



High-concentration polyaluminum chloride: Preparation and effects of the Al concentration on the distribution and transformation of Al species

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ABSTRACT

Using an instantaneous base-feeding method and a batch-feeding method, high-concentration polyaluminum chloride (PACl) products were prepared employing CaO as the basification reagent. The distribution and transformation of the Al species of PACl was explored for the first time over a broad total Al concentration (Al_T) range. There is always an optimal basicity (B value) at which the product has the highest Al_b content, whether Al_T is high or low. The optimal B value and the highest Al_b content obtained both decrease with increasing Al_T . When the B value is lower than optimal, the overall reaction mainly leads to the production of Al_b ; when the B value is higher, the reaction leads to the production of Al_c . During the dilution process, high B value PACl has a tendency towards Al_b formation, while low B value PACl favors Al_c formation. The transformation behaviors of Al species are influenced by Al_T in the preparation or dilution process.

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1. Introduction

Polyaluminum chloride (PACl) is a common inorganic polymeric coagulant used in the water supply and wastewater treatment process [1–7]. PACl is a dynamic intermediate product in the hydrolysis–aggregation–precipitation process of the Al ion. PACl is composed of several easily intertransformable Al species [8–10]. The coagulation performance of PACl is closely related to the Al species distribution [3,4,11,12]. According to the reaction rates of the Al species with Ferron reagent, the Al species of the Al-based coagulants are usually divided into Al_a (monomers that react with Ferron within 1 min), Al_b (reactive polymers that react with Ferron within 1–200 min) and Al_c (high polymers or colloidal Al species that cannot react with Ferron) [13]; or into Al_m (Al monomer), Al_{13} , and Al_u (which indicates undeterminable Al species using ^{27}Al NMR), based on whether and where the peak appears in the ^{27}Al NMR spectrograph [14,15]. It is commonly recognized that Al_b or Al_{13} is the most effective species for charge neutralization and particle bridging actions in coagulation and flocculation processes [1,16–19]. Their content is closely related to the basicity or OH/Al molar ratio (B value) of the product. Previous investigations show that the content of Al_b increases with an increase in the B value, and reaches a maximum when $B = 2.4$ or 2.5 [6,20,21]. As a result, to increase the Al_b content of the product,

a high B value is necessary. In addition, the total Al concentration (Al_T) of the product is also a factor that cannot be ignored in practical applications. High-concentration PACl can reduce the expense involved in the storage, transportation and dosing processes. Therefore, PACl has commercial value only when it has a high Al_T .

It is generally agreed upon that in the PACl preparation process, a low speed of base-feeding or base-injection is preferred for preparing a stable product with a high B value and Al_b content [8,19,22]. The method of base-microinjection is usually used in the laboratory. Using this method, with NaOH as the basification reagent, PACl can be produced with a high B value ($B = 2.2–2.5$), Al_b content ($>70\%$), and low Al_T ($<0.6 \text{ mol L}^{-1}$) [3,6,16,19]. A large number of previous investigations have proven that it is difficult to produce a stable high-concentration PACl using NaOH as the basification reagent because NaOH releases OH^- in water too rapidly [6,19,23]. The production of stable, high-concentration PACl thus requires other basification reagents. Furthermore, the high-concentration PACl can be obtained when a solid-state basification reagent is used [24]. A basification reagent should have the following qualifications: its solid-state should provide OH^- in $AlCl_3$ solution for the polymerization of Al^{3+} ; the rate of OH^- production should not be too slow or too fast; and the products should have no influence on the properties of PACl. Compared with Na-based basification reagents, Ca-based basification reagents have a low solubility in water. Therefore, Ca-based basification reactions are ideal for reducing the production and release of OH^- . Additionally, the Ca^{2+} produced compresses the electric double layer of the

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colloid particles, which may improve the flocculation performance to some extent [25]. As a result, an attempt should be made to use Ca-based basification reagents to produce high-concentration PACI in the laboratory.

At present, most processes and theories in the PACI research field are based on low-concentration PACI [3,6–11,13–20,22,26]. Whether these theories are adaptable to high-concentration PACI remains to be verified. In the body of practical work, there exist some phenomena that cannot be explained by the present theories. For example, the *B* value of some industrial PACI reaches 2.4, but the content of the Al_b is very low, reaching only about 30%. This cannot simply be attributed to the rough nature of industrial production. Other reasons, such as Al_T , a significant factor, must be taken into account as well.

A dilution process will occur once the coagulant is dosed in water. During this process, the Al_T , the system pH value, and other conditions change. The Al species change with the occurrence of these complicated physicochemical reactions, and these species changes have a direct effect on the entire flocculation process and the flocculation performance. With regard to the high-concentration PACI, these changes may be more complicated than those observed during the low-concentration PACI dilution process.

In this paper, high-concentration PACIs were prepared using CaO as the basification reagent, and the reaction characteristics were analyzed. Using the high-concentration PACI products, this study also explores the effects of the Al concentration and dilution function on the distribution and transformation of Al species. The results of this study will be of great significance in guiding the industrial production and application of these flocculants.

2. Materials and methods

2.1. Preparation

PACI products were prepared using an instantaneous base-feeding method and a batch base-feeding method, respectively. To obtain PACI with different Al_T , 100 mL of $AlCl_3$ solution with different Al concentrations (0.1, 0.2, 0.5, 1.0, 1.5, 2.0, and 2.1 mol L⁻¹) was added respectively to a 250-mL beaker at room temperature. Based on the definition of *B* value (OH/Al molar ratio) and the equation that 1 mol of CaO is equivalent to 2 mol of OH⁻¹, a calculated amount of solid CaO was added instantaneously or in batches to the $AlCl_3$ solution to achieve a target *B* value (for example, to obtain the PACI of Al_T 2.0 mol L⁻¹ and *B* 2.0, 11.2 g of CaO was added to 100 mL of 2.0 mol Al L⁻¹ $AlCl_3$ solution). For the batch addition, the total mass of CaO was divided averagely into five batches, and every 2 h one batch was fed instantaneously. During the experimental process, a magnetic stirring apparatus was used. The reaction was finished when the solid powders disappeared. The Al species measurement and other experiments on the prepared PACI were conducted 24 h after the aging of the product. All the reagents used in the experiment were of analytical grade.

2.2. Al species distribution measurement

The distribution of Al species was measured using Al–Ferron complexation timed spectrophotometry. Ferron reagent was purchased from Sigma–Aldrich (USA). A Ferron colorimetric solution was prepared using a method described previously [27]. High-concentration PACI samples were analyzed and tested directly, or after dilutions and settling for a period of time, according to the prescribed experimental setup. The amount of samples to be tested was determined according to the degree of dilution to ensure that the measured absorbency fell within the linearity range of such method. Ferron solution (10 mL) and the appropriate amount of the test sample were transferred into a graduated glass tube and then diluted to 50 mL. After homogeneous mixing, the sample was quickly transferred into a 1-cm quartz cell. A timed absorbance measurement was carried out immediately at 370 nm and absorbance was then recorded for an additional 2 h with a Specord® 200 spectrophotometer.

The Al species were artificially classified as Al_a , Al_b , and Al_c . The absorbance observed within the first minute of the experiment corresponded to the Al_a species, whereas the absorbance from the second minute up to 120 min corresponded to the Al_b species. Al_c values were calculated by subtracting Al_a and Al_b from the known Al_T [27].

2.3. Size measurement of the Al species

Tap water (1 L) was placed into the sample tank of the laser analysis instrument (Mastersizer 2000, Malvern Co.). The tap water (the raw water is from the groundwater in Peking University) is mainly characterized by pH of 7.7, HCO_3^- of 2.8 mmol L⁻¹, turbidity of 0.5 NTU, Al concentration of 1.3×10^{-3} mg L⁻¹, and undetectable TOC. After a 10-min ultrasonic treatment with stirring at 1800 rpm in the absence of air bubbles in the analysis instrument, the measurement program was started. After the background value was measured, a 0.1-mL sample of the PACI to be tested was added. The size distribution was measured at time intervals of 30 s. Measurement was suspended after 30 min. The particle diameter of the Al species was expressed as a volume-weighted average.

3. Results and discussion

3.1. Effect of the base-feeding method

Using the CaO solid and $AlCl_3$ solution, PACI was prepared via instantaneous base-feeding and batch base-feeding, respectively. The Al species distribution of the products was measured to study the differences between the two preparation methods. The results are shown in Table 1.

As can be seen in Table 1, the two base-feeding methods have nearly no effect on the Al species distribution of the products when high-concentration PACI is prepared. However, when prepar-

Table 1
Preparation of high-concentration PACI via different base-feeding methods.

Preparation method	Reaction time (h)	Al_T (mol L ⁻¹)	Al species distribution (%)			Phenomena in the reaction process
			Al_a	Al_b	Al_c	
Instantaneous base-feeding	20	2.0	23.61	35.46	40.93	No precipitates or gels appeared.
	12	0.2	17.24	71.77	10.99	White precipitates appeared later in the reaction process, and dissolved after the temperature was raised (80 °C).
Batch base-feeding ^a	20	2.0	21.48	36.05	42.47	No precipitates or gels appeared.
	12	0.2	13.19	78.26	8.55	No precipitates or gels appeared.

^a The total mass of CaO was divided averagely into five batches, and every 2 h one batch was fed instantaneously. The scheduled *B* values of all the products above were 2.0.

Table 2
Preparation of PACI with different Al_T using the instantaneous base-feeding method.

Al_T (mol L ⁻¹)	Reaction time (h)	The highest B value	Phenomena in the reaction process	Rate-determining step of the overall reaction
0.1	12	2.6	CaO hydrated relatively fast. Precipitates appeared and dissolved with temperature increasing.	The reaction of Al_b formation.
0.5	14	2.4		
1	18	2.3	CaO hydrated moderately. No precipitates appeared.	The reactions of Al_b formation and CaO hydration.
1.5	20	2.25		
2	26	2.2	CaO hydrated quite slowly. No precipitates appeared.	The reaction of CaO hydration.
2.1	30	2.1		

ing low-concentration PACI, the method of batch base-feeding can be optimized to increase the Al_b content. The time used for preparing low-concentration PACI is obviously shorter than that for high-concentration PACI. When using the method of instantaneous base-feeding to prepare low-concentration PACI, some precipitates appear in the reaction process. These precipitates could be dissolved at higher temperatures. In contrast, no precipitates appeared during the process of preparing the high-concentration PACI.

The reaction between CaO and $AlCl_3$ is the final reaction between the OH^- from CaO hydration and the H^+ from Al^{3+} hydrolysis. Therefore, the rate of OH^- production in water determines the rate of reaction of CaO and $AlCl_3$. The production of OH^- is associated with the hydration of CaO and the dissociation of $Ca(OH)_2$. Compared with NaOH and $Ca(OH)_2$, CaO has a lower rate of OH^- production, which leads to a low rate of reaction between CaO and $AlCl_3$. Moreover, a large amount of energy (63.89 kJ mol⁻¹) is released in the hydrating process of CaO, which leads to an increase in the system temperature. On the one hand, this increase in temperature promotes diffusion of the solute, the uniformity of the system and the production of Al_b from Al_c and Al_a (Reaction (2)), but on the other hand, it inhibits the hydration of CaO and the production of OH^- . This prevents the transformation of the soluble Al_c into the gel precipitate, which occurs in cases of excessively high OH^- production rates.

3.2. Reaction characteristics of the instantaneous base-feeding method

Using the instantaneous base-feeding method and CaO as the basification reagent, stable PACI samples were prepared. The highest B value (it is the highest B value of PACI before precipitation occurred in the preparation process) and the corresponding reaction time at different Al_T values are shown in Table 2. The highest B value reached decreased with an increase in the Al_T value. When $Al_T = 0.1$ mol L⁻¹, the B value reached a maximum of 2.6, whereas $Al_T = 2.1$ mol L⁻¹, the highest B value was only 2.1. Conversely, the Al_T would reach its maximum of 2.1 mol L⁻¹ at the B value of 2.1 only when the stability of the PACI is ensured. Notably, though increasing the Al_T increases the concentration of Al species in the system, it also increases the system viscosity. These two features have opposing influences on the diffusion of Al_c formed at the moment of base-feeding. As a consequence, Al_c cannot react adequately with Al_a , and thus, the formation of the $Al(OH)_3$ gel increases. Consequently, the maximum amount of base that can be fed to the system is reduced, which means the highest B value is decreased.

When Al_T is lower than 0.5 mol L⁻¹, the appearance of precipitates can be distinctly observed. Increasing the temperature can make the precipitates disappear quickly, which means the reaction between Al_c and Al_a to produce Al_b is the rate-determining step of the total reaction. When $Al_T > 1.5$ mol L⁻¹, the hydration of CaO becomes quite slow, leading to an increase in total reaction time. This indicates that, with high Al concentration, the hydration reaction of CaO becomes the rate-determining step. When $0.5 \text{ mol L}^{-1} \leq Al_T \leq 1.5 \text{ mol L}^{-1}$, the two reactions are thought

to reach a balance. As a result, according to the different rate-determining steps, the total reaction can be divided into three regions in the whole Al concentration range: a region controlled by Al_b formation, a region with a balanced reaction, and a region controlled by CaO hydration (Table 2).

3.3. Effect of the Al concentration on the Al species distribution

The distribution of Al species of PACI with different Al concentrations is shown in Fig. 1. With the rise of the B value, the Al_a content of the PACI samples decreased, whereas the Al_b content rose, and then declined after reaching the highest point. The Al_c content increased gradually, and when the B value exceeded the value corresponding to the maximum value of Al_b , the Al_c content began to rise.

Based on the experimental phenomena in the present work and some reports in relevant literatures [19,22,24,28,29], it is assumed that the following reactions took place in the polymerization of aluminum salt forced by base-feeding.



The production of Al_c depended on the OH^- concentration (having a positive correlation with the B value of the products) and Al_T . Reaction (1) mainly occurred at low OH^- concentrations, whereas Reaction (3) was likely when the OH^- concentration was high. The reason for this might be that the activation energy of Reaction (3) is higher than that of Reaction (1). When Al_T is low, the contact probability between the Al_b species decreases because of the repulsive forces between the positively charged Al_b species. Thus, there is little probability of Reaction (4) taking place. When Al_T is high, Al_b species are dehydrated by hydrogen bond association and polymerize with each other to form high-molecular-weight species, which means the probability of Reaction (4) increases. Reactions (3) and (4) might both be exothermic reactions, and therefore, an increase in temperature would promote the reverse reactions.

In regions with low B values, there was little probability of Reactions (3) and (4) taking place, and the main reactions in the system were Reactions (1) and (2) because of the low OH^- and Al_b concentration in the system. With increasing B value, the Al_a content decreased rapidly, and the Al_b and Al_c contents increased substantially in the low Al concentration system; but in the high Al concentration system, the Al species changed relatively slowly. When the B value reached a certain level, the probability of Reactions (3) and (4) occurring increased. This resulted in the maximum content of Al_b and increased Al_c content. Increases in the Al concentration led to the reduction of the maximum Al_b content and the corresponding B value. When the B value increased, Reactions (3) and (4) became the dominant reactions. In this case, Reactions (1) and (2) still occurred, but Reaction (2) proceeded more slowly. In

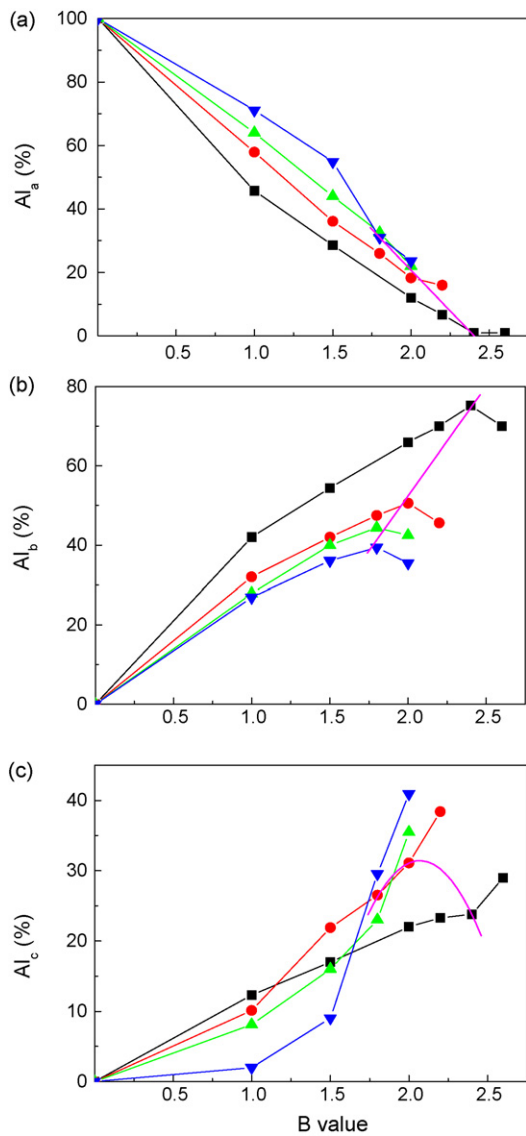


Fig. 1. Effect of B value and Al_T on the distribution of the Al species: (a) Al_a , (b) Al_b , (c) Al_c . $Al_T = 0.1 \text{ mol L}^{-1}$ (■), $Al_T = 0.5 \text{ mol L}^{-1}$ (●), $Al_T = 1.0 \text{ mol L}^{-1}$ (▲), $Al_T = 2.0 \text{ mol L}^{-1}$ (▼), trend-line of Al species at the optimal B value (---).

total, the Al_a and Al_b contents decreased from the maximum, while the Al_c content increased rapidly.

Based on the analysis above, Fig. 1 demonstrates that, whether in a high or low Al concentration system, an optimal B value does exist. The Al_b content was the highest at this B value. The effect of Al_T on the optimal B value and the Al species distribution of the products can be demonstrated using the trend-lines constructed by comparing the Al species contents to the optimal B value at different Al concentrations (Fig. 1). The relationship between the Al concentrations and the optimal B value or the maximum Al_b content is shown in Fig. 2. The optimal B value decreased with the increase in the Al_T , similar to the maximum value of the Al_b content. When the B value was lower than the optimal B value, the overall reaction mainly represented the formation of Al_b ; when it was higher, it represented the formation of Al_c (Fig. 2). In the low Al concentration system, the total reaction involved the transformation of Al_a into Al_b almost in the entire range of B values, since the optimal B value was high. This finding is consistent with most of the research results for the low Al concentration system [6,19]. In the high Al concentration system, the optimal B value was low, and the B value

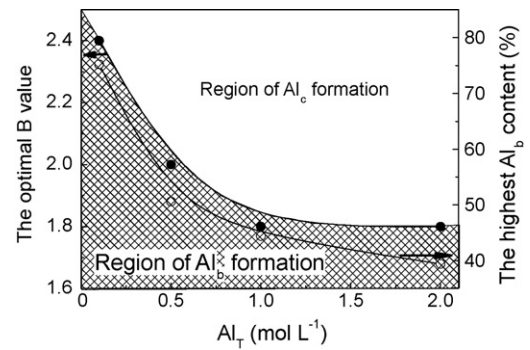


Fig. 2. Effect of Al_T on the optimal B value (●) and highest Al_b content (○) of PACI.

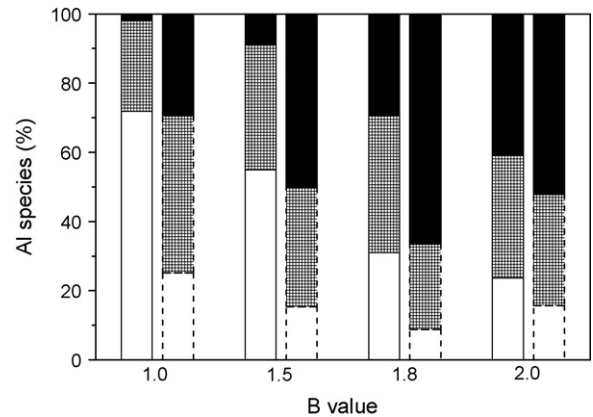


Fig. 3. Effect of dilution on the transformation of the Al species in PACI ($Al_T = 2 \text{ mol L}^{-1}$) with different B values. Al_a (□), Al_b (▨), Al_c (■). Undiluted sample (—), samples diluted 25,000 times (---).

chosen in the study was always higher than the optimal B value. The total reaction involved the transformation of Al_a into Al_c , or of the produced Al_b into Al_c . This is the reason that Al_c content is usually high in commercial PACI products with a high Al_T .

3.4. Effect of dilution on the Al species distribution

Before and after the dilution of PACI samples with different B values, the distribution of the Al species was measured. The distribution of Al species changed after the dilution of PACI, and this distribution differed depending on the B value (Fig. 3). The content of Al_a in the diluted sample decreased, while the content of Al_c increased after dilution. The ratio of Al_c content decreased with the increase in B value. The Al_b content first increased, then remained stable, and then finally decreased with the increase in B value.

When PACI was diluted in water, the species distribution was influenced by two opposing factors, the first being Al_T . According to the theory of coordination chemistry, a decrease in the metal complex concentration leads to a decrease in the polymerization degree of the coordination compounds [30]. The other factor that influences species distribution is the pH value. The pH value of the deionized water for dilution is much higher than that of the PACI sample. Dilution (adding deionized water) is similar to adding base in a sense, as both will increase the polymerization degree of the coordination compounds [8,14,31,32]. The effects of the two factors have a dynamic balance under certain conditions. Whether the polymerization degree of the polyaluminum increases or decreases depends upon which factor is more influential. The results of this paper demonstrate that the polymerization degree of the PACI species increases because of the dilution effect, which shows that the effect of the pH is much more obvious than that of Al_T . This

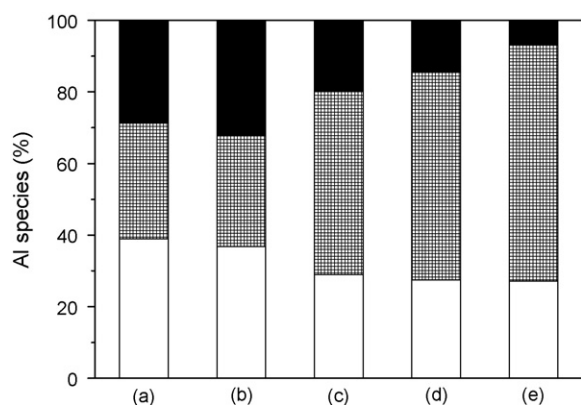


Fig. 4. Effect of the aging time after dilution on the transformation of the Al species in PACI ($B = 1.8$, $Al_T = 2 \text{ mol L}^{-1}$). Al_a (□), Al_b (▨), Al_c (■). Undiluted sample (a), samples aged 0 h (b), 6 h (c), 12 h (d), and 36 h (e), respectively, after diluted 100 times.

is not consistent with other studies. The reason for this difference may be that, after dilution, the aging time used in our study was 2 h, whereas it was greater than 8 h in other studies [30,33]. The effect of Al_T will not be obvious when the aging time is short. In the dilution process, for PACI with a low B value, the Al_b content increases, while for PACI with a high B value, the Al_b content decreases and the Al_c content increases. The Al_b content remains unchanged in the dilution process when PACI had a moderate B value. This means that in the dilution process, PACI with a low B value has a tendency to form Al_b , PACI with a high B value has a tendency to form Al_c , and the Al_b species have some stability when PACI has an optimal B value. Similarly to the results on PACI preparation, it has been found that the B value influences the change of the Al species distribution with the Al concentration. The similarity of the results in these two research areas shows that the dilution process carried out by adding deionized water to PACI is equal to the polymerization process that occurs in response to the addition of base. The gap in the pH value between PACI and deionized water narrows with an increase in the B value of the PACI. Under these conditions, the polymerization in response to the addition of base diminishes gradually, so the species of PACI with a high B value remain relatively stable during the dilution process (Fig. 3).

Aging time has been found to have an effect on the Al species distribution of PACI after dilution (Fig. 4). When the PACI samples were initially diluted, the Al_a content decreased slightly, and the Al_b content increased appreciably. Six hours later, the Al_a and Al_c contents decreased to a greater extent, while the Al_b content increased substantially and became the dominant species. In the following 30 h, there was little change in the Al_a content, while the Al_c content continually decreased and the Al_b content continually increased up to 66%. The above results suggest that a complex species transformation took place in the period of time after the PACI samples were diluted. The result of the species transformation was also dependent upon two opposing factors: Al_T and pH value. When the PACI was initially diluted, the effects of pH were more prominent, and the polymerization degree increased appreciably. With aging, the effect of Al_T became dominant. There was a trend of macromolecules being converted into smaller molecules. During this process, the average polymerization degree of PACI decreased. In addition, the Al_a and Al_c contents decreased and the Al_b content increased (Fig. 4), which meant that Reaction (2) could occur in the system. The considerable decrease in the Al_c content meant that the decomposition of Al_c could also occur. In the dilute solution, the high-molecular-weight Al species were gradually transformed into low-molecular-weight species via the solvation effect of water. Al_c was spontaneously transformed into Al_b or Al_a in dilute solution, and the opposing process was achieved via the addition of

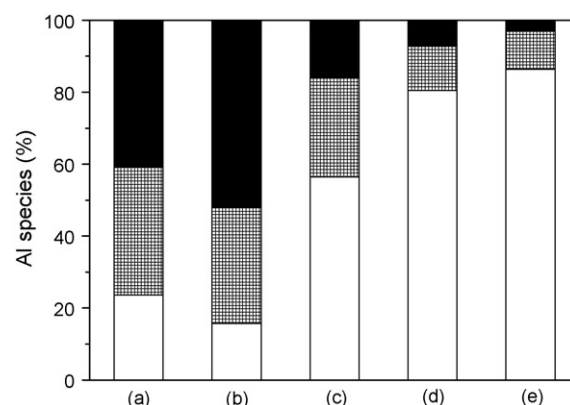


Fig. 5. Effect of dilution pH on the transformation of the Al species in PACI ($B = 2.0$, $Al_T = 2 \text{ mol L}^{-1}$). Al_a (□), Al_b (▨), Al_c (■). Undiluted sample (a), samples diluted 25,000 times at pH of 5.5 (b), 4.7 (c), 4.0 (d), and 3.5 (e), respectively, and measured 2 h after dilution.

base. The transformation balance of Al_c was dependent upon the concentration of the solution and the content of base, that is, the dilution degree and the B value (the higher the dilution degree, the lower the B value, and the greater the transformation degree of Al_c). In the present study, the B value was comparatively high, and the dilution degree was low, so Al_c was transformed only into Al_b and not Al_a . The longer the aging time after dilution, the higher the Al_b content became.

The PACI samples with $B = 2.0$ and $Al_T = 2 \text{ mol L}^{-1}$ were diluted 25,000 times at different pH values (5.5, 4.7, 4.0, and 3.5), and the Al species were measured 2 h after dilution (the final pH values were 4.8, 4.1, 3.8, and 3.5, respectively). The results of this process are shown in Fig. 5. The system pH had a considerable effect on the Al species in the dilution process. The average polymerization degree of the Al species increased after dilution at a pH of 5.5. These results were similar to those presented in Fig. 3, and the dilution under these conditions could be regarded as a base-adding process. When the pH value of the dilution medium was reduced, the high-molecular-weight Al species were transformed into low-molecular-weight species, and the average polymerization degree of the Al species decreased. As a result, the Al_c content decreased and the Al_a content increased. The Al_a content reached 86% at a pH of 3.5. In the low pH system, the dissociated H^+ decreased the polymerization degree of the high-molecular-weight Al species by breaking the hydroxyl and oxygen bridge in the Al species [34]. Therefore, dilution at low pH was no longer a base-adding process, but it rather became an acidolysis process. Different Al species distributions appeared when PACI was diluted in different pH systems, producing PACI with different flocculation performances at different pH values in water.

After 0.1 mL of PACI with $B = 2.0$ and $Al_T = 2 \text{ mol L}^{-1}$ was diluted in 1 L of tap water (the final system pH was 7.5), the species particle diameter was measured over time. As shown in Fig. 6, the average particle diameter of the Al species proceeded from high to low then to high again over time. This resulted from the PACI in the dilution first diffusing and then hydrolyzing. Specifically, the high-concentration sample was mainly composed of Al species polymers with larger particle diameters. After diffusing through the system, the Al polymer gradually changed into Al species with a smaller particle diameter. The Al species diffused completely 400 s later, and at this time the average particle diameter was the smallest. Then, because of the high pH of the tap water, the diffused Al species in the water hydrolyzed and polymerized. Species with high polymerization degrees were gradually formed, as demonstrated by the gradual increase in the average particle diameter. Our research indicates that the Al_a and Al_b species transform into Al_c species dur-

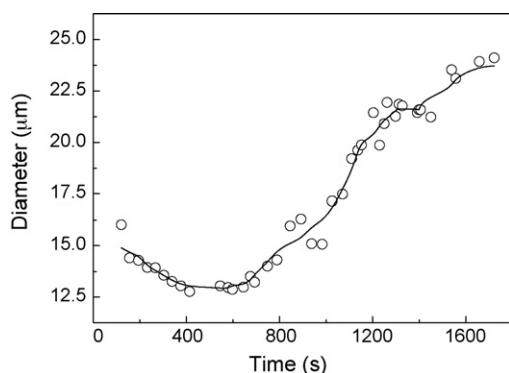


Fig. 6. Time course of the diameter of the Al species in PACl ($B = 2.0$, $Al_T = 2 \text{ mol L}^{-1}$) in the dilution process (diluted 10,000 times in tap water, the final pH 7.5).

ing the dilution of PACl (Fig. 3). The results in Fig. 6 demonstrate this point more directly.

4. Conclusions

To expand upon the current theories about PACls, high-concentration PACls with maximum Al_T and B value of 2.1 mol L^{-1} and 2.1, respectively, were prepared using the instantaneous base-feeding method, with solid CaO employed as the basification reagent. Whether in a high or low Al concentration system, there was an optimal B value at which the Al_b content of the products was the highest. When the B value was lower than optimal, the overall reaction mainly embodied the formation of Al_b ; when the B value was higher, it embodied the formation of Al_c . The optimal B value and the highest Al_b content both decreased with an increase in Al_T . The range of the B values suitable for Al_b formation was broad at low Al_T , and narrow at high Al_T . The range of B values suitable for Al_c formation was broad at high Al_T . With Al_T decreasing in the dilution process, PACl with a low B value exhibited a tendency towards Al_b formation, and PACl with a high B value exhibited a tendency towards Al_c formation. The dilution of PACl in a high pH system could be considered a polymerization process in the case of base-addition, and a process of acidolysis in the case of a low pH system.

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