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Influence of velocity gradient and rapid mixing time on flocs formed by polysilica iron (PSI) and polyaluminum chloride (PACl)

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ABSTRACT

The aim of this investigation was to evaluate the effect of different velocity gradient and rapid mixing time on coagulation and floc properties, using polysilica iron (PSI) and polyaluminum chloride (PACl) with different rapid mixing speeds and times. The growth, breakage, and re-growth of flocs were monitored by a particle-size analyzer during the rapid mixing, in order to determine the underlying mechanisms. For PSI and PACl, three different zones were found to depending on the G value in the coagulation process. At low G values ($G < 546\,\mathrm{s}^{-1}$ for PSI flocs and $G < 390\ \mathrm{s}^{-1}$ for PACl flocs), the aggregation dominated over breakup. At mean G values ($G = 546\ \mathrm{s}^{-1}$ for PSI flocs and $G = 390\,\mathrm{s}^{-1}$ for PACl flocs), floc formation was maximized with the largest flocs produced at the lowest residual turbidity. For large G values ($G > 546\,\mathrm{s}^{-1}$ for PSI flocs and $G > 390\,\mathrm{s}^{-1}$ for PACl flocs), breakup dominated over floc formation. Broken flocs did not fully re-grow after breakage, probably as a result of a change in the floc surface properties arising from the rupture of bonds within the hydroxide precipitate. A rapid mixing time of 60 s led to maximum floc formation. In addition, PSI flocs were more durable to high than PACl.

Keywords: Velocity gradient; Rapid mixing time; Polysilica iron (PSI); Polyaluminum chloride (PACl); Particle-size distribution

1. Introduction

Coagulation is one of the most important processes used in the removal of suspended particles in water treatment plants. The coagulation process involves many factors: besides the velocity gradient and duration of time, the solution pH, type of coagulant and its dosage, particle properties and concentration, all play a role [1]. The interparticle collision frequency, the growth of aggregates, and the uniformity of the dispersion of hydrolyzed products of the coagulants have been shown to be especially dependent on the velocity gradient (G) and duration [2].

The initial phase of 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 [3]. At this stage, when there are fewer but

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larger flocs, the growth rate of the flocs decreases. Finally, the floc-size distribution reaches a steady state, where a balance is reached between floc breakup and floc formation, and the size spectra are essentially constant [4,5].

Several researchers have investigated the influence of the velocity gradient and rapid mixing time on the coagulation process. For example, Rossini et al. [6] found that a rapid mixing time had a significant influence on coagulation efficiency. Although a short rapid mixing time provided the lowest values of residual turbidity, a prolonged rapid mixing time resulted in a poorer performance. Vrale et al. [7] suggested that an instant rapid mixing time below 1 s provided the best coagulation efficiency. Mhaisalkar et al. [8] obtained similar conclusions, although the effects of rapid mixing conditions were not marked. At much lower G values ($25\,\mathrm{s}^{-1}$), floc growth is much slower and the flocs are smaller in size [9]. Colomer et al. [10] investigated the aggregation and breakup of particle flocs under a low-velocity gradient (from 0.70 to $27.36\,\mathrm{s}^{-1}$) and reported an increase in floc size with increasing G values. These authors suggested some values for G and an optimal time for rapid mix design, but these were probably specific to the systems investigated. Also, despite mentioning other conflicting data from the literature on rapid mixing effects, they were unable to explain their results.

In addition, the effect of the type of coagulant has also been studied by many researchers. Yukselen and Gregory [11] found that while larger flocs were formed with aluminum-based coagulants with shorter rapid mixing times, only limited re-growth occurred in all cases, indicating a significant irreversibility of the flocs break-up process. However, in the case of cationic polyelectrolytes, while longer rapid mixing times were needed for floc formation, they promoted the growth of very large flocs, about four times the size of those with an alum coagulant and twice the size of those with polyaluminum chloride (PACl). Yu et al. [12] reported that when charge neutrality dominates the coagulate mechanisms, the strength and re-growth factor of flocs formed by alum was higher than that of flocs formed by PACl. When sweep flocculation dominates the coagulation mechanism, it is difficult to compare their re-growth factor. Zhao et al. [13] showed that the average size of flocs formed by titanium tetrachloride ($TiCl_4$) was larger than those by PACl during the growth period. However, flocs formed by PACl had a better reformation capacity based on the higher recovery factors of flocs formed with this coagulant than flocs formed by $TiCl_4$.

To date, numerous studies have been done about the influence of the velocity gradient and rapid mixing time on flocs formed by aluminum-based coagulants such as PACl. However, research on the genetic determinants of Alzheimer's disease has suggested that aluminum may be an important factor in this disease because of its neurotoxicity [14–16]. To avoid the use of aluminum, a new inorganic polymer coagulant, polysilicate iron (PSI) was developed to provide a substitute for aluminum coagulant, and its coagulation characteristics have been investigated by the authors in previous studies [17–19]. However, the floc properties of PSI under different G values and rapid mixing times have yet to be investigated.

Therefore, the aim of this study is to investigate the effect of different velocity gradients and rapid mixing times on the formation, breakage, and re-growth of kaolin flocs formed by PSI and PACl.

2. Materials and methods

2.1. Preparation of kaolin suspension

Kaolin clay was used as the model suspension (Kanto Chemical, Japan). Five grams of kaolin was dispersed completely in 500 mL of distilled water with a high-speed blender. After blending, the kaolin suspension was made up to 1 L and allowed to stand for 24 h to allow for the complete hydration of the kaolin. The stock solution was diluted with tap water to adjust the sample turbidity to 10 NTU. The alkalinity was adjusted to 35 mg/L (as $CaCO_3$) by adding $NaHCO_3$. The diluted synthetic water samples were kept in reservoirs at room temperature before use.

2.2. Coagulants agents

Polyaluminum chloride (PACl) and polysilicato iron (PSI) were obtained from Kanto Chemical Co. Inc and Suido Kiko Kaisha Ltd., respectively. Stock solutions with PACl and PSI were prepared in distilled water.

2.3. Coagulation tests and rapid mixing

Coagulation experiments were conducted with a high-speed jar test apparatus with four paddle-stirrers (60 mm × 30 mm). The paddles and the shafts were made of stainless steel in the Technical Division in Tohoku University. The procedure comprised of 3 min of rapid mixing at 150 rpm for the addition of varying coagulant doses and subsequent pH adjustment using the appropriate amounts of HCl and NaOH to maintain pH of 7.0. This was followed by a 30 min flocculation period at 30 rpm and a settling period of 60 min.

After settling, samples were taken from 2 cm below the surface for measurements. The residual turbidity of the supernatant was measured once the specimen had settled. The zeta potentials of the suspensions were also measured with a zeta analyzer.

Rapid mixing experiments were performed with the same mixing device. Rapid mixing according to changes in the velocity gradient and mixing time was conducted to investigate the growth of flocs in each condition. A range of rapid mixing speeds between 100 ($\sim 20\,s^{-1}$) and 1,600 rpm ($\sim 1{,}110\,s^{-1}$) was tested and the mixing times varied between 30 and 180 s.

2.4. Floc size

The floc growth rate and the average floc size after slow mixing were analyzed by particle-size analyzer (Microtract ASVR, HRA X-100, Nikkiso, Japan). Samples were taken using a pipette and added to the particle-size analyzer cell.

2.5. Floc strength and recovery factors

The floc-strength factor (S_f) and the recovery factor (R_f) are well-established parameters used to describe flocs, and can be calculated as follows [20,21].

$$S_f = \frac{d_2}{d_1} \times 100$$
$$R_f = \frac{d_3 - d_2}{d_1 - d_2} \times 100 \qquad (1)$$

where d_1 is the fully-grown floc size before breakage, d_2 is the floc size after floc breakage period, and d_3 is the floc size after regrowth to the new state stay phase. Here, the strength factor is indicative of floc strength, which refers to the ability to resist rupture by a velocity gradient, while the recovery factor indicates the recovery ability of the floc. Larger values of strength factors indicate that the flocs are less susceptible to the flocs that are to breakage as a result of increased shear rate and are, therefore, considered stronger. Flocs with larger recovery factors show better recovery ability after high shear, that is, better re-growth after breakage.

2.6. Zeta potential measurements

Zeta potential measurements were obtained using a Micro-Electrophoresis Apparatus Mk II (Rank Brothers, UK). The zeta potential was evaluated at a room temperature of 20 ± 1℃ and in suspension under an applied electric field of 80 mV. The zeta potential was calculated according to the Smoluchowski equation.

3. Results

3.1. Coagulant dosage optimization

The optimum coagulant dose for kaolin suspension was determined by the standard jar tests. Several PSI and PACl dosages, ranging from 10 to 50 mg/L, were added to coagulate the kaolin suspension. The zeta potential of the kaolin particles in the suspension and the residual turbidity after mixing and settling are shown in Fig. 1. The results showed that the maximum turbidity removal efficiency for PSI was achieved with a coagulant dose of 30 mg/L. The corresponding value for PACl was 40 mg/L.

The zeta potential of kaolin solution with a PSI of 30 mg/L was −1.9 mV. On the other hand, the zeta potential of kaolin solution was −1.1 mV with a PACl of 40 mg/L. This indicates that the predominant mechanism for kaolin coagulation is charge neutalization in the PACl experiments, and that charge neutralization is not the only factor in the PSI experiment. Shi et al. [22] reported that the coagulation of turbid materials was the result of charge neutralization and interparticle bridging in PSI.

Residual turbidity and the zeta potential of kaloin system increased gradually at PSI doses of over 30 mg/L and PACl doses of over 40 mg/L, due to restabilization by the charge reversal of particles. From these results, the optimum dosage of PSI and PACl was determined to be 30 and 40 mg/L, respectively.

3.2. Effect of velocity gradient on the floc property of kaolin system

3.2.1. The floc size

In order to determine the effect of the G values and the rapid mixing time on the growth properties of kaolin suspension with PSI and PACl, the size of the flocs during rapid mixing were analyzed over a

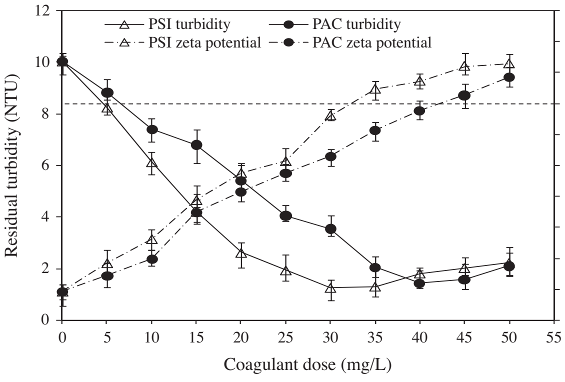

Fig. 1. Effect of coagulant dose on residual turbidity and zeta potential.

range of G values from 100 ($\sim 20\,\mathrm{s}^{-1}$) to 1,600 rpm ($\sim 1{,}110\,\mathrm{s}^{-1}$) and mixing times from 30 to 180 s.

The results reveals that the floc-formation process can be divided into three regions: a growth region with a low-velocity gradient ($G < 200\,\mathrm{s}^{-1}$), a steady state region with a mean-velocity gradient ($200\,\mathrm{s}^{-1} < G < 546\,\mathrm{s}^{-1}$ for kaolin-PSI flocs and $200\,\mathrm{s}^{-1} < G < 390\,\mathrm{s}^{-1}$ for kaolin-PACl flocs), and a re-growth range with a high-velocity gradient ($G > 546\,\mathrm{s}^{-1}$ for kaolin-PSI flocs and $G > 390\,\mathrm{s}^{-1}$ for kaolin-PACl flocs). The results are illustrated in Fig. 2. In addition, the size data are expressed as an equivalent volumetric diameter, and d_{50} was selected as the representative floc size in this paper, which refers to the 50 percentile floc size. Kaolin-PSI flocs reached a steady state floc size between G values of 200 and 546 s^{-1} with a mixing time of 60 s. The d_{50} value of kaolin-PSI flocs reached a maximum size (347.6 μm) at G 546 s^{-1} with a mixing time of 60 s, which corresponded with the lowest residual turbidity. When the G value was over 546 s^{-1} and the mixing time was over 60 s, the amount of d_{50} flocs significantly decreased as a result of floc breakage. Through the breakage period, the size of the kaolin-PSI flocs changed from 347.6 to 105.3 μm when the G value was increased from 640 to 1,110 s^{-1}. Following the breakage period, a slight re-growth in the kaolin-PSI floc, from 105.3 to 113.3 μm was observed.

Fig. 3 shows the kaolin-PACl flocs formation at different velocity gradients. The maximum d_{50} value of kaolin-PACl flocs (287.5 μm) was obtained at G 200 s^{-1} with the rapid mixing time of 60 s. The kaolin-PACl flocs were almost the same size between G values 200 and 390 s^{-1}. At G values over 390 s^{-1}, the kaolin-PACl flocs significantly decreased in size from 284.6 to 63.4 μm. The re-growth rate of the kaolin-PACl flocs was very low, as it was in kaolin-PSI experiments. In addition, the results also showed that the PSI flocs had better shear strength than the PACl flocs.

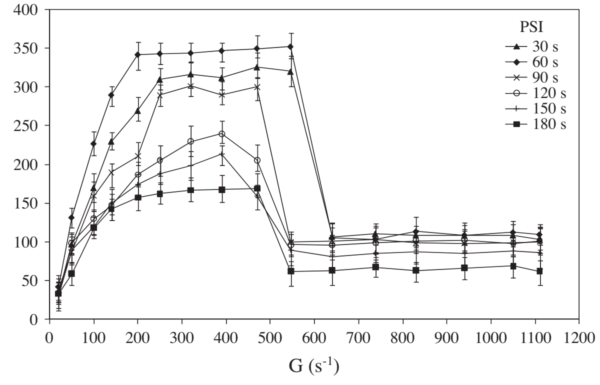

Fig. 2. The kaolin floc size formed by PSI under various G values.

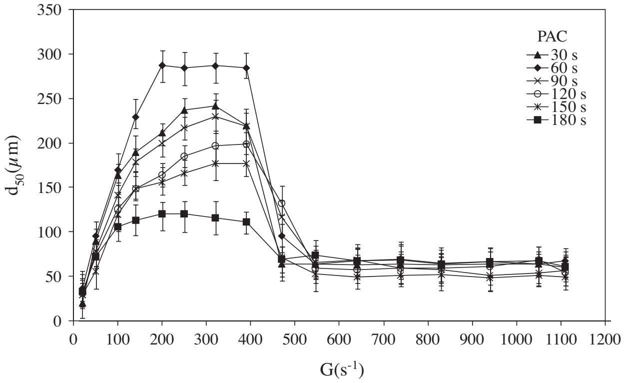

Fig. 3. The kaolin floc size formed by PACl under various G values.

3.2.2. Strength and recovery factor

In order to compare the floc strength and reformation of PSI and PACl flocs under different G values, the strength and recovery factors were calculated as Eqs. (1) and (2), respectively. The results are summarized in Table 1. The general trend was that the strength factor of the flocs decreased with increasing d_{50}. For PSI, when G was increased from 200 to 470 s^{-1}, the floc size increased from 341.3 to 349.8 μm, and the floc strength of the PSI flocs, therefore, decreased from 51.0 to 45.6. Then, when G was increased from 470 to 1,110 s^{-1}, the d_{50} decreased from 349.8 to 109.6 μm due to floc breakeage and the strength factor began to increase from 45.6 to 78.7 again. A similar trend with regard to floc strength was also observed for PACl flocs. When G was increased from 200 to 390 s^{-1}, the d_{50} slightly increased from 284.5 to 287.6 μm, and correspondingly, the floc strength decreased from 62.2 to 57.1 for PACl. At highest G value (1,110 s^{-1}), d_{50} decreased to 67.2 μm and the floc strength increased to 72.7 in parallel with the decreasing size of the flocs.

Kaolin flocs formed by PSI are larger flocs than PACl, which means PSI formed the most compact kaolin flocs, while PACl formed the loosest kaolin flocs. As a result, the floc strength measurement shows a clear relationship between floc strength and floc size. This means that floc strength is dependent on the floc size. This relationship is plotted in Fig. 4.

Additionally, it should be considered that the strength and reformation of flocs are not only controlled by the G value but also by the characteristics of the different coagulants. For instance, the higher strength and therefore low break-up behavior of PSI flocs under high exposure to increased shear can be attributed to the activated silica in PSI. Because, it is known that activated silica improves the bonding properties of PSI [22].

Table 1
Strength and recovery factor of flocs formed by PSI and PACl with different *G* values

G value (s^{-1})	Floc diameter (d_{50}) (μm)		Strength factor (%)		Recovery factor (%)	
	PSI	PACl	PSI	PACl	PSI	PACl
200	341.3	284.5	51.0	62.2	0.0	0.0
390	346.3	287.6	48.3	57.1	0.0	0.0
470	349.8	95.3	45.6	72.7	0.0	0.0
640	105.3	67.2	77.2	72.0	0.0	0.0
738	103.6	69.4	77.6	73.6	13.1	0.0
830	113.3	64.8	76.9	70.5	18.3	20.0
940	108.9	66.5	78.4	72.0	38.3	65.7
1,110	109.6	67.2	78.7	72.7	11.7	45.4

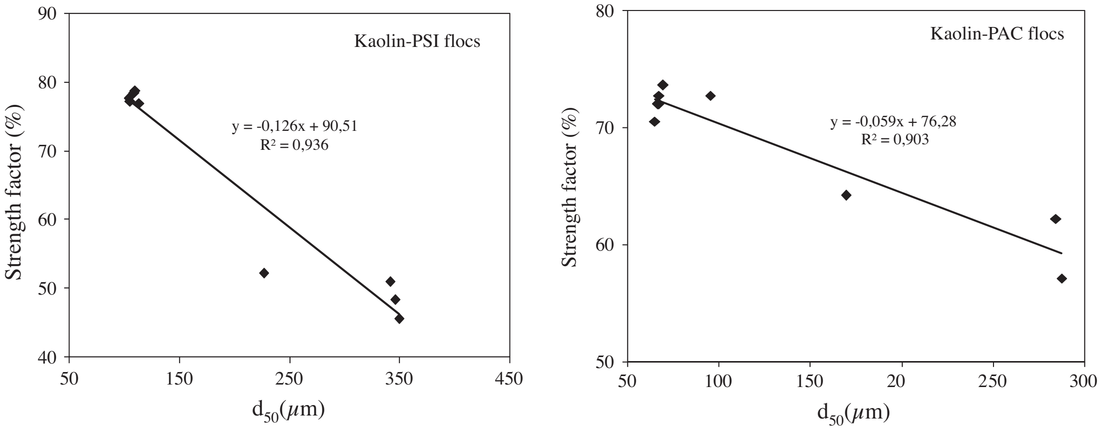

Fig. 4. Relationship between the strength factor and floc size.

3.3. *Effect of rapid mixing time on the floc property of kaolin system*

In order to determine the optimum rapid mixing time for growth of flocs, the particle-size distribution during rapid mixing was analyzed for mixing times from 30 to 180 s. Fig. 5 shows the effect of rapid mixing time on the formation of kaolin floc with PSI and PACl. After mixing for 30 s, the analysis of the floc particle-size distribution shows that a peak was

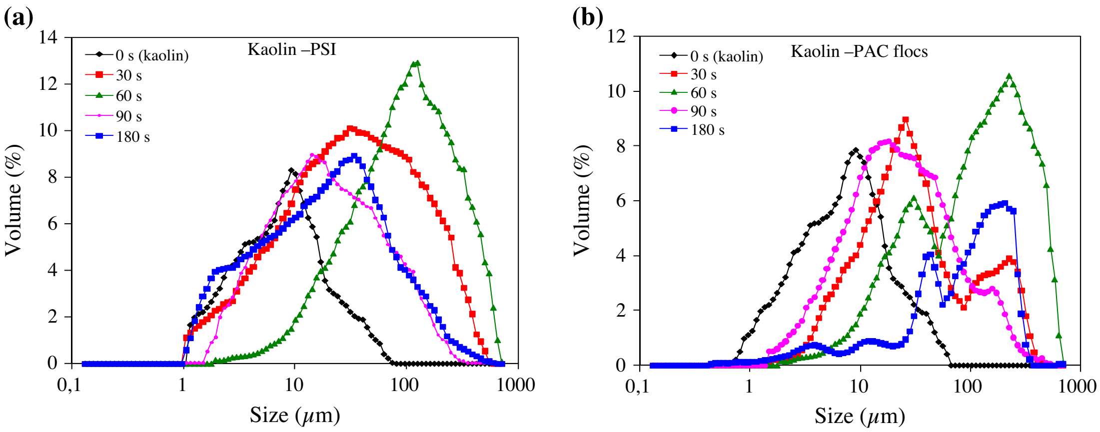

Fig. 5. Kaolin floc-size distributions for different rapid mixing times.

observed in the smaller floc size between 10 and 100 μm for PSI. When the rapid mixing time was increased from 30 to 60 s, the peak shifted to the right, and the major peak coincided with the formation of the largest flocs, with sizes between 100 and 1,000 μm. The volume of the kaolin-PSI flocs reached a maximum particle-size distribution of 12.87% by a mixing time of 60 s. When the rapid mixing time was over 60 s, the major peak was considerably smaller, and the shift to the left was due to the breakage of the flocs. There was also a considerable increase in the smaller peak at floc sizes between 10 and 100 μm. The volume of the kaolin-PSI flocs decreased to a particle-size distribution of 8.89% at a rapid mixing time of 180 s. Similar results were also obtained for kaolin-PACl flocs. However, two different peaks were observed at different floc sizes for each rapid mixing time. The maximum volume of kaolin-PACl was slightly lower than kaolin-PSI. The volume of kaolin-PACl flocs reached a maximum particle-size distribution 10.56% at a mixing time of 60 s. Therefore, a rapid mixing time of 60 s is recommended for the formation of both larger kaolin-PSI flocs and kaolin-PACl flocs.

4. Discussion

Coagulation experiments were conducted in order to characterize the floc formation ability of the kaolin system with PSI and PACl at different rapid mixing conditions. In the literature, there are rather contradictory recommendations for the optimal rapid mixing parameters. Some authors have suggested instantaneous mixing based on the contact-destabilization theory, whereas others have recommended a mixing time of a few minutes [6,23].

In this study, it was noted that the floc size of the kaolin system first increased and then decreased with increased *G* in the coagulation process, which demonstrated that there were three different ranges for floc formation: the low-velocity gradient range ($G < 200\,\mathrm{s}^{-1}$), which was the aggregation-dominated range; the mean-velocity gradient range ($G = 546\,\mathrm{s}^{-1}$ for kaolin-PSI flocs and $G = 390\,\mathrm{s}^{-1}$ for kaolin-PACl flocs), where coagulation rates were maximized and breakup was minimal; and the high-velocity gradient range ($G > 546\ \mathrm{s}^{-1}$ for kaolin-PSI floc and $G > 390\ \mathrm{s}^{-1}$ for kaolin-PACl flocs), where floc breakup was dominant. In the aggregation-dominated range, the aggregate size increased in proportion to the share rate, but resulted in smaller flocs due to the low–particle-collision rates. Although breakup most likely occurred, the breakage was considered to have relatively insignificant impact on the overall aggregate sizes. In the mean-velocity gradient range, a balance between the rate of aggregation and the rate of breakage was reached; the flocs reached their maximum size. In the breakup dominant range, collisions between the flocs produced more, but smaller flocs. In the beginning of this range, the size of the flocs in the kaolin system significantly decreased due to the large-scale fragmentation caused by tensile stress. The kaolin flocs could not re-growth to anywhere near their previous size.

The velocity gradient range used in this study was slightly higher than that used in earlier studies. In addition, the velocity gradient which provided the largest floc formation for the kaolin system was also slightly higher than suggested in previous studies. In previous studies, a *G* value ranging between 180 and 400 rpm has been recommended for the kaolin system [24–26]. However, here we showed that the maximum floc formation for the kaolin system occurred at *G* values ranging between 390 and 546 s^{-1}. Therefore, in contrast to previous studies, a relatively high velocity gradient is suggested for the coagulation process in drinking water treatment.

Kan et al. [23] suggested that the rise in residual turbidity during coagulation when time of rapid mixing time was increased was caused by the breakage of flocs. In this work, it was observed that although the long rapid mixing time increased the number of collisions between flocs, the nature of the floc surface is changed by the repeated breakage and reconnection of floc fragments [26], which causes lower collision efficiency. Therefore, after longer rapid mixing times, there were fewer large flocs and more small flocs. Thus, the size of the re-grown flocs was smaller the flocs before breakage and the re-grown flocs are almost all the same size. In this study, it was noted that the volume of formed kaolin flocs first increased and then decreased with the increased rapid mixing time. The kaolin floc volume was maximized at the rapid mixing times of 60 s with PACl and PSI, respectively, where the formed floc size was mostly distributed in the range between 100 and 1,000 μm. From these results, it can be concluded that a rapid mixing time of between 60 s is suitable for raw water treatment.

5. Conclusions

This is the first study to determine the effect of different velocity gradient and rapid mixing time on coagulation and floc properties, using PSI and PACl under different rapid mixing speed and time. For this purpose, the effect of the *G* values and rapid mixing time was investigated. Several jar tests were performed using various combinations of G values and mixing times, and the main conclusions are as follows.

(1) Three different zones were found to be dependent on the G value in the coagulation process. At low G values, aggregation dominated over breakup. At mean G values, flocs formation was maximized, producing the largest flocs. For large G values, breakup dominated over floc formation. In the last case, an increase in the shear rate caused a reduction in the mean particle size (d_{50}).

(2) A longer rapid mixing speed caused a decrease in the volume of formed flocs. Although the rapid mixing time influenced coagulation efficiency, the size of the re-grown flocs after breakage was nearly the same. A rapid mixing time of 60 s is recommended for kaolin systems.

(3) The ability of the kaolin flocs with PSI to withstand shear in the jar tester was higher than the kaolin flocs with PACl flocs.

(4) The re-growth of flocs was not only controlled by the G value or the rapid mixing time but also by the characteristics of the different coagulants.

(5) The G value range which provided maximum floc formation was between 390 and $546\,s^{-1}$ for the kaolin system. Therefore, in contrast to previous studies, relatively high-velocity gradients are suggested for the coagulation process in drinking water treatment.

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