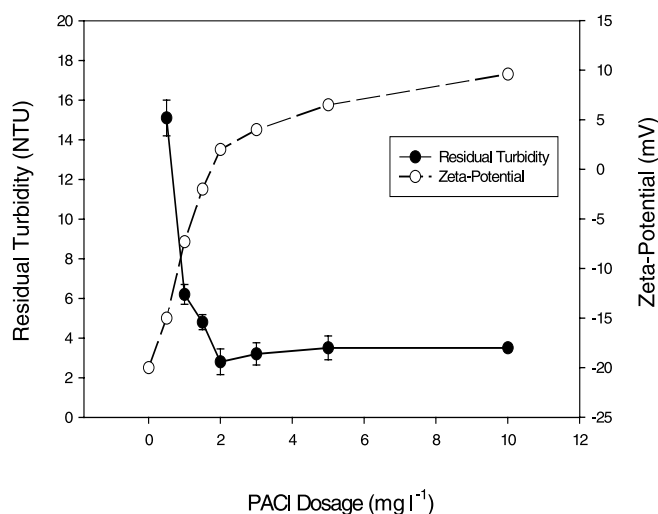


Figure 1 | Effect of PACl dosage on residual turbidity and zeta potential of the synthetic sample.

evaluate coagulant dosing. The optimum dosage was evaluated by residual turbidity as well as zeta potential. As shown in Figure 1, the turbidity was reduced with increasing PACl dosage, and then reached stabilization at a dosage considered to be the optimal coagulant dosage. The original zeta potential of the synthetic water sample was  $-20$  mV. Zeta potentials increased with the increasing coagulant concentration. At the optimal dosage determined by the lowest residual turbidity, the water sample exhibited near zero ZP, which indicates that the coagulation mechanism with PACl is charge neutralization as PACl contains mostly polymeric aluminium salts of high charge valency.

Although charge reversal occurred at dosages over the optimal, the corresponding residual turbidity did not increase. This can be explained by the theory proposed by Dentel that there are sites on the colloidal surfaces which remain adsorbable by the polymeric aluminium (Dentel 1991). Through the 'mosaic effect', these charge-reversed particles can further combine to form flocs through a mechanism similar to polymer bridging (Yao 1987).

The optimal coagulant dosage determined by ZP and residual turbidity measurement was  $2 \text{ mg l}^{-1}$ . In the subsequent tests, a slightly insufficient dosage was chosen to

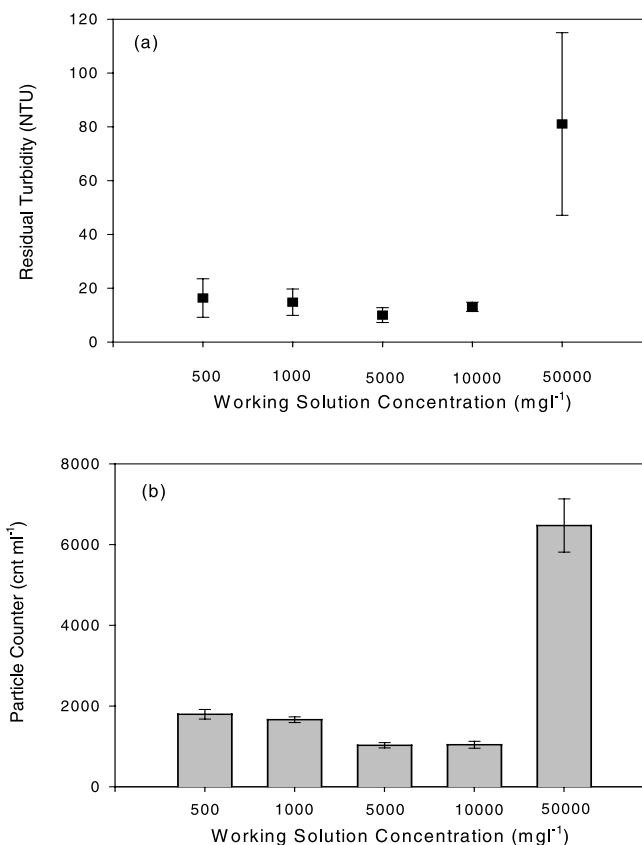
amplify the effect of rapid mixing on the highly turbid water coagulation. Dosages slightly lower than the optimal dosage, i.e.  $1 \text{ mg l}^{-1}$  and  $0.5 \text{ mg l}^{-1}$ , were chosen. Furthermore, because of the high pH buffer capacity contributed by the abundant alkalinity, the pH was maintained within a range of 0.3 units throughout the experiments.

### Effect of coagulant concentration

Five concentrations of PACl working solutions, 50,000, 10,000, 5,000, 1000 and 500  $\text{mg l}^{-1}$ , were prepared from the stock solution. Various volumes of the working solution, 0.04, 0.2, 0.4, 2 and 4 ml were added into the 2 l sample to make a dosage of  $1 \text{ mg l}^{-1}$ . The pH values of the working solutions were all less than 4. Working solutions were effective for only 2 h after preparation. During these effective times, Al composition in the PACl solution was assumed to be stable before dosing. In this experiment, the coagulant was injected at the sample surface. The rate of injection was maintained at  $1 \text{ ml s}^{-1}$ ; therefore, all injections could be finished within 4 s. The mixing intensity of rapid mixing was controlled at  $25 \text{ s}^{-1}$ . After the slow mixing and settling, the residual turbidity and particle count in the size range of 2–15  $\mu\text{m}$  were measured.

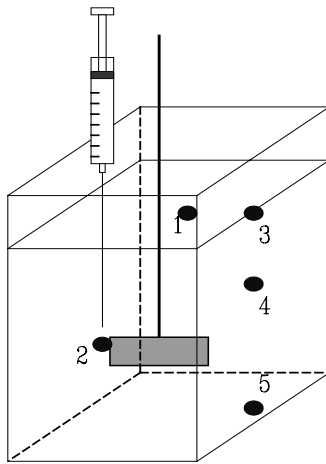
The average residual turbidities and particle counts from the dosing by PACl with five concentrations are shown in Figure 2. The results indicate that residual turbidities were in the range of 15–20 NTU and particle counts were about  $1,800 \text{ cnt ml}^{-1}$  in the supernatant for the trials with 500 and 1,000  $\text{mg l}^{-1}$  coagulant concentrations. For the 5,000 and 10,000  $\text{mg l}^{-1}$  coagulant concentrations, the results were 10–15 NTU and  $1,100 \text{ cnt ml}^{-1}$ . The highest turbidity and particle counts appeared with the 50,000  $\text{mg l}^{-1}$  coagulant concentration. The evidence showed that dilution of coagulant during rapid mixing was an important factor affecting coagulation performance. It also recommended that dilution factors of 5–10 are appropriate.

PACl is a prehydrolysed inorganic coagulant. The extent of prehydrolysis of pure PACl is described in terms of the ligand ratio  $r$ , which is the molar ratio of hydroxide ( $[\text{OH}^-]$ ) to the total Al present in solution ( $[\text{Al}_{\text{total}}]$ ) (Yao



**Figure 2** | Average residual turbidity (a), and average counts of particles (b) in the size range 2–15  $\mu\text{m}$  for each coagulant concentration at  $1 \text{ mg l}^{-1}$  PACl dosage.

1987). At adequate  $r$  ( $1.25 < r < 2.5$ ), a polymeric species,  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}(\text{H}_2\text{O})_{12}^{+7}$  or  $\text{Al}_{13}$  polymer, is formed. When the  $r$  is greater than 2.5, aluminium hydroxide solids are produced. Therefore, the pH can be used as an index for the chemical composition of PACl in the working solution.  $\text{Al}_{13}$  contains high positive charge, which dictates the behaviour of the PACl coagulation. As a result, charge neutralization is the principal mechanism for the coagulation. The performance of the PACl coagulant species can be improved by enhanced diffusion via dilution as long as the hydrolysis of  $\text{Al}_{13}$  does not occur. Over-dilution will drive the  $\text{Al}_{13}$  into  $\text{Al}(\text{OH})_{3(\text{s})}$  and reduce the coagulation efficiency of the PACl. The dilution factors of 5–10 are appropriate because the pH of the PACl working solution remains stable after the dilution. The pH of the working solution rises slightly when the dilution factor is more than 10.



**Figure 3** | Schematic representation of coagulant injection site in the mixing vessel.

### Effect of dosing site

The effects of dosing site on coagulation performance were investigated. Five locations within the vessel were selected based on the suggestion by Clark & Srivastava (1993), as indicated in Figure 3. Site 1 is at the liquid surface, which is the location of least turbulence. Site 2 is near the tip of the mixer, which experiences the maximum turbulence. Sites 3, 4 and 5 have median turbulent inten-

sity. PACl was injected directly into the selected spot with a syringe. The long and thin syringe was used to minimize the impact on the flow in the vessel. The PACl concentration of  $1,000 \text{ mg l}^{-1}$  was used for injection. Several coagulant dosages and mixing conditions were used and the residual turbidity and particle count after 20 min of sedimentation were measured. The experimental parameters and results are shown in Table 1. No apparent difference among the dosing sites was observed.

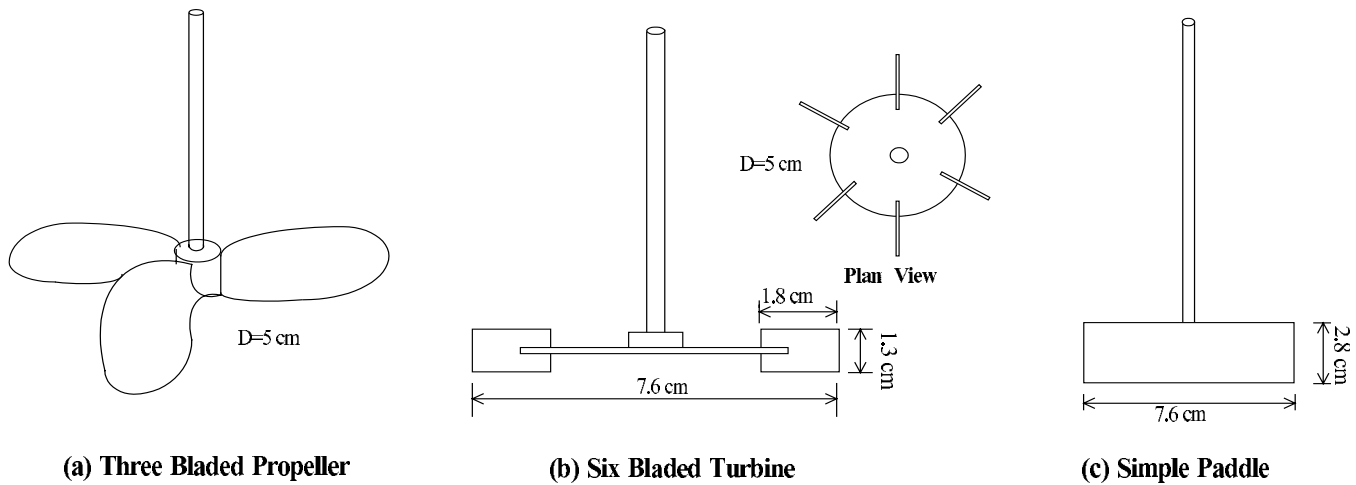
### Effect of mixer types

The geometric shape of the mixers might affect the efficiency of mixing. Three types of mechanical mixers were chosen in this study. They are the 'three-blade propeller', 'six-blade disk turbine' and 'simple paddle', which were the most commonly used mixers for coagulation processes in water treatment facilities. The configuration and dimension of these mixers are shown in Figure 4. The dimensions of the square agitator vessel were the same as those used in the jar test. The conversion factors between the rotation velocity of the mixer and the gradient velocity were given by AWWA (1992). The rotation velocities were 300 rpm, 100 rpm and 120 rpm for the three-blade propeller, disk turbine and paddle, respectively. The rotation

**Table 1** | Residual turbidities and particle counts for various coagulant injection sites

Test no.	Experimental conditions			Residual turbidity (NTU)					*Particle count (cnt/ml)				
	Coagulant dosage ( $\text{mg l}^{-1}$ )	Rapid mixing (G)	Slow mixing (Gt)	1	2	3	4	5	1	2	3	4	5
A	0.5	25	30,000	14.2	14.0	—	—	—	2,456	3,060	—	—	—
B	0.5	25	12,000	29	35	31	31	24	—	—	—	—	—
C	0.5	25	9,000	30	28	—	—	28	—	—	—	—	—
D	1	25	30,000	10.6	10.3	—	—	—	1,344	1,050	—	—	—
E	1	350	30,000	4.2	4.8	4.6	—	5.5	581	584	615	—	714
F	5	350	30,000	0.47	0.57	—	0.45	0.55	176	153	—	113	147

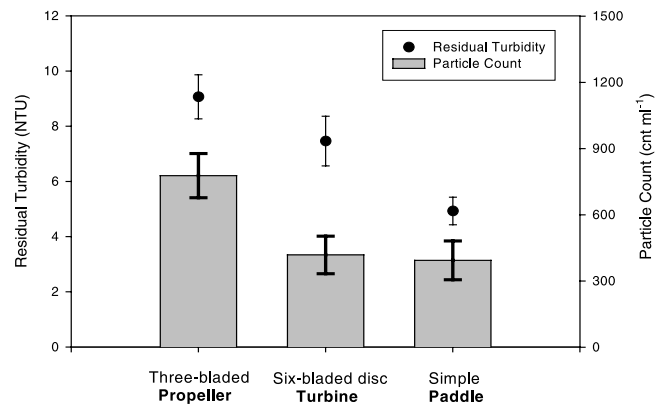
\*Particles in the size range 2–15  $\mu\text{m}$ .



**Figure 4** | The shape and dimension of three-bladed propeller (a), six-bladed turbine (b) and simple paddle (c).

velocity of each mixer was varied in order to obtain the same  $G$  value,  $160 \text{ s}^{-1}$ . In this series of tests, the rapid mixing retention time was 1 minute for all three mixers, with a PACl dosage of  $1 \text{ mg l}^{-1}$ . The simple paddle was used for the slow mixing process. The position of the agitators was fixed in the vessel in all trials.

The results of average residual turbidity and particle count for each mixer are shown in Figure 5. The average residual turbidity was  $9.1 \text{ NTU}$  and particle number was  $780 \text{ cnt ml}^{-1}$  for the propeller,  $7.5 \text{ NTU}$  and  $390 \text{ cnt ml}^{-1}$  for the turbine, and  $5.0 \text{ NTU}$  and  $280 \text{ cnt ml}^{-1}$  for the paddle, which showed the best results. Although all flows were turbulent, the flow patterns were different in each mixing device. Propeller agitators drive the liquid straight down to the bottom of the tank, from where the stream spreads radially in all directions towards the wall, flows upwards along the wall, and returns to the suction of the propeller at the top. Turbine impellers drive the liquid radially against the wall, from where the stream divides, one portion flowing downwards to the bottom and back to the centre of the impeller from below, and the other flowing upwards toward the surface and back to the impeller from above. The flow pattern of the paddle mixing is similar to that of the turbine. Furthermore, the simple paddle has the highest  $D/T$  ratio (the ratio of agitator diameter to vessel width) and the largest projectional cross-section area of these three agitators. Therefore, it

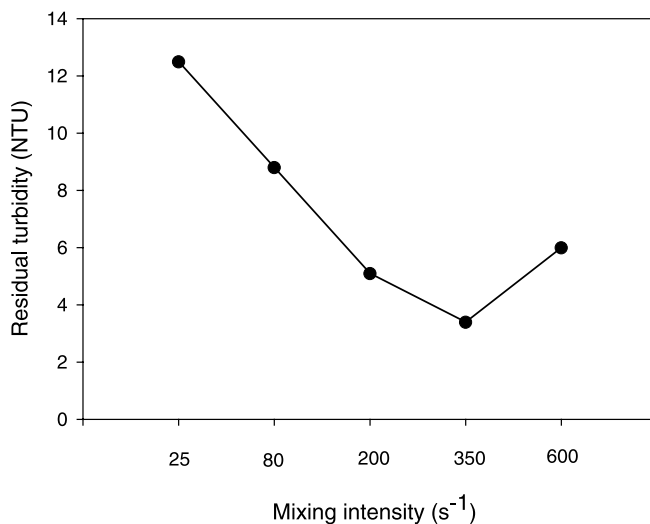


**Figure 5** | Average residual turbidities and average counts of particles ranging from 2 to  $15 \mu\text{m}$  for each mixer at  $1 \text{ mg l}^{-1}$  PACl dosage.

can create the largest turbulent zone and the highest mixing efficiency. The result in Figure 5 indicates that mixing condition is important in the coagulation of high turbidity water by PACl.

### Effect of mixing intensity

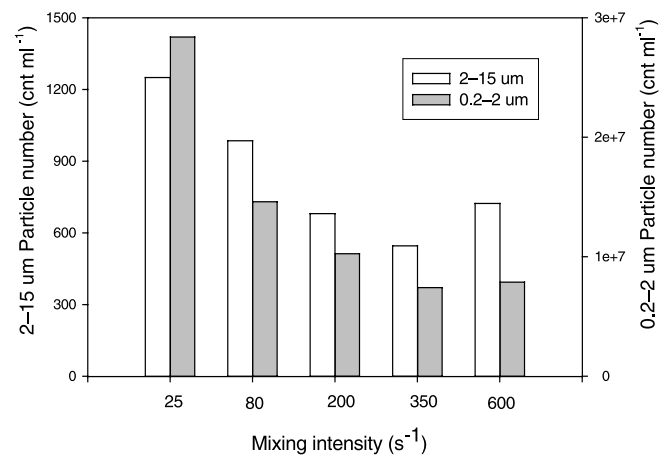
Five intensities of rapid mixing with the simple paddle, 25, 80, 200, 350 and  $600 \text{ s}^{-1}$ , were used in each test. PACl at a dosage of  $1 \text{ mg l}^{-1}$  was injected into the water sample. The resulting residual turbidities are shown in Figure 6.



**Figure 6** | Average residual turbidity for various mixing intensities at  $1 \text{ mg l}^{-1}$  PACl dosage.

The highest turbidity, 12.5 NTU, was discovered at the mixing intensity of  $25 \text{ s}^{-1}$ . Turbidity decreased with increasing mixing intensity, and reached the lowest point, 3.4 NTU, at  $G = 350 \text{ s}^{-1}$ . Stronger mixing resulted in the rebound of the residual turbidity.

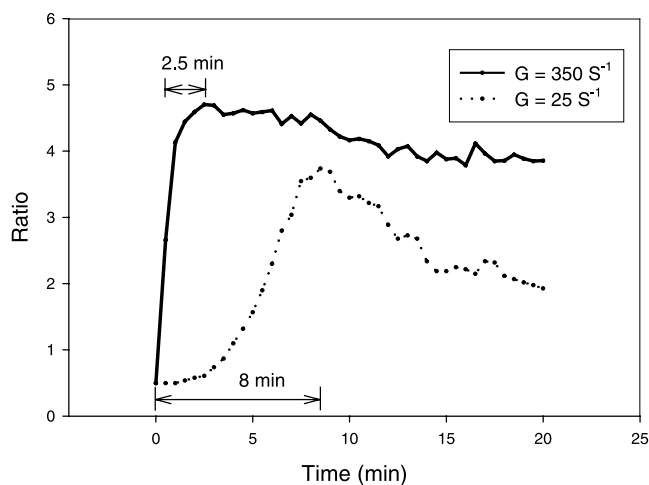
The number of particles remaining in the supernatant was measured in two size categories. One was from 0.2 to  $2 \mu\text{m}$  and the other from 2 to  $15 \mu\text{m}$ . The results are shown in Figure 7. In the range 2–15  $\mu\text{m}$ ,  $G = 25 \text{ s}^{-1}$  resulted in the highest particle count,  $1,250 \text{ cnt ml}^{-1}$ . The lowest particle number,  $580 \text{ cnt ml}^{-1}$ , was found at  $G = 350 \text{ s}^{-1}$ . The highest particle count at  $25 \text{ s}^{-1}$  was twice the lowest one. A more dramatic change was discovered in the size range 0.2–2  $\mu\text{m}$ . The highest particle count at  $25 \text{ s}^{-1}$  was four times the lowest one at  $350 \text{ s}^{-1}$ . Since no significant difference was observed at  $G$  between 200 and  $600 \text{ s}^{-1}$  for both size ranges, we define a term of ‘good mixing’ for the mixing intensity beyond  $200 \text{ s}^{-1}$ , and ‘poor mixing’ for less than  $200 \text{ s}^{-1}$  in the following discussion. From the analysis of the residual turbidity and particle count, a phenomenon was observed. The particles contributed to the high residual turbidity from poor mixing were mainly in the size range 0.2–2  $\mu\text{m}$ . Particles in this size range are the most difficult to remove from the downstream filtration process in water treatment (Yao *et al.* 1971). Good mixing resulted in fewer



**Figure 7** | Average particle counts of supernatants in the range 0.2–2 and 2–15  $\mu\text{m}$  for various mixing intensities at  $1 \text{ mg l}^{-1}$  PACl dosage.

particles in the supernatant than poor mixing. This suggests that more fine particles were transformed to aggregates in the good mixing condition, owing to the different aggregations for the two types of mixing. Therefore, the dynamics of particle aggregation at different rapid-mixing intensity are studied in the next section.

In this study,  $G = 350 \text{ s}^{-1}$  and  $25 \text{ s}^{-1}$  were selected to simulate the good and poor mixing conditions, and the state of aggregation was presented with the PDA measurement. The ratio outputs of both trials were adjusted to 0.5 at the outset. The profiles of ratio output versus time, at coagulant dosage of  $1 \text{ mg l}^{-1}$ , are shown in Figure 8. For the good mixing condition, the ratio output reached a maximum value of 4.71 at 2.5 min. For the poor mixing condition, the ratio output reached a maximum value of 3.74 at 8 min. After the maximum value, they both tailed off. Li and Gregory (1991) suggested that the decrease in the ratio value results from the settling of large aggregate (flocs) during the flocculation process. At low mixing intensity during flocculation, it is difficult to keep the large flocs suspended in the water, and a significant number of these flocs settled. Since the ratio value is proportional to the square root of the concentration of suspended particles, a lower value is reached when the flocs begin to settle. The time required to reach the maximum ratio output can be viewed as the time required for particles to aggregate into a ‘settleable’ floc.



**Figure 8** | Profiles of PDA ratio output over a period of 20 minutes for mixing intensity of  $G=350\text{ s}^{-1}$  and  $25\text{ s}^{-1}$  at  $1\text{ mg l}^{-1}$  PACl dosage.

As mentioned above, the time for the particles to aggregate into ‘settleeable’ flocs was 2.5 min for  $G = 350\text{ s}^{-1}$  and 8 min for  $G = 25\text{ s}^{-1}$ . The steeper increase in ratio output means a faster aggregation. The difference between the two conditions can be explained by the collision theory proposed first by Smoluchowski (Thomas et al. 1999). Camp and Stein modified Smoluchowski’s theory, as shown in Equation (2):

$$-\frac{dn_1}{dt} = \frac{1}{6} G \alpha (d_1 + d_2)^3 n_1 n_2 \quad (2)$$

In this formula,  $n_1$  is the number of primary particles with diameter  $d_1$ ,  $n_2$  is the number of other particles with diameter  $d_2$ ,  $G$  is the mean velocity gradient in the fluid, and  $\alpha$  is the collision efficiency coefficient. In this article,  $n_2$  represents the particle number initially destabilized by the adsorption of PACl coagulant species. Judged from the difference in aggregation velocity, it is obvious that the  $n_2$  for  $G = 350\text{ s}^{-1}$  was much larger than that for  $G = 25\text{ s}^{-1}$ .

## CONCLUSION

In this study, the coagulant dosage, coagulant concentration and mixing intensity all exhibited considerable

impacts on the residual turbidity and particle count. However, dosing site and mixer type showed insignificant effect. The state of flocculation was also influenced by rapid-mixing intensity. The key factor in flocculation is the number of destabilized particles in the initial period, which is determined by the dispersion of PACl during the rapid mixing. Poor rapid mixing cannot induce aggregation to effectively coagulate the smaller and hard-to-settle particles. Small particles remain as stable dispersion even after long-term settling. We also found that the characteristics of flocs formed from the flocculation by various rapid-mixing conditions were quite different. In the future, studies such as size distribution and fractal dimension analysis of flocs need to be pursued.

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