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Influence of velocity gradient on aluminum and iron floc property for NOM removal from low organic matter surfacewater by coagulation

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ABSTRACT

Prehydrolyzed coagulants (polyaluminum chloride, denoted as PAC, and polyferric chloride, denoted as PFC) were used to study the effect of different velocity gradient values (G ranging from 4.4 s^{-1} to 28.3 s^{-1}) on the flocculation performance and floc properties from the low organic matter (OM) surfacewater. For different coagulants there were three distinct zones observed in terms of the magnitude of G. At low G values ($G < 7.6\ s^{-1}$), mean particle diameter increased with G, showing that aggregation dominated over breakup. The flocs were loosely compacted with poor settling ability and high strength and recovery factors. At intermediate G values ($7.6\ s^{-1} < G < 15.1\ s^{-1}$ for PFC; $7.6\ s^{-1} < G < 11.2\ s^{-1}$ for PAC), flocculation rates were maximized, resulting in large flocs with low strength and recovery factors. The residual turbidity was lower and the flocs were more compact. At higher G values ($G > 15.1\ s^{-1}$ for PFC; $G > 11.2\ s^{-1}$ for PAC), the dominant effect of breakup was shown through reduced maximum floc size with increasing G values. The strength and recovery factors increased and the residual turbidity also increased due to the small flocs. In addition, the PFC-NOM flocs properties were more influenced by the velocity gradients. In terms of NOM removal, the differences of the residual UV_{254} and DOC in the range of G studied were slight. The lowest SUVA value was achieved at G 7.6 s^{-1} and 15.2 s^{-1} respectively for PFC and PAC.

1. Introduction

Coagulation by metal salts is an important and widely applied process in water and wastewater treatment. In the process the repulsive potential of electrical double layers of colloids is reduced and micro-particles can be produced [1]. Coagulation of small particles into larger aggregates requires increasing the collision frequency between particles and minimizing breakup. Aggregates formed by coagulation are not uniform in size, and vary over a wide range. Maximizing the size of these aggregates is important for obtaining fast-settling particles [2].

The first step in the coagulation process is dominated by a rapid initial growth rate in the mean diameter of the particles. During this initial period, if particles are fully destabilized they aggregate as soon as they come into contact. In the flocculation process, aggregates become larger and have more tenuous and fragile structures that are susceptible to breakup by fluid shear. At this stage, there are fewer but larger aggregates, and the growth rate of aggregate size decreases. Finally, the aggregate size distribution reaches a pseudo-steady state, where breakup balances aggregation, and size spectra is essentially constant [3–5]. The flocculation stage facilitates aggregation to some extent, but as shear rate increases it limits the growth of aggregates [5–8]. Since the application of a specific velocity gradient value (G) determines the production of different types of flocs which may be more or less easy to be eliminated by physical processes, the flocculation process requires particular attention [9].

Previous studies found that the larger the shear the smaller the average aggregate size under steady-state conditions [3,5]. However, only G above 25 s^{-1} was examined. Colomer et al. [10] investigated the aggregation and breakup of particle flocs under low-shear flow (G ranging from 0.70 s^{-1} to 27.36 s^{-1}) and found that there was an increase of floc size at increasing G values. Spicer and Pratsinis [4] examined the effect of different impellers on the aggregation and breakup of flocs over a wide range of G values (ranging from 4 s^{-1} to 102 s^{-1}). In addition, Serra et al. [2] studied the effect of three different laboratory devices (paddle mixer, oscillating grid and Couette) on the particle size spectra with G values ranging from 4 s^{-1} to 102 s^{-1}. Xiao et al. [11] found that the steady floc sizes of the particles in the suspension became smaller and more regular when slow-mixing speed was increased. In the studies cited, only coagulation of particles was examined. Except for the study by Rojas et al. [9] who only applied different G values (27, 47, 87 and 104 s^{-1}) in studying the effect on the membrane clogging

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Table 1
Raw water characteristics and the methods of measurement.

Characteristic	Value	Methods of measurement
Dissolved organic carbon (DOC) (mg/L)	2.571 to 2.679	Prefiltered sample measured on a TOC analyzer (Shimadzu, Japan)
Ultraviolet light absorption at 254 nm (UV_{254}) (cm^{-1})	0.050 to 0.054	Prefiltered sample measured on a UV-754 UV/VIS spectrophotometer (Shanghai Jinhua Group Co., Ltd., China)
Turbidity (NTU)	1.51 to 1.98	Turbidimeter 2100P (Hach, America)
Zeta potential (mV)	−13.4 to −10.3	Zetasizer 3000HSa (Malvern Instruments, UK)
pH	8.17 to 8.51	pH analyzer
Specific UV absorbance (SUVA) (L/(mg m))	1.87 to 2.10	$\mathrm{SUVA} = \frac{UV_{254}}{DOC} \times 100$

for natural organic matter (NOM) removal, few studies have investigated the coagulation performance and floc properties for NOM removal under various low-shear flows at the flocculation stage. Additionally, there is limited information in the literature on the floc properties for NOM removal from waters with low organic matter (OM). These waters may be characterized by having hydrophilic OM which is difficult to be removed and shows appreciable DBPs yields [12].

In this study coagulation experiments were conducted on the low OM surfacewater in the flat paddle mixers under different slow-mixing rates in order to evaluate the effect of *G* on coagulation efficiency and floc properties. Prehydrolyzed coagulants (polyaluminum chloride, denoted as PAC, and polyferric chloride, denoted as PFC) were used as coagulants which have many advantages over conventional salts including less sludge production, less dependence on temperature and pH [13,14]. The effects of *G* on the floc properties between PFC-flocs and PAC-flocs were compared.

2. Materials and methods

2.1. Preparation of coagulant

The PFC used in this experiment was prepared with $FeCl_3{\cdot}6H_2O$ (A.R.) and Na_2CO_3 (A.R.). Firstly, $FeCl_3{\cdot}6H_2O$ was dissolved in distilled water, then slowly mixed with Na_2CO_3 powder at room temperature to obtain the desired basicity (*B*, OH/Fe molar ratio) [15]. The solution was stirred until foam disappeared and became transparent. Then, Na_2HPO_4 (A.R.) was added to the solution as a stabilizer. The characteristics of PFC were as follows: $Fe_a = 17.4\%$, $Fe_b = 29.8\%$, $Fe_c = 52.8\%$, pH = 0.7, $B = 0.5$. The dosages of PFC were calculated as mg/L of Fe during coagulation experiments.

PAC with a basicity value (*B*, OH/Al molar ratio) of 2.0 was synthesized by adding pre-determined amount of Na_2CO_3 (A.R.) slowly into $AlCl_3$ (A.R.) solution under intense agitation. The temperature was kept at $80.0 \pm 0.5\,^{\circ}C$ by using recycling water bath [16]. The properties of PAC were as follows: $Al_a = 24.8\%$, $Al_b = 32.1\%$, $Al_c = 43.1\%$, pH = 2.85, $B = 2.0$. The dosages of PAC were calculated as mg/L of Al during coagulation experiments.

2.2. Raw water

All experiments were conducted with the raw water collected from Queshan reservoir, an important drinking water source in Jinan, China. The raw water characteristics, along with the methods of measurement, were shown in Table 1. The raw water had DOC values <3 mg/L which can be regarded as low OM water. The specific UV absorbance (SUVA) of the raw water was less than 4 mg/L, which indicated that the DOC was hydrophilic, non-humic, low in molecular weight, and difficult to be removed.

Table 2
The rotary speed (rpm) and its corresponding *G* value (s^{-1}).

Rotary speed (rpm)	20	30	40	50	60	70	80
G^a (s^{-1})	4.4	7.6	11.2	15.1	19.2	23.7	28.3

[a] $T = 15\,^{\circ}C$.

2.3. Coagulation tests

Coagulation experiments were performed on a program-controlled jar-test apparatus (ZR4-6, Zhongrun Water Industry Technology Development Co. Ltd., China) at 15 °C. For coagulant dosage optimization, the water samples (1.0 L) were mixed rapidly at 200 rpm for 1 min after dosing, followed by slow stirring at 40 rpm for 15 min. The flocs were allowed to settle for 25 min, and the supernatant sample was withdrawn by a syringe from about 2 cm below the water surface for analysis. For investigating the effects of slow-mixing intensity, slow-mixing speed was adjusted to 20 rpm, 30 rpm, 40 rpm, 50 rpm, 60 rpm, 70 rpm and 80 rpm, respectively. The *G* values of different rotary speeds were listed in Table 2.

2.4. Floc size

The floc size was measured by a laser diffraction instrument (Malvern Mastersizer 2000, Malvern, UK). Coagulation test was conducted on a cylindrical jar tester with the diameter of 100 mm and with a 50 × 40 mm flat paddle impeller. The suspension was monitored by drawing water through the optical unit of the Mastersizer and back into the jar by a peristaltic pump at a flow rate of 2.0 L/h. Dynamic floc size measurements were taken every 0.5 min for the duration of the jar test and logged onto a PC.

2.5. Floc strength and recovery factor

Floc strength and regrowth experiments were performed under the following conditions: a rapid mix at 200 rpm for 1 min, followed by a slow stir phase at different *G* values for 15 min, then a 5 min breakage phase at 200 rpm. After the breakage phase, a slow stirring at 40 rpm was reintroduced for a further 15 min for flocs regrowth. Dynamic floc size was measured during the breakage and regrowth of the flocs using the laser diffraction instrument (Malvern Mastersizer 2000, Malvern, UK). Floc strength and recovery factors, which had previously been used to compare the relative strength and regrowth of flocs in different flocculated systems, were calculated as follows [8,17,18]:

$$\text{strength factor} = \frac{d_2}{d_1} \times 100 \tag{1}$$

$$\text{recovery factor} = \frac{d_3 - d_2}{d_1 - d_2} \times 100 \tag{2}$$

where d_1 (μm) is the average floc size of the steady phase before breakage, d_2 (μm) is the floc size after the floc breakage period, and d_3 is the floc size after regrowth to the new steady phase. The strength factor value indicates the ability of flocs to withstand shear. A larger value of strength factor suggests that flocs are stronger than the flocs with a lower factor. Likewise, the floc with a larger recovery factor shows better regrowth after high shear.

2.6. Floc fractal dimension

Light scattering method was used here for determining aggregate mass fractal dimension. The theory of the mass fractal

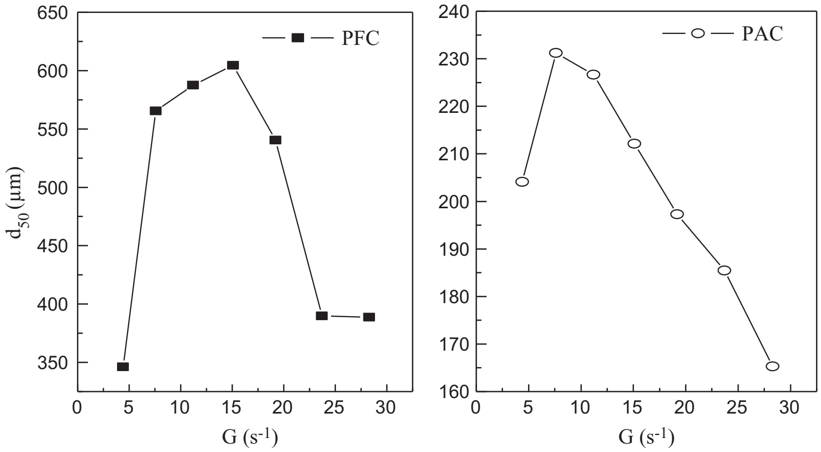

Fig. 1. The NOM floc size formed by PFC and PAC under various G values.

dimension has been reported in details in a few literatures [19,20]. The light scattering technique involves measurement of light intensity I as a function of the scatter vector Q. The vector is defined as the difference between the incident and scattered wave vectors of the radiation beam in the medium [20], which is given as follows:

$$Q = \frac{4\pi n \sin(\theta/2)}{\lambda} \tag{3}$$

where n, θ, and λ are the refractive index of the medium, the scattered angle, and the wavelength of radiation in vacuum, respectively.

For independently scattering aggregates, the relationship between I and Q can be represented by:

$$I \propto Q^{-Df} \tag{4}$$

D_f is the fractal dimension and can be determined by the slope of a plot of I as a function of Q on a log–log scale. High D_f means the primary particles in an aggregate are arranged compactly, while low D_f results from highly branched and loosely bound structures.

3. Results

3.1. Coagulant dosage optimization

Through standardized jar testing procedures, it showed that the optimum dosage for PFC was 14 mg/L, which yielded an optimum UV_{254} and DOC removal efficiencies of 36% and 31%, respectively. Whilst the corresponding values for PAC were 39% and 40% respectively at a dosage of 12 mg/L.

3.2. Effect of velocity gradient on the floc property

3.2.1. Floc size

Flocs were formed under different slow-mixing conditions in a series of jar tests to examine the effect of G on the properties of NOM flocs. Floc size was measured at the steady stage of the floc growth by Mastersizer 2000. d_{50} was selected as the representative floc size and the results were shown in Fig. 1. For PFC-NOM flocs, d_{50} reached a maximum at G 15.1 s^{-1} and at higher slow-mixing rates the maximum particle size decreased. For PAC-NOM flocs, the d_{50} increased with G up to 7.6 s^{-1}. And at higher rates, the maximum floc size decreased. With the G investigated, the PFC-NOM flocs size changed greatly from 610 μm to 390 μm when G value was increased from 15.1 s^{-1} to 23.7 s^{-1}. While the PAC-NOM flocs size changed from 230 μm to 160 μm with G from 7.6 s^{-1} to 28.3 s^{-1}. The result was consistent with the study that the Fe-floc size decreased more easily with the increase of slow-mixing rate [11].

3.2.2. Strength and recovery factor

In order to characterize the floc strength and re-formation under different G values in the flocculation phase, strength and recovery factors were calculated as Eqs. (1) and (2), respectively. The results were summarized in Table 3. It was found that the strength factor of the flocs decreased when G was increased from 4.4 s^{-1} to 7.6 s^{-1}. And then the strength factor began to increase. The trend of the recovery factor was similar to that of the strength factor. The recovery factor decreased from 96 to 35 for PFC-NOM flocs, and from 32 to 26 for PAC-NOM flocs with G increased to 11.2 s^{-1}. And at higher G values, the recovery factor increased.

However, it should be pointed out that the strength and re-formation of flocs were not only controlled by the G value but also by the characteristics of different coagulants. In the middle range of 7.6 $s^{-1} < G <$ 19.2 s^{-1}, PAC-NOM floc had higher strength factor which indicated that the break-up behavior of these flocs under exposure to an increased shear was very low. While the flocs formed by PFC presented a better re-formation capacity based on the higher recovery factors as shown in Table 3. Additionally, the PFC-NOM flocs seemed to be more influenced by G than the PAC-NOM flocs.

3.2.3. Floc fractal dimension

A comparative study on the floc structure at different G values at the flocculation stage was carried out in terms of fractal dimension. The fractal dimension (D_f) for the flocs in the steady-state phase (16 min) was shown in Fig. 2. The D_f of PFC-NOM flocs initially increased and then gradually decreased. And the D_f values reached the maximum at G 15.2 s^{-1}. In terms of PAC-NOM flocs, a continuous and gentle increase in D_f was observed with the increase of G at the flocculation stage. It also could be seen from Fig. 2 that PFC gave more compact floc structure than PAC except at lower velocity gradients. In addition, the differences of D_f at various velocity

Table 3
Strength and recovery factors of flocs under different G values.

Coagulant	G/s^{-1}						
	4.4	7.6	11.2	15.1	19.2	23.7	28.3
Strength factor							
PFC	73	33	34	35	39	51	49
PAC	45	41	44	44	46	51	57
Recovery factor							
PFC	96	36	35	33	42	46	50
PAC	32	28	26	26	33	35	38

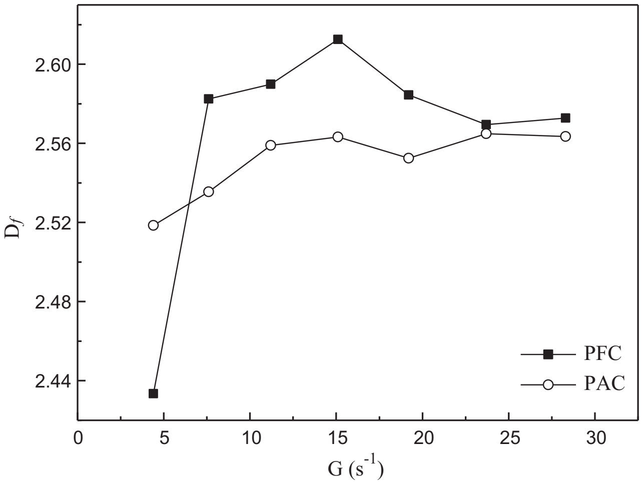

Fig. 2. The fractal dimension of NOM flocs formed by PFC and PAC under various G values.

gradients were smaller for PAC-NOM flocs compared to PFC-NOM flocs.

3.3. Effect of velocity gradient on the coagulation efficiency

The effect of different slow-mixing rates on turbidity and NOM removal was investigated. The variations of residual turbidity, UV_{254} and DOC were shown in Fig. 3. As shown in Fig. 3, the residual turbidity by PFC was sharply decreased with the increasing G and slightly increased when G was above 23.7 s^{-1}. For PAC the lowest residual turbidity was achieved at G 11.2–19.2 s^{-1}. In terms of UV_{254}, the residual UV_{254} by PFC showed a slight rise when G increased. The trend of residual UV_{254} curve by PAC coagulation was not clear and the values were in the range of 0.034–0.037. The residual DOC after coagulation increased a little at first and then decreased when the slow-mixing rate increased. However, the differences of residual DOC at various G values were not obvious.

The SUVA values after coagulation by PFC and PAC were displayed in Fig. 4. The SUVA value is an index of the relative aromaticity and hydrophobicity/hydrophilicity of the DOC present in water [21,22]. It could be found from Fig. 4 that the SUVA value decreased and then increased with the increasing G. At low slow-mixing rate, an increase in slow-mixing rate generally resulted in a decrease of SUVA value, which indicated that higher-SUVA components of NOM were preferentially removed. For higher slow-mixing rate, higher SUVA values indicated that none or lower UV-absorbing fractions were preferentially removed. The lowest SUVA was achieved at G 7.6 s^{-1} and 15.2 s^{-1} respectively for PFC and PAC.

4. Discussion

Mooyoung and Lawler [23] claimed that the major significance of mixing in coagulation is to keep the largest possible number of particles suspended. It was noted that the floc size first increased and then decreased with the increased G in the flocculation process which demonstrated that there were three different ranges for NOM flocs formation: low velocity gradient ($G < 7.6$ s^{-1}), which was the aggregation-dominated range; mean velocity gradient (7.6 s^{-1} < G < 15.1 s^{-1} for PFC; 7.6 s^{-1} < G < 11.2 s^{-1} for PAC), where coagulation rates were maximized and breakup minimized; and high velocity gradient ($G > 15.1$ s^{-1} for PFC; $G > 11.2$ s^{-1} for PAC), where flocs breakup dominated. In the aggregation-dominated range, aggregates size increased in proportion to the shear rate, but resulted in small floc sizes that were limited by low particle collision rates. Although breakup probably occurred, it was considered relatively insignificant to the overall aggregate sizes. In the mean velocity gradient, the balance between the rate of aggregation and the rate of breakage was reached; the flocs reached the maximum size. In the breakup-dominated range, there were sufficient collisions to produce larger flocs, but those flocs were not stable due to breakup. Thus, the floc size began to decrease.

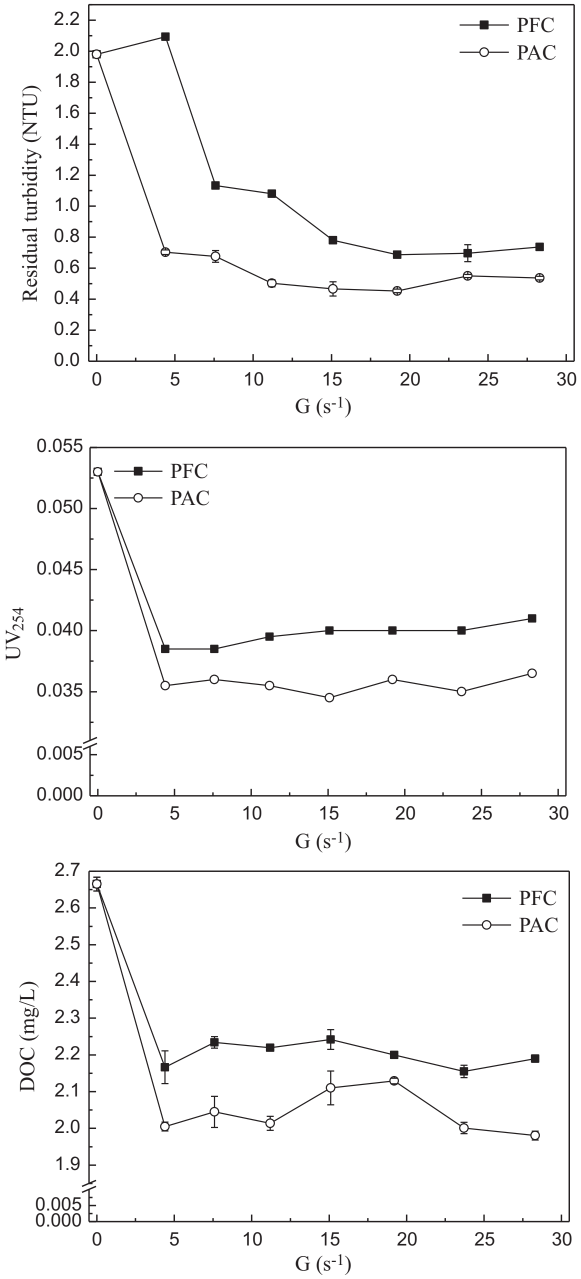

Fig. 3. Coagulation efficiency by PFC and PAC under various G values.

The maximum size of the aggregates produced in these experiments depended not only on G value but also on the type of coagulant. PFC produced maximum particle sizes 600 μm at 15.1 s^{-1}, while PAC produced the largest flocs with d_{50} 230 μm at 7.6 s^{-1}. It was consistent with the findings by Jarvis et al. [18] that the Fe-NOM flocs were larger than Al-NOM flocs.

The trend of the strength factor was contrary to that of the floc size. This may be due to the different floc size formed at different G values. Larger flocs became more affected by the microscale eddies that were attributed to floc breakage. Smaller flocs were more likely

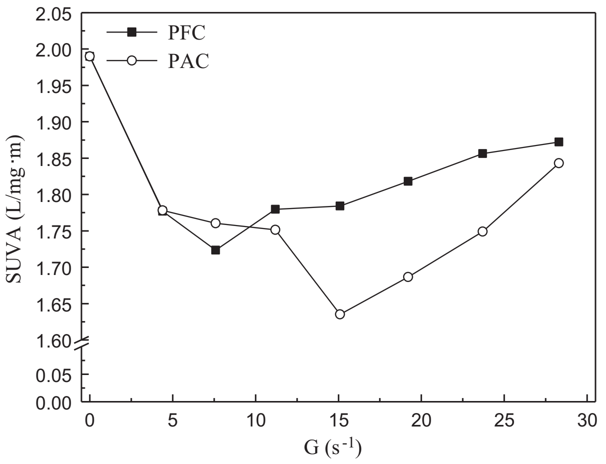

Fig. 4. SUVA values by PFC and PAC under various *G* values.

to become entrained within the eddies rather than being broken by them [18]. So in the mean velocity gradient, the strength factor of the flocs was small due to the large flocs which were easily affected by the increasing shear. At higher slow-mixing rate, the strength of the floc increased as the floc size was reduced which was consistent with the finding by Xiao et al. [11]. After the increasing shear, the small flocs could continue to grow to a large extent and the recovery factor was high which may be attributed to the internal bonds of the small flocs which were not greatly affected by the increasing shear. At the mean slow-mixing rate, the chemical bonds of the large flocs were greatly affected by floc disruption which caused the low recovery ability [8].

During flocs aggregation, the floc structure changed continuously because the flocs' internal bonds broke and re-formed at more favorable points where the attractive force was greater or the repulsive force was lower [24]. So the floc structures became more compact during the coagulation process and were stable by restructuring and rearrangement [25]. At the lower *G* values, the aggregation was restricted by the limited collision rates. Restructuring and rearrangement were limited and the flocs were more loosely packed. As *G* increased, restructuring probably occurred and thus was the governing mechanism together with the aggregation kinetics to the final aggregate properties. The aggregates were restructured to a more compact arrangement, resulting in the higher D_f. At the higher *G* values, the aggregates reached their compact size early in the process [25]. Thus, aggregation and fragmentation seemed to be the governing mechanisms in the evolution to the final aggregate size, although restructuring also took place. The PFC-NOM flocs became loosely packed due to fragmentation. While for the small PAC-NOM flocs, fragmentation was not important. The D_f did not decrease and remained almost constant. Hence the decrease in PAC aggregate size was largely due to the compaction of aggregates rather than fragmentation.

Overall, *G* values had a stronger influence on the PFC-NOM flocs than the PAC-NOM flocs. This may be attributed to the different floc size at the flocculation stage. PFC-NOM flocs were larger than the PAC-NOM flocs which were easily affected by *G* values [18] in the flocculation process. Additionally, PAC-NOM flocs presented lower rates of change in size at different slow-mixing rates, indicating that they had stronger bond [11]. And this may be another reason why the floc properties formed by PAC were less affected by the slow-mixing rates.

Istv'an [26] showed that the hydrolysis of metal ions occurred immediately after contacting with water. The mechanisms of destabilization NOM not only include the charge-neutralization and adsorption but also the entrapment [27,28]. These mechanisms were mainly influenced by the rapid-mixing process which was important for coagulants rapid dispersal [29]. Hence the NOM removal was not greatly influenced by the slow-mixing rate investigated. The differences in residual UV_{254} and DOC under different slow-mixing rates were not obvious.

At the low *G* values, the flocs were small and loosely packed with poor settling ability which increased the residual turbidity [30]. Similarly, in the case of $G > 23.7\,s^{-1}$, the floc breakage and the formation of smaller particles did not settle well in the settling period and thus contributed to the residual turbidity [11,31]. So the lower and greater slow-mixing intensity slightly increased the residual turbidity.

5. Conclusions

Three different zones were found to be depending on *G* values in the flocculation process. At low *G* values, the mean particle diameter increased with *G* values; in this case the aggregation dominated over breakup. At intermediate *G* values flocculation rates were maximized, producing the largest flocs. For large *G* values, breakup dominated over aggregation. In the last case, an increase in the shear rate caused a reduction in the mean particle size. The trend of the strength and recovery factors was contrary to that of the floc size: first decreased and then increased with increasing *G* in the flocculation process. Additionally, the flocs became more compact with increasing *G* except the PFC-NOM flocs at higher *G* value. The UV_{254} and DOC removal were slightly influenced by the *G* values and the lowest SUVA value was achieved at *G* $7.6\,s^{-1}$ and $15.2\,s^{-1}$ respectively for PFC and PAC. The residual turbidity was low under the intermediate slow-mixing rates. Overall, the *G* values in the slow-mixing phase had greater influence on the PFC-NOM flocs compared to the PAC-NOM flocs.

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