



Enhanced particle destabilization and aggregation by flash-mixing coagulation for drinking water treatment



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ABSTRACT

In water treatment plants (WTPs), the operation of rapid mixing in coagulation units dominates particle destabilization and the formation of flocs. The goal of this study was to investigate the effect of enhanced rapid-mixing intensity (velocity gradient [G]) on the performance of coagulation for natural turbid water treatment. Two surface-water samples of low and high turbidity were coagulated by using polyaluminum chloride (PACl), with G values ranging from 200 to 800 s^{-1} , and the destabilization of particles and dissolved organic matter (DOM) was determined, followed by measurement of filterability of the supernatant. Furthermore, the effect of high rapid mixing on the coagulation efficiency was studied by using a pilot-scale, in-line instantaneous flash mixer (IFM) which could generate much higher G than a conventional mechanical mixing of WTP. The results of Jar test showed that the enhanced reduction in turbidity was accompanied by the improved filterability of the supernatant, which was due to the formation of larger flocs with strong structure in response to higher rapid-mixing intensity, especially for low-turbidity water. It was found that the fractal dimension (D_f) of coagulated flocs is inversely proportional to the recovery factor of broken flocs. The in-line IFM with intensive mixing ($G > 5000 s^{-1}$) substantially improved the removal of particles and DOM, as a result of the formation of large flocs.

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1. Introduction

Coagulation processes are important in solid–liquid separation in water treatment practice. In principle, a suitable coagulant, such as aluminum or iron salts, must be added during intense agitation to rapidly disperse hydrolyzed coagulant species to particles, followed by a period of slower mixing to form flocs. Usually the velocity gradient (G) is calculated to evaluate the intensity of mixing in a turbulent flow system [1].

Mixing conditions have a significant effect on the performance of coagulation. To achieve particle aggregation in coagulation, the particles need to collide to form flocs. This process can be greatly improved by agitation [2]. Generally, slow-mixing is crucial to form flocs by the cohesion between primary particles during flocculation. Many studies have suggested that the intensity of gentle agitation or mixing dictates the size and structure of flocs during floc growth, as well as aggregate breakage and restructuring [3–5]. However, the initial mixing conditions are the most critical in the coagulation process, because rapid and uniform dispersion of metal coagulants benefits the destabilization of particles [6]. By contrast, poor rapid-mixing can lead to local overdosing and resta-

bilization of particles. Studies have indicated that coagulation kinetics is mainly governed by the initial rapid-mixing process that strongly affects the models of floc aggregation and breakage [7–9].

It is well known that mechanical mixing with intensive agitation can bring about instantaneous and uniform dispersion of coagulant to improve the efficiency of particle removal after coagulation/sedimentation [10,11]. To reduce the use of coagulant, an in-line hydraulic jet and flash mixer have been designed as a rapid-mixing unit to produce high mixing intensity in a turbulent flow, which facilitates collision efficiency between colloids in the initial aggregation process, and subsequent flocculation efficacy [12,13]. Although many researchers have observed the enhanced coagulation performance of colloidal particles or organic matter at appropriate rapid-mixing conditions [14–16], they only examined the effect of rapid-mixing intensity or duration on the coagulation of synthetic raw water in the laboratory. In these studies, they found that optimal rapid-mixing intensity varies with rapid mixing time, coagulant dosage and turbidity of water samples. In addition, few investigations have considered the application of optimal rapid mixing in water treatment practice. In conventional water treatment plants (WTPs), rapid mixing is typically conducted in a basin with a mechanical mixer or by hydraulic jump at a fixed G value less than 1000 s^{-1} . To ensure the quality of the treated water, operators normally apply high concentration of coagulant to remove

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turbidity through sweep flocculation. Although rapid-mixing is not critical in the formation of sweep floc [8], it can effectively promote particle aggregation, resulting in reduced turbidity and organic matter [17]. Thus, it is crucial to understand the effect of enhanced rapid-mixing intensity on coagulation with naturally turbid water in order to improve the coagulation performance of WTP, especially at rapid-mixing intensity much higher than that conducted in conventional WTP.

The aim of this study was to investigate the effect of enhanced rapid-mixing on the coagulation of naturally turbid water at optimal coagulant dosage by means of a laboratory jar test and a pilot-scale in-line instantaneous flash mixing (IFM) test. The removal efficiency in turbidity and dissolved organic matter at various mixing intensities were first evaluated in the laboratory, followed by the corresponding filterability of supernatant after coagulation/sedimentation was measured by a suction time index (STI) test. And, the pilot-scale in-line IFM was established to provide high speed dispersion of the hydrolyzing coagulant to investigate the effect of high rapid-mixing intensity ($G > 5000 \text{ s}^{-1}$) on the efficiency of coagulation/sedimentation.

2. Materials and methods

2.1. Water samples

Two surface-water samples were collected from the Fengyuan Water Treatment Plant in Fengyuan, Taiwan. Characteristics of the raw water samples are given in Table 1. The water sample with turbidity of 200 NTU was designated high-turbidity water, and that with 15 NTU low-turbidity water. The concentration of dissolved organic carbon (DOC) in high-turbidity water was higher than that in low-turbidity water, which may explain why the zeta potential of particles in high-turbidity water was lower than in low-turbidity water. In addition, particles in high-turbidity water were larger than in low-turbidity water. Experiments were conducted immediately after the collection of the raw water samples.

2.2. Coagulants

A commercial PACl product ($\text{Al}_2\text{O}_3 = 10\%$) containing an insignificant amount of sulfate was provided by the Fengyuan WTP as the coagulant for the experiment. Working solutions containing 1000 mg/L as Al were freshly prepared before each experiment. The Al concentration was analyzed by inductively-coupled plasma-atomic emission spectrometry (JY24, Jobin-Yvon Inc., France). The initial composition of PACl was determined by the Ferron method as we reported previously [18]. It contains 54% monomeric Al (Al_a), 35% polymeric Al (Al_b), and 11% colloidal Al (Al_c).

2.3. Coagulation experiments

Standard jar tests were used for the coagulation experiments in the lab. A square acrylic vessel ($11.5 \text{ cm} \times 11.5 \text{ cm} \times 21.0 \text{ cm}$

Table 1
Characteristics of raw water taken from Fengyuan water treatment plant.

Sample	High-turbidity water	Low-turbidity water
pH	7.8	7.8
Temperature ($^{\circ}\text{C}$)	25	26
Turbidity (NTU)	200	15
DOC (mg/L)	2.2	1.3
Alkalinity (mg/L as CaCO_3)	55	69
Conductivity ($\mu\text{S}/\text{cm}$)	192	223
Zeta Potential (mV)	-18.2	-15.3
Particle Size (μm)	2.7	1.9

DOC: dissolved organic matter.

high), commonly known as a gator jar, was used as a mixing device. A flat rectangular blade (76.0 mm long \times 25.0 mm wide) driven by a single thin spindle via a motor was located at the center of the vessel. Rapid mixing was initially carried out from 100 to 300 rpm ($G = 200\text{--}800 \text{ s}^{-1}$) for 15 s, followed by a slow mixing at 30 rpm ($G = 25 \text{ s}^{-1}$) for 10 min. After a 10-min settling period, the supernatant was withdrawn to measure the residual turbidity and DOC. The residual turbidity and DOC of the supernatant was quantified by turbidimeter (2100P, Hach, USA) and TOC analyzer (TOC-5000A, Shimadzu, Japan). The zeta potentials (ZPs) of the suspension were measured via a laser zeta analyzer (Zetasizer nano ZS, Malvern Inc., UK) immediately after the rapid mixing without dilution. The coagulant (Al) dosage is expressed in milligrams per liter. A particle size analyzer (Mastersizer 2000, Malvern, UK) coupled with small-angle laser light scattering (SALLS) was used to observe the aggregation dynamics of particles during coagulation. The properties of flocs – size, fractal dimension, strength, and recovery factor – formed at various G values of rapid mixing in coagulation process were also determined.

2.4. Filterability test

A suction time index (STI) test was first reported by Ives [19] to evaluate the filterability of water samples. The filtration times of 500 mL of supernatant and distilled water were determined separately by use of a suction filtration device (membrane filter with a pore size of $1 \mu\text{m}$) to calculate the STI as follows:

$$\text{STI} = \frac{\text{Suction time of 500 mL of supernatant (s)}}{\text{Suction time of 500 mL of distilled water (s)}} \quad (1)$$

2.5. Floc strength and recovery factor

The flocs formed after slow mixing were further mixed for 15 min at four different mixing intensities – 25, 70, 130 and 350 s^{-1} – to break the flocs. The size of the broken flocs was measured. The slope of the log–log plot of floc size and the applied shear stress represents floc strength constant (γ). Determination of floc strength was as described in the previous study [20]. Higher floc strength constants means that flocs are easier to break by shear forces. After flocs breakage induced by a constant mixing shear force ($G = 350 \text{ s}^{-1}$), slow stirring at 30 rpm was reintroduced for a further 15 min for floc re-growth. Floc recovery factors, which had previously been used to compare the re-growth of flocs in coagulation, were calculated as follows [15]:

$$\text{Recovery factor (\%)} = \frac{(d_3 - d_2)}{(d_1 - d_2)} \times 100 \quad (2)$$

where d_1 is the average floc size of the steady phase before breakage, d_2 is the floc size after the floc breakage period, and d_3 is the floc size after regrowth to the new steady phase. Flocs with larger recovery factor have better re-growth after breakage.

Static light scattering has been extensively employed to evaluate the size and structure of aggregates as well as in the study of coagulation of various suspensions [21]. The scattered light intensity is measured as a function of the magnitude of the scattering vector, Q . The relationship between the scattered intensity from the aggregate, $I(Q)$, and the scattering wave vector, Q , is shown as follow:

$$I(Q) \propto Q^{-D_f} \quad (3)$$

where

$$Q = \frac{4\pi n \sin(\theta/2)}{\lambda} \quad (4)$$

where n is the refractive index of the fluid, λ is the wavelength in the vacuum of the static light used, and θ is the scattering angle. The mass fractal dimension (D_f) can be estimated from the absolute slope of $\log I(Q)$ versus $\log Q$ by fitting a straight line through the fractal regime section of the scattering plot.

2.6. Mixing test

Two mixing techniques were used in this study, including instantaneous flash mixing (IFM) and mechanical mixing. As shown in Fig. 1, the effluents from the in-line IFM and rapid-mixing WTP unit were simultaneously flocculated at a constant slow-mixing intensity by the jar test, followed by the determination of the residual turbidity and DOC after sedimentation. During flocculation, the formation of flocs was monitored by a continuous optical flocculation monitor (photometric dispersion analyzer [PDA] 2000, Rank Brothers Ltd., Cambridge, UK). For dynamic monitoring, a sample from one beaker was circulated through transparent plastic tubing (3 mm id) by means of a peristaltic pump. The pump was located after the PDA instrument to avoid possible effects of floc breakage. The PDA 2000 measures the mean transmitted light intensity (dc value) and the root mean square (rms) value of the fluctuating components. The ratio of rms to dc is a sensitive index of particle aggregation. In this study, the ratio is called the flocculation index (FI). The FI value is highly correlated with mean floc size.

The IFM with an inner diameter of 20 cm has a plug-flow stream with no back mixing or dead zones during complete mixing inside the pipe. It consists of a fill-up pipe in the center of the IFM, a nozzle, deflector plate and raw water inlet-pipe. The deflector plate in the nozzle area was installed to help the coagulant to disperse evenly immediately after collision. The velocity of influent was from 0.1 to 2.0 m/s (V_f). A fill-up pipe served the pressurized water by the mix pump. The velocity of the fill-up water was from 1.2 to

6.0 m/s (V_F). The coagulants were dispersed at various ratios of V_f/V_F to cause high agitation intensity at turbulent flow [13]. The dispersion characteristics in the IFM were investigated by CFDs (computational fluid dynamics) analysis using the Fluent 6.0 software, and the G value obtained by the jet dispersion of the coagulant ranged from 4501 to 22 802 s^{-1} .

3. Results and discussion

3.1. Determination of optimal coagulant dosage

To determine the optimal dosage of PACl coagulation, low- and high-turbidity water was coagulated at various dosages at fixed rapid-mixing intensity and mixing time ($G = 560 s^{-1}$; $t = 15 s$), simulating the rapid-mixing condition of the WTP. The final pH after rapid-mixing is around neutral for each coagulation test. The corresponding zeta potential of particles after rapid mixing and the residual turbidity after sedimentation were determined. The residual turbidity at charge reversal is lower than that at negative potential regardless of the original turbidity, as illustrated in Fig. 2. Optimum coagulation was achieved when zeta potential increased to between +5 mV and +10 mV. Our previous study suggested that the optimal turbidity removal of natural turbidity water by commercial PACl coagulation preferentially occurs under sweep flocculation corresponding to charge reversal [22]. In this study, because the PACl coagulant we used contained more than 50% monomeric Al which hydrolyzes into aluminum hydroxide ($Al(OH)_3$) during coagulation, sweep flocculation dominated coagulation. To further verify the significance of enhanced rapid-mixing in coagulation, the optimal dosage, which was 1.75 mg/L as Al and 2 mg/L as Al at low and high turbidity, respectively, as determined from the results given in Fig. 2, was adopted to evaluate the effect of rapid-mixing on turbidity and DOC destabilization by coagulation in the following experiments.

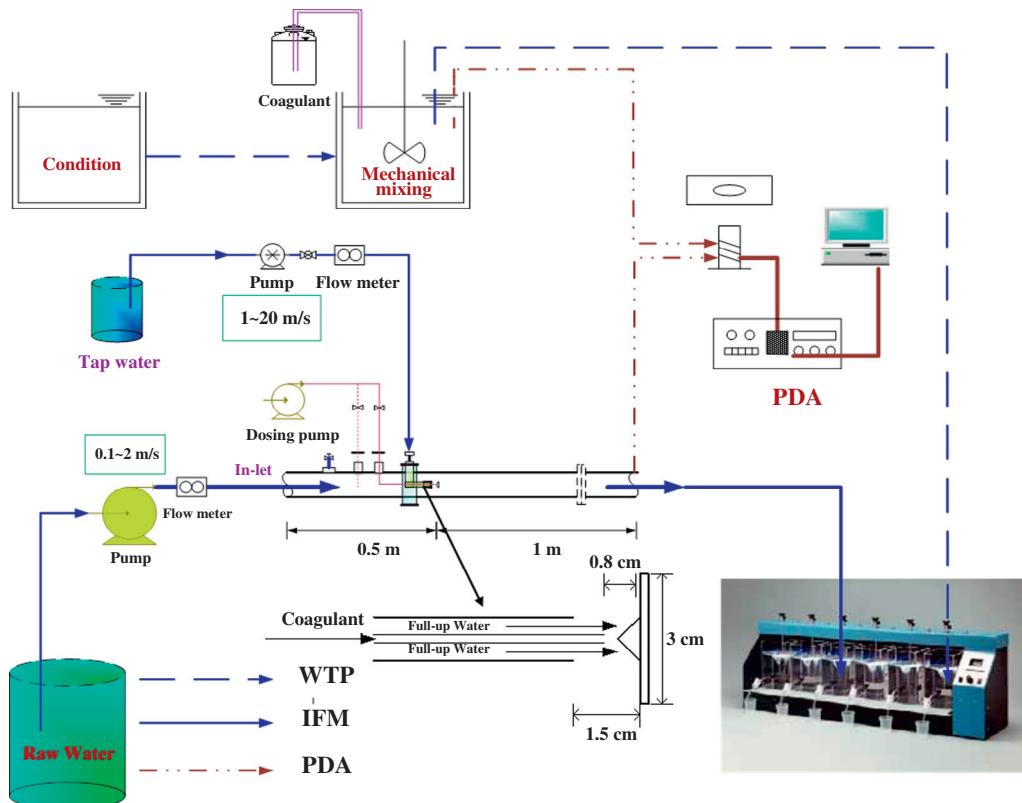


Fig. 1. Schematic diagram of in-line IFM and full-scale mechanical mixing test.

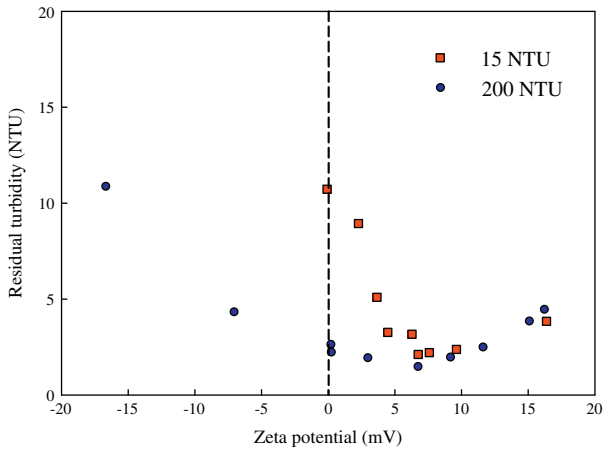


Fig. 2. Variation in residual turbidity with zeta potential after rapid mixing for coagulation of low- and high-turbidity water. The final pH after rapid-mixing is around neutral for each coagulation test.

3.2. Effect of rapid-mixing intensity on coagulation performance

Turbidity removal and filterability can be improved by coagulation with intensive rapid mixing in a mechanical mixing system [10,11]. Our results show that the residual turbidity varied with the velocity gradient of rapid mixing (Fig. 3a). For low turbidity water, the residual turbidity continuously decreased with increasing G -values of rapid mixing, but the residual turbidity only decreased slightly with increasing rapid-mixing intensity for high-turbidity water. This result could be explained by the effect of rapid mixing on particle aggregation. For the low-turbidity water coagulation, increasing rapid-mixing intensity enhanced the collision between particles, by which particles aggregated into larger

micro-flocs, as shown in Fig. 3b. These micro-flocs become larger flocs at the end of flocculation, which results in the reduction of residual turbidity [23]. However, at high turbidity, the collision efficiency of particles is high enough even at low rapid-mixing intensity. Therefore, the effect of rapid-mixing intensity on particle aggregation is not significant for high-turbidity water coagulation. As a result, the flocs do not grow with increased G value, which results in a limited improvement in the reduction of residual turbidity at high rapid-mixing intensity. Furthermore, it is found that the flocs formed at high-turbidity condition are smaller than that formed at low turbidity due to the breakage of flocs formed at high turbidity in the flocculation process. At high turbidity, because the rate of particle aggregation is faster, the larger flocs formed in the initial coagulation process are easily broken at the end of flocculation [20]. The variation in residual turbidity with rapid-mixing intensity also indirectly affects the filterability of supernatant after coagulation/sedimentation. At low turbidity, the decrease in STI value of the supernatant is also more sensitive to rapid-mixing intensity (Fig. 3c). Therefore, the reduced residual turbidity and enhanced filterability of the supernatant is attributed to the formation of larger flocs with intensive rapid mixing.

As illustrated in Fig. 3d, the residual DOC is highest at low velocity gradient (200 s^{-1}) but decreases substantially at high velocity gradient range ($400\text{--}800 \text{ s}^{-1}$) for the coagulation of low- and high-turbidity water. In this study, most of the PACl coagulant used was monomeric Al, which resulted in a large amount of $\text{Al}(\text{OH})_3$ precipitates at neutral pH during PACl coagulation. DOC removal by coagulation/sedimentation relies on complexation and adsorption/co-precipitation [24]. Intense mixing promotes the adsorption of DOC on the surface of $\text{Al}(\text{OH})_3$, which can subsequently be removed from water after sedimentation [10]. The results indicated that the destabilization of natural organic matter can be effectively improved by coagulation with efficient rapid-mixing intensity.

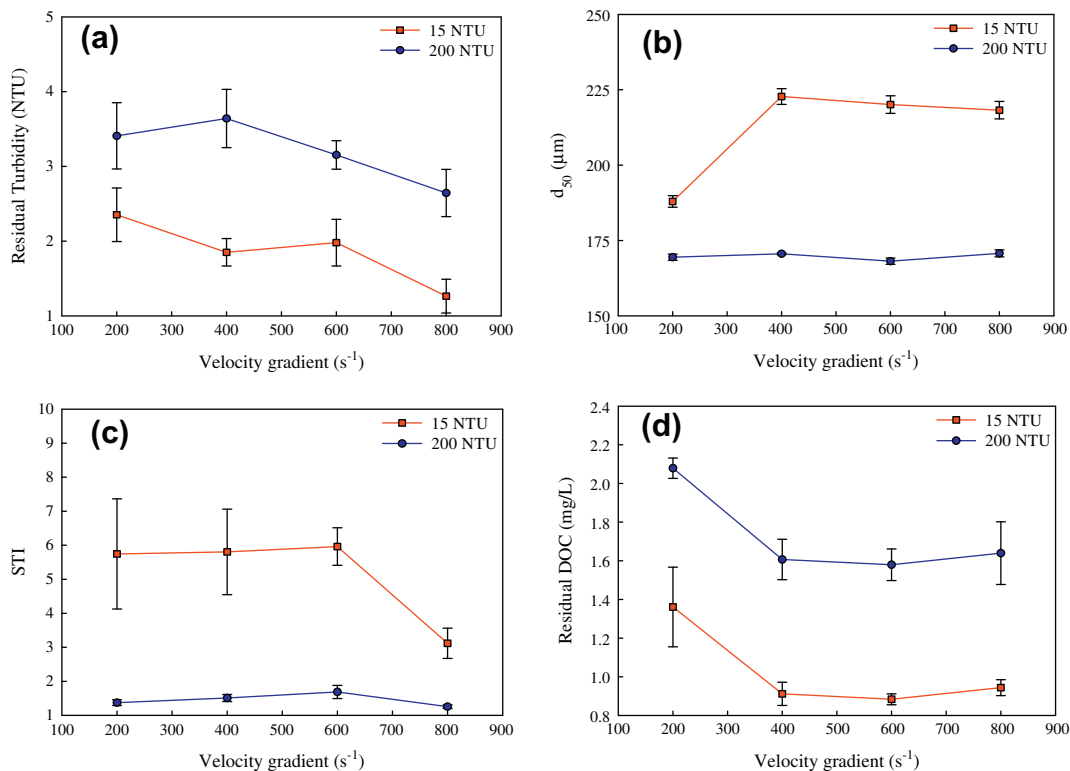


Fig. 3. Variation in coagulation efficiency and filterability with velocity gradient of rapid mixing: (a) residual turbidity, (b) floc size (d_{50}), (c) suction time index (STI), and (d) residual DOC.

3.3. Effect of rapid-mixing intensity on floc strength and re-growth ability

To understand the effect of rapid-mixing intensity on the strength of coagulated floc, the floc strength constant (γ) was determined after coagulation. γ decreased slightly with G value at low turbidity but did not change much with G at high turbidity (Fig. 4). This observation might be explained by the variation in particle collisions. Bache et al. [25] has suggested that the strength of floc is strongly related to the bonding force between particles within flocs. By intensive mixing, the coagulants can be effectively dispersed onto the surface of particles, which prompt the collision and the bonding between particles to form strong floc [26,27]. At low turbidity, larger floc forms at higher rapid-mixing intensity (Fig. 3b), suggesting the stronger bonding force between particles. By contrast, at high turbidity, the particles bond to each other effectively because of the high collision frequency, even at low mixing intensity. Thus, the rapid-mixing intensity has only a minor effect on the strength of the floc.

However, floc strength constant at low turbidity is higher than that at high turbidity, suggesting that the strength of flocs formed by coagulation at high turbidity is superior to that at low turbidity. As the formation pathway of floc governs the strength of flocs [15], the aggregation and breakage of floc during flocculation could affect the strength of coagulated floc [20]. In theory, the size of floc formed by coagulation at high turbidity should be larger than that at low turbidity. However, smaller floc was found in the coagulation of high turbidity water (Fig. 3b). When the turbidity is high, the floc may grow quickly in the beginning because of a high rate of collision, but the breakage of flocs could also occur with the longer mixing time in flocculation. Lin et al. [21] suggested that the structure of flocs could become stronger through structure rearrangement after the breakage of floc. As a result, stronger floc could be formed by coagulation at high turbidity through breakage and structure rearrangement.

On the other hand, the floc structure can affect the re-growth of flocs after floc breakage. As shown in Fig. 5, the fractal dimension (D_f) is inversely proportional to the recovery factor in our study, meaning that floc with low D_f is easier to re-grow after flocs are broken by shear force. Studies have suggested that more compact flocs with high D_f are likely to suffer surface erosion induced by shear during breakage, and more primary particles or small cluster are formed [28,29]. When floc breaks by surface erosion, the small aggregates could be eroded off the parent floc, which significantly

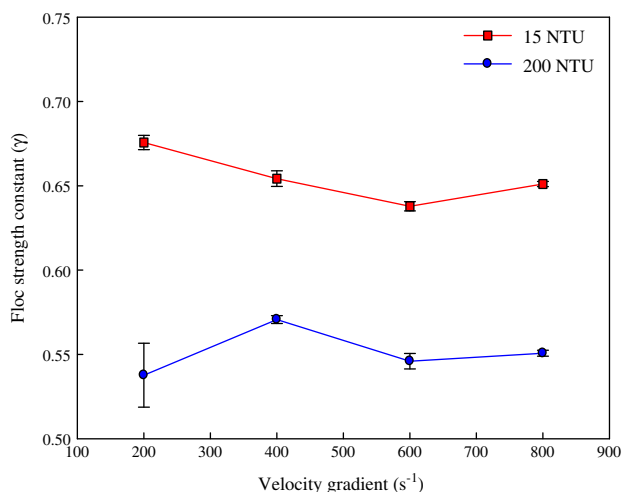


Fig. 4. Variation in floc strength constant with velocity gradient of rapid mixing.

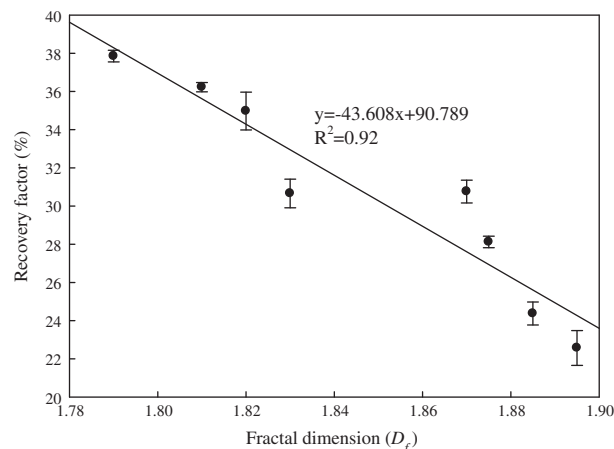


Fig. 5. The relationship between recovery factor and fractal dimension of flocs.

changes the effective binding sites between particles or clusters during floc reformation. Because the PACI coagulation favored sweep flocculation by aluminum hydroxide in our study, aggregates formed by chemical binding between particles. Once these flocs are broken by shear forces, they are difficult to re-aggregate because of the weak binding forces of broken aluminum hydroxide flocs, resulting in low recovery factor of flocs.

3.4. Effect of flash-mixing coagulation on particle destabilization and aggregation

The in-line IFM which yields much higher G than conventional mechanical mixing was used to investigate the effect of flash mixing on the efficiency of coagulation/sedimentation. The effluents from the in-line IFM and rapid-mixing unit of WTP were simultaneously flocculated at a constant slow-mixing intensity using jar test to determine the residual turbidity and DOC after sedimentation. During flocculation, the formation of flocs was monitored by a photometric dispersion analyzer (PDA). Three turbidities were used in this experiment. The variation in residual turbidity and DOC with velocity gradient induced by IFM and mechanical mixing was determined at the same optimal dosage as shown in Fig. 6. As can be seen, the highest residual turbidity and DOC was observed in coagulation with mechanical rapid mixing ($G = 560 s^{-1}$), which decrease substantially by IFM coagulation with G value of more than $5000 s^{-1}$. This significant decrease in residual turbidity and DOC is attributed to the effect of high mixing intensity on floc formation, even though the turbidity and DOC concentration of raw water are different in various IFM coagulation tests. With increased mixing intensity, the FI value increases quickly – Fig. 6c. This indicates that the primary particles formed by IFM mixing are easier to aggregate and form larger flocs during flocculation than those formed by mechanical mixing. At turbulent flow, the collision efficiency of suspended containments can be significantly improved at extremely high rapid-mixing intensity, which facilitates the growth of floc [13]. This phenomenon is especially prominent in coagulation of high-turbidity water because of the higher collision efficiency. Because the extremely high rapid-mixing intensity formed by in-line IFM can provide extremely high collision rate between particles in a very short period of time, large floc could form by cluster-cluster aggregation model during coagulation [9,30]. The results suggest that the extremely high rapid-mixing intensity ($G > 5000 s^{-1}$) in a short mixing time (t) can be induced by in-line IFM coagulation to form large flocs, facilitating the removal of particles and dissolved organic matter. In engineering application, operating in-line IFM is as easy as operating mechanical mixer.

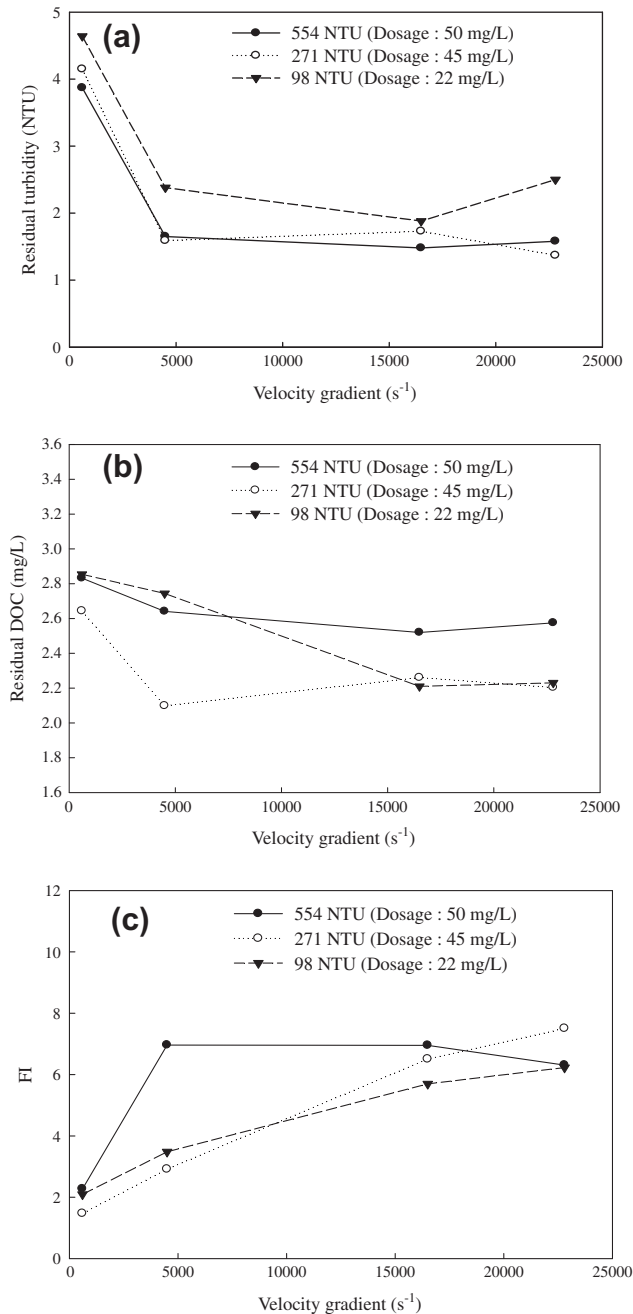


Fig. 6. Variation in coagulation efficiency with velocity gradient induced by in-line IFM and by mechanical mixing of WTP: (a) residual turbidity, (b) residual DOC, and (c) FI. All data of each IFM coagulation test was determined in duplicate.

By quickly adjusting the location of deflector plate in the nozzle area, the agitation with various velocity gradients can be immediately conducted to perform effective rapid-mixing for the coagulation of low- and high-turbidity water. Therefore, the in-line IFM is a simple and convenient technology to enhance the performance of coagulation in water treatment practices.

4. Conclusions

The efficiency of removing turbidity and DOC can be optimized by sweep flocculation for naturally turbid water. At optimal dosage, the residual turbidity and DOC can be effectively reduced by increasing the G value of rapid mixing. With intensive rapid mixing, the formation of large flocs can decrease residual turbidity

and DOC, thus resulting in increased filterability. The flocs become stronger with intense rapid mixing at low turbidity, but the rapid-mixing intensity has limited effect on the strength of flocs at high turbidity. In addition, the fractal dimension (D_f) of coagulated flocs is inversely proportional to recovery factor of broken flocs. By using an in-line IFM, the efficiency of removing turbidity and DOC can be improved more than with the mechanical WTP mixer, especially at $G > 5000 \text{ s}^{-1}$. The use of in-line IFM coagulation in water treatment is technically feasible for WTPs, so as to effectively enhance the efficiency of removing turbidity and organic matter.

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