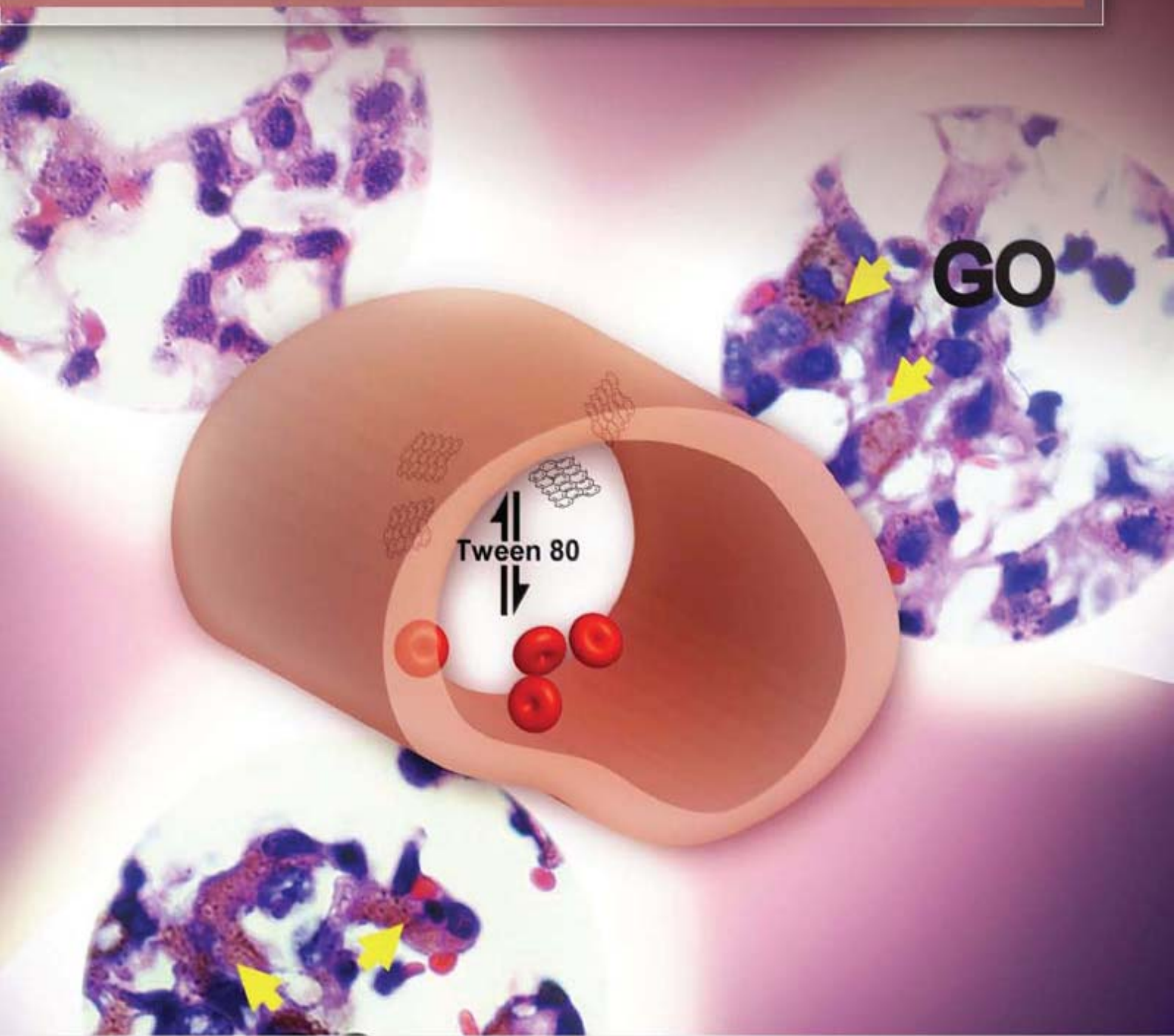


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## Influential factors of formation kinetics of flocs produced by water treatment coagulants

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### Abstract

The growth rate and size of floc formation is of great importance in water treatment especially in coagulation process. The floc formation kinetics and the coagulation efficiency of synthetic water were investigated by using an on-line continuous optical photometric dispersion analyze and the analysis of water quality. Experimental conditions such as alum dosage, pH value for coagulation, stirring intensity and initial turbidity were extensively examined. The photometric dispersion analyze results showed that coagulation of kaolin suspensions with two coagulants (alum and polyaluminium chloride) could be taken as a two-phase process: slow and rapid growth periods. Operating conditions with higher coagulant doses, appropriate pH and average shear rate might be particularly advantageous. The rate of overall floc growth was mainly determined by a combination of hydraulic and water quality conditions such as pH and turbidity. The measurement of zeta potential indicates that polyaluminium chloride exhibited higher charge-neutralizing ability than alum and achieved lower turbidities than alum for equivalent Al dosages. Under the same operating conditions, the alum showed a higher grow rate, but with smaller floc size.

**Key words:** floc; flocculation index; growth rate; kinetics

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### Introduction

Coagulation-flocculation, based on hydrolyzing metal salts, is a well-established treatment technology with a wide range of applications in drinking water and wastewater treatment (Choksuchart et al., 2002; Bes-Pia et al., 2005; Harrelkas et al., 2009; Harif et al., 2012). It is popular as a process for the removal of suspended colloid particles, natural organic matter and wastewater turbidity (Jarvis et al., 2005). Generally, there are two primary coagulation mechanisms, charge-neutralization coagulation and enmeshment or sweep coagulation (Yukselen and Gregory, 2004). The hydrolyzed metal ions, such as 'alum' or aluminum sulfate, produce numerous positive intermediate polymeric species in test suspension before precipitation of the hydroxide, which can adsorb negatively charged particles and destabilize their charge leading to charge-neutralization coagulation. Sweep coagulation occurs to form an amorphous precipitate hydroxide and enmeshed

particles.

In the last few decades, advances in analytical approach and measuring technology have made it possible for researchers to understand many aspects of coagulation. Several authors have found that the coagulation processes are influenced by coagulant type and dosage, solution pH, mixing intensity and particle concentration (Chakraborti et al., 2003; Wang et al., 2011; Yu et al., 2011; Zhao et al., 2012). Another significant factor influencing coagulation/flocculation performance is initial turbidity. Quantitative study about the effects of initial turbidity on floc growth rate may be able to provide available information for a water-treatment. However, those studies focused on the effects of initial turbidity on floc growth rate have not been well established yet (Wu et al., 2012).

Generally, the aggregates formed by coagulation are known as flocs (Xu et al., 2010). Floc growth rate depends on not only the characteristics of coagulants, water temperature, water quality parameters (e.g., pH, ionic strength, total hardness, the concentrations of certain anions and cations,

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and the concentrations of particles), but also various coagulation conditions, such as coagulant dosage and stirring condition. A few publications, so far, were mainly focused on the impact of some factors, especially coagulant dosage and mixing intensity, on the floc formation kinetics of alum species. However, the effects of parameters have not been studied systematically and roundly yet. Therefore, in order to form larger, more compact and higher strength flocs, study on kinetics of floc growth rate is very important. The main goal of the present study is to further investigate the effect of alum dosage, coagulation pH, stirring strength and initial turbidity on floc formation kinetics in clay suspensions, using aluminum sulfate and polyaluminum chloride (PACl) as coagulants. General information concerning the kinetics of floc growth rate using a photometric dispersion analyzer (PDA) is provided.

## 1 Materials and methods

### 1.1 Suspension

Kaolin clay (Tianjin, China) was used as the synthetic suspension. The stock suspension was prepared in a way similar to that of Yukselen and Gregory (2004). The top 600 mL was decanted and its solid content was determined gravimetrically and found to be 60.55 g/L. This was diluted to give a final solid content of 50 g/L.

For the flocculation tests, the stock suspension was diluted using tap water in Guangzhou, China, to give a clay concentration of 50 mg/L. Guangzhou tap water has soft total hardness (ca. 100 mg/L as  $\text{CaCO}_3$ ), alkalinity (ca. 115 mg/L as  $\text{CaCO}_3$ ) and a pH of 7.4. Coagulating pH of the test solution was maintained at 7.3 in most cases. Prior to the addition of alum, an appropriate dose of 0.1 mol/L HCl or 0.1 mol/L NaOH was added to achieve the target coagulation pH of 5.3, 5.9, 6.3, 6.9 and 7.6. The diluted samples had an initial turbidity of 45 NTU determined by a turbid meter (2100P, HACH Company, USA). The temperature of all test solution was  $(21 \pm 2)^\circ\text{C}$ .

### 1.2 Coagulants

Aluminum sulfate hydrate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , analytic reagent) and PACl (30 weight percent as  $\text{Al}_2\text{O}_3$ , the basicity of which was 76.99%) was used. Stock alum solutions were prepared at a concentration of 0.1 mol/L as Al. These reagents were kept in refrigerator at  $4^\circ\text{C}$  and renewed every two weeks.

### 1.3 Apparatus

Experiments on the floc formation kinetics were performed using an on-line continuous photometric dispersion analyzer (I-PDA, Econovel Company Ltd., Korea) in a modified jar test procedure. The experiment process of model suspension was similar to that of Yukselen and Gregory (2004). The average transmitted light intensity (dc value) and the root mean square (rms) value of the

fluctuating component were measured. The ratio (rms/dc) provided a sensitive measure of particle aggregation (Gregory and Nelson, 1986). In this study, the ratio value was called flocculation index (FI). The FI value was highly correlated with mean floc size and always increased as flocs grow larger (Gregory and Chung, 1995). Although it was not possible to attain quantitative information on floc size in the present system, the FI value did provide a very useful relative indication of floc formation in a semi-empirical manner.

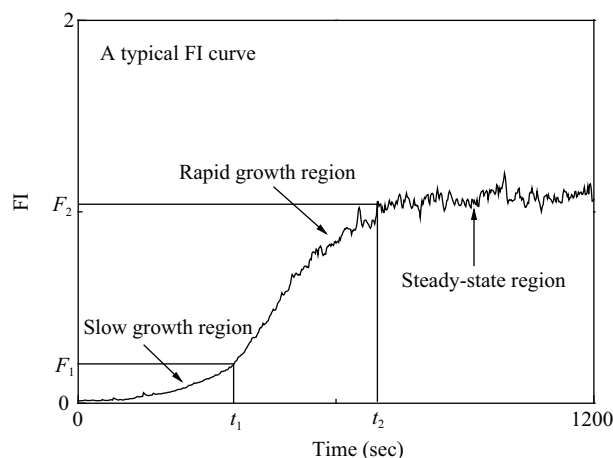
The floc growth rate was calculated to analyze the data collected during the flocculation experiment to explain floc formation kinetics (Wang et al., 2009). As shown in **Fig. 1**, the FI curve could be categorized into three regions: slow growth region, rapid growth region and steady-state region. In the slow growth region, coagulants came to contact and react with small particles, but floc growth rate was so slow that the FI value increased only a little. The rapid growth region described the stage where the floc size increased significantly. The steady-state region depicted a dynamic balance between floc growth and breakage. It has been generally accepted that floc growth was held in check by floc breakage so that the rate of floc growth was considered between floc formation and floc breakage (Spicer and Pratsinis, 1996; Ducoste and Clark, 1998; Biggs and Lant, 2000).

The changing rate of FI value in the initial stage of slow flocculation process is much slower than that in the secondly rapid stage (**Fig. 1**). Consequently, it seems that floc formation of the synthetic suspension can be taken as a two-phase process, and the growth rate ( $R$ ) of the two phases can be determined by

$$R_1 = \frac{F_1 - F_0}{t_1} \quad (1)$$

$$R_2 = \frac{F_2 - F_1}{t_2 - t_1} \quad (2)$$

where,  $R_1$  and  $R_2$  are growth rate of the initially slow grow the region and the secondly rapid grow the region, respectively.  $t_1$  is the first inflexion point on the FI curve and  $t_2$  is



**Fig. 1** Floc growth rates from the typical flocculation index (FI) distribution curve.

the moment when the FI value ends the growth and reaches the steady-state region.  $F_1$  and  $F_2$ , correspondingly, are the values of FI at  $t_1$  and  $t_2$ .  $F_0$  is the initial FI value at  $t = 0$ . The growth of FI value between  $t_1$  and  $t_2$  is nearly linear.

#### 1.4 Procedure

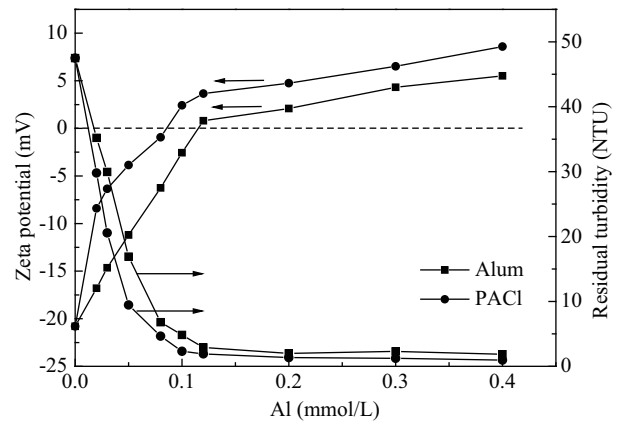
To characterize the relationship between alum dose and settling performance, jar tests were conducted with the synthetic water (model ZR4-6, Shenzhen, China). A two-stage mixing process (30 sec at 400 r/min,  $G = 344.6 \text{ sec}^{-1}$ ) and flocculation (10 min at 50 r/min,  $G = 26.9 \text{ sec}^{-1}$ ) were followed by a 30 min settling period, subsequently supernatant samples were collected and analyzed for turbidity. The  $G$  values represent average shear rates in the test suspensions and were calculated according to the literature (Mejia and Cisneros, 2000). Zeta potential (Zetasizer, Nano series, Malvern Instruments, UK) of the samples were measured after the rapid mix stage of approximately 30 sec, and all measurements were conducted in triplicate on unfiltered samples. The general accuracy of the Zetasizer was determined using standard solutions (Malvern Instruments, UK).

For dynamic tests, the samples during coagulation passed through the photo-detector of the PDA by the peristaltic pump at 25 mL/min, and then returned to the beaker. After the FI value reached an initial steady value, coagulant was added into the suspension, readings were taken every 2 sec. The standard test procedure was modified as follows. After 30 sec rapid mix phase 400 r/min ( $G = 344.6 \text{ sec}^{-1}$ ) for homogenizing water samples, coagulant was dosed into the samples, then the suspension were rapidly mixed for 2 min at 200 r/min, followed by a slow mixing for 10–20 min at 50 r/min ( $G = 26.9 \text{ sec}^{-1}$ ). In order to investigate the impact of mixing condition on floc formation kinetics, the suspension was stirred for 15 min at the required speed (from 30 r/min ( $G = 10.6 \text{ sec}^{-1}$ ) to 250 r/min ( $G = 183.1 \text{ sec}^{-1}$ )).

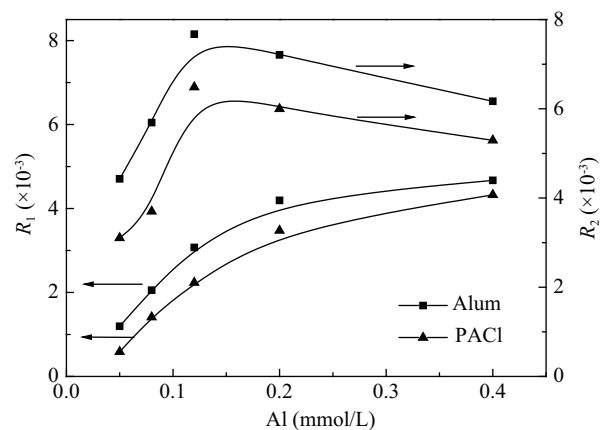
## 2 Results and discussion

### 2.1 Effect of coagulant dosage

Previous study had demonstrated that a link between zeta potential and coagulation performance whereby residual turbidity were both low and stable when the zeta potential values were maintained between  $-10$  and  $+3$  mV (Sharp et al., 2006). In present study, the effect of coagulant dosages on coagulation performance was examined as shown in **Fig. 2**. The value of zeta potential of kaolin suspensions reversed from negative to positive charge and the residual turbidity dramatically decreased with increasing dosage. It could be also seen that charge neutralization ability of PACl was higher than that of alum. This result can be explained from the change of zeta potential of particles after added coagulants. At the dose 0.12 mmol/L for two coagulants, particle destabilization due to charge neutralization oc-



**Fig. 2** Variation of zeta potential and residual turbidity on different coagulant dosages. Coagulant: 0.02–0.4 mmol/L as Al, pH = 7.3.

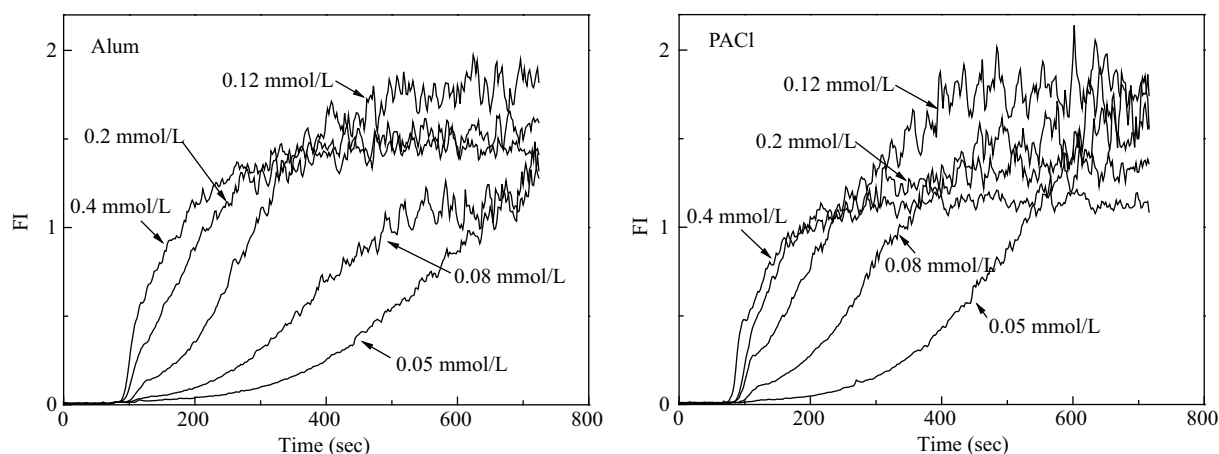


**Fig. 4** Effects of coagulant dosage on floc growth rate ( $R_1$  in slow growth period, and  $R_2$  in rapid growth period). Coagulant: 0.05–0.4 mmol/L as Al, pH = 7.3.

curred, zeta potential of 0.801 mV (produced by alum) resulted in a turbidity residual of 2.9 NTU, whereas, the PACl sample produced a lower turbidity of 1.92 NTU though a higher value of zeta potential is 3.0 mV.

**Figure 3** shows the effects of coagulant dosage on floc formation kinetics for two coagulants (alum and PACl) at the different dosages (0.05–0.4 mmol/L as Al). Analysis of the growth profile revealed an increase in floc size, as indicated by the FI value, with the zeta potential changing from positive to negative values. The FI results indicated that at coagulant dose 0.12 mmol/L as Al, the flocs grew to a limiting size. However, when the added coagulant dosage was higher than 0.2 mmol/L, the FI curves showed a different pattern, i.e., the FI value increased rapidly, but attained lower value. Yu et al. (2010) also discovered that there is a tendency for the FI plateau value to decrease slightly when the alum dosage increases above 0.2 mmol/L Al. The FI plateau value with PACl was higher than that with alum (**Fig. 3**).

**Figure 4** displays the relation of coagulant dose with floc growth rate in coagulation process with alum or PACl. As shown in **Fig. 4**, coagulant dosage exerted a significant effect on  $R_1$ , i.e., the rate of the initially slow growth. As a



**Fig. 3** Dynamic monitoring of flocculation at different dosages. Coagulant: 0.05–0.4 mmol/L as Al, pH = 7.3.

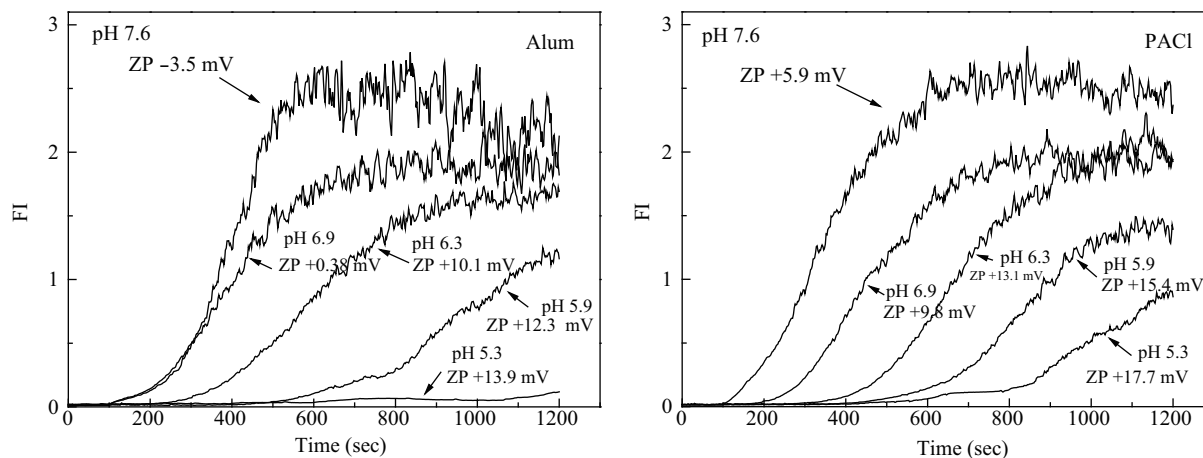
general trend, the values of  $R_1$  increased as coagulant dose increased, and the  $R_1$  values reached higher with alum than that with PACl. On the other hand, coagulant dose also had obvious impact on  $R_2$ , i.e., the rate of the rapid growth. For alum and PACl, the change of  $R_2$  as a function of coagulant dosage clearly showed a maximum in this case. When coagulant dosage was 0.12 mmol/L, the  $R_2$  of alum and PACl reached to the peaked values, which demonstrated that it took less time for floc aggregation to attain the plateau FI value (**Fig. 4**). The trend may be explained. Under the same shear condition for the coagulant of same synthetic water, different floc growth profiles were direct reflection of different coagulation characteristics with different hydrolyzed Al species distribution.

## 2.2 Effect of initial pH

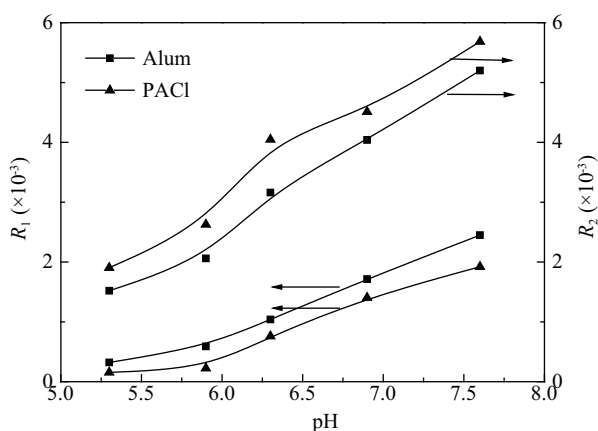
One of the most important factors that affect coagulation is pH (Xie et al., 2012). In order to get deeper insight into the effect of pH on the dynamics of floc formation, jar tests with different pH control were carried out. It can be seen from **Fig. 5** that the values of zeta potential of floc particles decreased with increasing solution pH. For alum coagulant, the zeta potential changed from +13.9 mV (pH

5.3) to  $-3.5$  mV (pH 7.6). In contrast, for PACl, a pronounced reduction was observed when the zeta potential was reduced from +17.7 mV (pH 5.3) to +5.9 mV. Since the magnitude and sign of the zeta potential was in relation to the floc formation, it can be seen from **Fig. 5** that the FI plateau values increased and the time required for attaining FI plateau values decreased with increasing solution pH under given stirring condition. For PACl and alum, at pH 5.3 the FI plateau values was very low, the time required for attaining FI plateau values were 20 min or so. However, for samples controlled at pH 7.6, the FI plateau values was 2.5 or so, the time required for attaining FI plateau values were only 8 min or so. At the same time, it is found that the FI plateau values with PACl were almost equal to or higher than those with alum though all the values of zeta potential of flocs with PACl, corresponding to solution pH, were higher than those with alum. It was revealed that the adsorb-bridging with PACl played an important role in coagulation process besides charge neutralization.

**Figure 6** shows the effect of pH on floc growth rate. It can be seen from **Fig. 6** that  $R$  increased with increasing pH values. It can be partially explained by the zeta potential of floc particles. At lower pH level, the zeta



**Fig. 5** Dynamic monitoring of flocculation at different pH levels. Coagulant: 0.12 mmol/L as Al, pH 5.3–7.6. ZP: zeta potential.



**Fig. 6** Effects of pH on floc growth rate. Coagulant: 0.12 mmol/L as Al, pH 5.3–7.6.

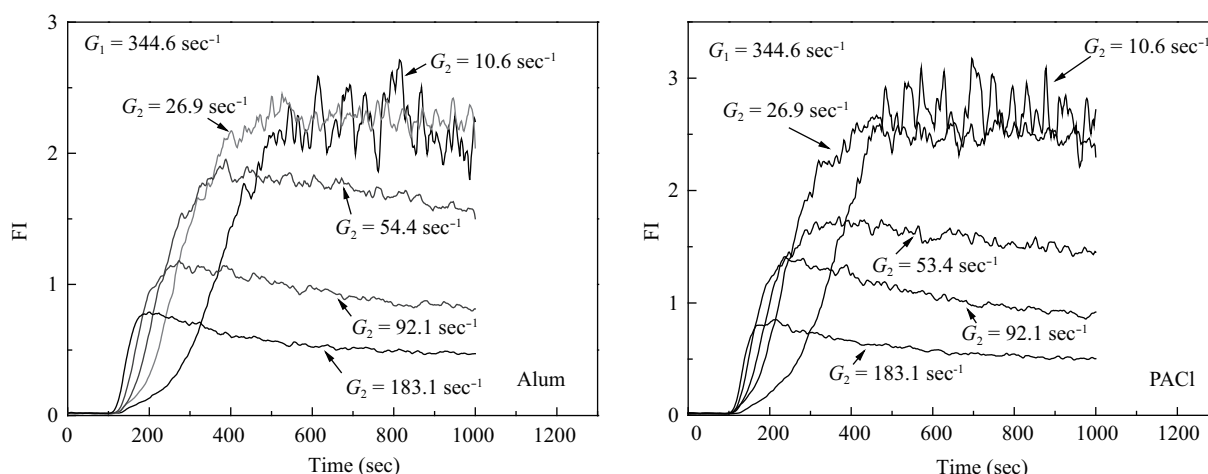
potential of floc particles remained higher (**Fig. 5**), which led to lower collision efficiency in coagulation process, the precipitation was gradually formed and  $R$  was very small. When the precipitated hydroxide particles reached a certain size, the surface precipitation began and caused rapid aggregation. With increasing pH, the zeta potential of floc particles decreased rapidly, precipitate formation was accelerated and the rate of second phrase of flocculation was increased markedly, i.e.,  $R_2$  were increased, too. The optimum coagulation pH value for the faster floc growth rate of synthetic water was about 7.6 or more. It seems that the initially precipitated hydroxide particles could aggregate rapidly to form larger particles at the optimum pH value. For alum samples, the aggregated precipitate formed from positively charged patches to the negatively charged surfaces of the particles. Attractive forces between a patch and an oppositely charged surface as particles collide might account for the rapid rise of the FI curves. But for PACI samples, a significant “patch coagulation” could occur, as suggested from coagulation with cationic polymers (Wang et al., 2002).

### 2.3 Effect of stirring condition

**Figure 7** shows the effect of slow-mixing intensity on floc size (FI value) of kaolin suspensions for alum and PACI with the same dosage of 0.12 mmol/L as Al. Coagulation tests were undertaken on a jar tester as described above. Following the 30 sec rapid-mixing stirring phrase (400 r/min,  $G_1 = 344.6 \text{ sec}^{-1}$ ), the impact of increased shear on floc growth rate was investigated by varying mixing rate (with average velocity gradient) of: 30 r/min ( $G_2 = 10.6 \text{ sec}^{-1}$ ), 50 r/min ( $G_2 = 26.9 \text{ sec}^{-1}$ ), 100 r/min ( $G_2 = 53.4 \text{ sec}^{-1}$ ), 150 r/min ( $G_2 = 92.1 \text{ sec}^{-1}$ ), and 250 r/min ( $G_2 = 183.1 \text{ sec}^{-1}$ ) for 15 min.

It can be seen from **Fig. 7** that  $G_2$  was related to the FI value (floc size) and the FI value attained a maximum value at  $G_2 10.6 \text{ sec}^{-1}$  (30 r/min) for two coagulants (alum and PACI). Beyond this value ( $G_2 10.6 \text{ sec}^{-1}$ ), there is not much further effect on floc size, though the FI values increased rapidly within a shorter time. Increased  $G_2$  gave higher particle collision efficiency but flocs were subject to greater shearing force. When  $G_2$  for shear equals to  $26.9 \text{ sec}^{-1}$  or higher, the FI values attained a maximum value and then declined, even if the slow stirring remained constant. It is well known that a limiting floc size is found at a given shear rate (Torres et al., 1991). Usually, a constant floc size is found under given slow stirring conditions, representing a balance between floc growth and breakage. Yu et al. (2011) also found that increasing the rapid mixing time led to a decrease in the final floc size. This suggested that the increased shearing energy was sufficient to cause floc breakage. It can be also seen from the figure that the flocs with PACI had a larger FI plateau value than those with alum, corresponding to a larger floc size.

The growth rate calculations showed that the floc size at high  $G_2$  value was smaller (**Fig. 7**) but with much higher coagulation rate especially  $R_1$  for alum and  $R_2$  for PACI (**Fig. 8**). Increasing the slow-stirring intensity within the



**Fig. 7** Dynamic monitoring of flocculation at different stirring conditions.  $G_1$  and  $G_2$ , respectively, were the average velocity gradient of rapid mixing and slow mixing. Coagulant: 0.12 mmol/L as Al, pH = 7.3.



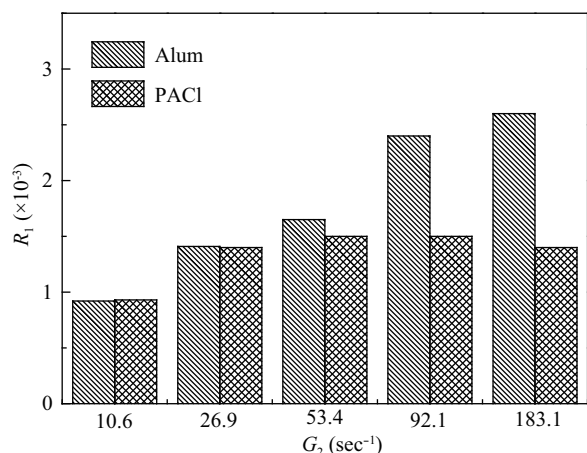
range of 10.6–183.1  $\text{sec}^{-1}$ , markedly influenced the growth rates. As shown in **Fig. 8**,  $R_1$  was not significantly differ as  $G_2$  increased, but the increase of  $G_2$  displayed a greatly dramatic effect on  $R_2$ . The improved  $R_1$  with the change of  $G_2$  did not obviously differ between PACI and alum. However, the  $R_2$  of alum could not reach the  $R_2$  of PACI with the change of  $G_2$ , suggesting that PACI played more positive effect on the  $R_2$ . The reasons were speculated for the results: (1) the variation of the PDA output FI value reflected actually the change in floc size; (2) the collision efficiency of particles became higher as slow stirring intensity increased and became lower as particles size increased. To summarize the formation and breakage of flocs, flocculation test solutions were governed by the prevailing shear conditions and would reach a steady state. When the stirring rate increased above a critical level, flocs would break until a new steady state was reached.

#### 2.4 Effect of initial turbidity

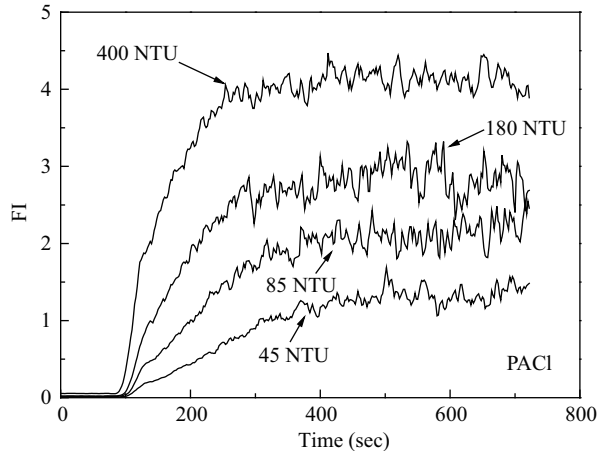
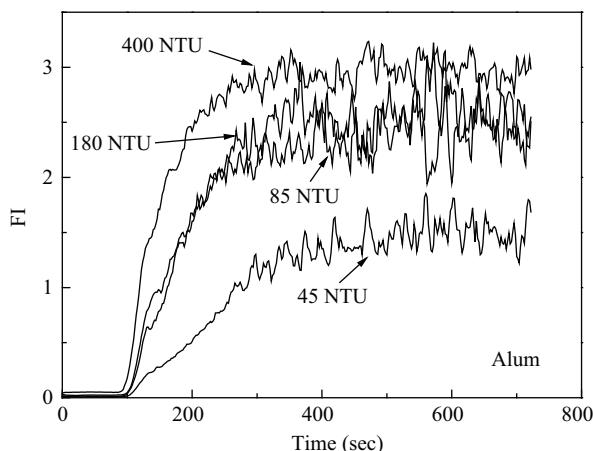
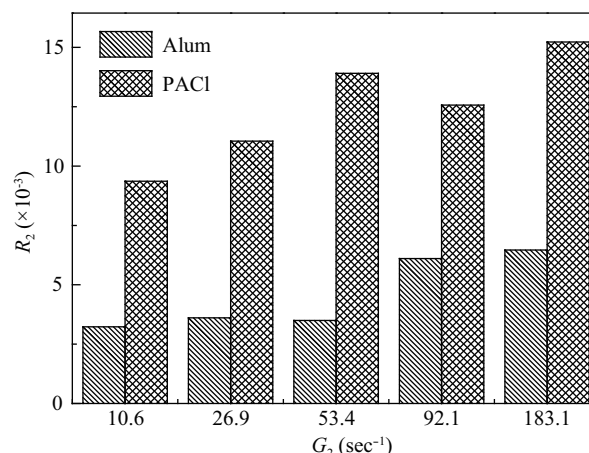
The kaolin suspensions with different initial turbidity (i.e., 45, 85, 180 and 400 NTU) were prepared and the effects of initial turbidity on the kinetics of floc formation were investigated. **Figure 9** indicates that the suspension turbidity

ranging from 45 to 400 NTU exerted significant effects on the steady-stage FI values. The FI values increased rapidly with increasing suspension initial turbidity. Furthermore, the higher the test solution turbidity was, the higher the plateau FI value reached for the two coagulants. The FI curves demonstrated that at the initial turbidity of 400 NTU with a set coagulant dose of 0.12 mmol/L as Al, the flocs grew to a limiting size ( $FI_{\text{max}}$ ) and there were great differences of  $FI_{\text{max}}$  among the different initial turbidities. Moreover, the FI values with PACI attained higher than that with alum and the maximum for PACI was more pronounced.

The initial turbidity also exerted a dramatic influence on growth rates, as shown in **Fig. 10**. The increase of the raw water turbidity in the range of 45–400 NTU significantly improved the  $R$  values with alum and PACI. Initial turbidity had no significantly effect on  $R_1$ , but the  $R_1$  values with the alum were larger than those with the PACI as initial turbidity increased; on the other hand, the  $R_2$  values with the PACI were sharply increased with initial turbidity increased. However, **Fig. 10** does not show excellent relation between the  $R_2$  values and the initial turbidity for alum.



**Fig. 8** Effects of mixing intensity on the growth rate. Coagulant: 0.12 mmol/L as Al, pH = 7.3.



**Fig. 9** Dynamic monitoring of flocculation at different initial turbidity of 45–400 NTU. Coagulant: 0.12 mmol/L as Al, pH = 7.3.

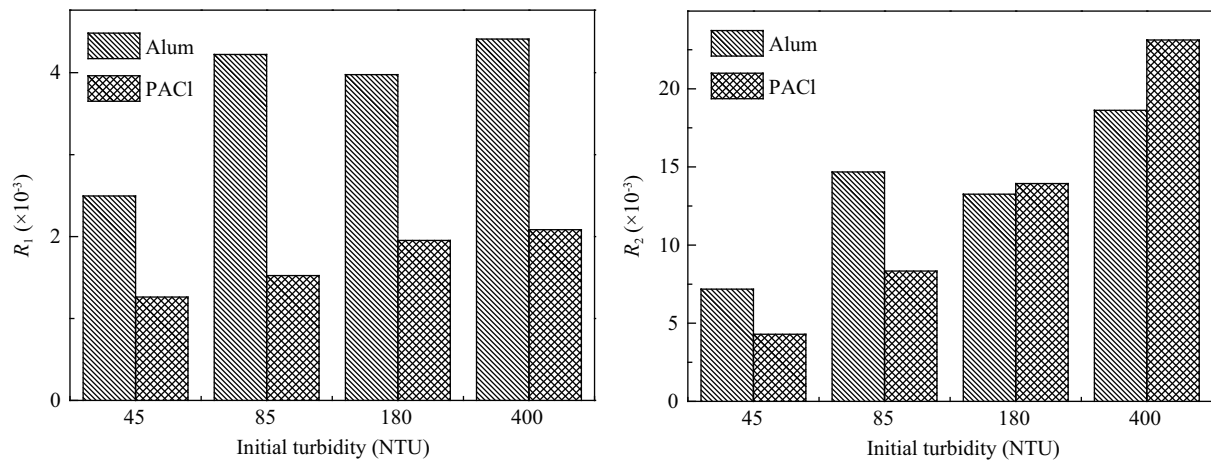


Fig. 10 Effects of initial turbidity on the growth rate. Coagulant: 0.12 mmol/L as Al, pH = 7.3.

### 3 Conclusions

Based on the experiments reported here, it can be safely concluded: (1) The coagulation of kaolin particles can be taken as a two-phase process, involving the slow growth of small flocs and then the rapid growth into larger flocs based on the different growth rates and FI values by the PDA analysis. (2) The coagulant dosage exerted a significant effect on floc formation. The FI plateau value and the removal efficiency of turbidity with PACI are higher than those with alum at 0.12 mmol/L as Al, although the alum exhibited a more rapid floc growth rate. (3) For PACI and alum, the FI values increased with increasing solution pH (5.3–7.6). The FI values with PACI still were very high though zeta potential of floc particles was away from zero. (4) Higher or lower velocity gradients for flocculation will not be favorable to improve FI plateau values though their initial FI values increased rapidly within a shorter time. Moreover, initial turbidity exerted significant effects on the steady-stage FI values.

Dynamic monitoring of floc formation gives much more detailed information on the process than a simple jar test procedure. The findings cannot be adequately explained on the basis of current models, further studies on other systems should be fruitful. This aspect should be analyzed in more detail in future work.

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