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Coagulation optimization for low temperature and low turbidity source water using combined coagulants: a case study

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ABSTRACT

Coagulation optimization using coagulants of ferric chloride ($FeCl_3$), polymer–polyaluminum ferric chloride (PAFC) and their combinations ($FeCl_3$/PAFC) was evaluated by performing jar tests for treating source water with low temperature (<10°C) and low turbidity (1.5–8 NTU) i,n a typical North-China water treatment plant (WTP), Tanggu WTP. The results indicated that compared to single coagulant, the combined coagulants showed superior coagulation performance in terms of turbidity, UV_{254}, COD_{Mn}, iron, and aluminum removal. The optimal dosage was determined as 20 mg/L using the combined PAFC/$FeCl_3$ (3:1 by mass) and dosing PAFC with a 5–20 s interval followed by $FeCl_3$. Scanning electron micrographs further showed that the coagulations combination can form a more compact structure that increases the settleability of the flocculation flocs.

Keywords: Combined coagulant; Coagulation optimization; Low turbidity and low temperature source water

1. Introduction

The presence of natural organic matters (NOMs) in source water can lead to the formation of disinfection by-products (DBPs), such as trihalomethanes and haloaceic acids, during the disinfection water treatment process [1,31]. Thus, the US environmental protection agency has mandated the water utilities remove NOMs as a means to reduce DBP precursors [2].

Conventional coagulation is the most commonly used treatment process for turbidity and particle removal, which uses aluminum and ferric salts to remove the particles and NOMs [3]. The hydrolysis of single nucleus metallic salts in coagulation was very rapid, and the precipitates of metal hydroxide were produced afterward, resulting in difficulty to control the coagulation process [4,5]. The optimal pH levels for aluminium- and ferric-based coagulation were reported to be 5.0–6.5 and 4.5–6.0, respectively, for obtaining high removal of turbidity, dissolved organic carbon, and UV_{254} [6–9]. When the alkalinity in the raw water is high, coagulation using metallic salts can lower the pH level in water to achieve better metal hydrolyzation and the formation of hydroxide precipitate afterward. However, it leads to high demand of the dosage of the metal salts. On the other hand, the

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hydrolysis of polymeric salts can be controlled to a specific degree during production. Thus, the use of polymeric salts can reduce the complicated reactions triggered by iron–salt hydrolysis, which provide a simpler and more precise control on the coagulation reactions [10,11]. These coagulants may have some advantages over the metallic salts on a wider working pH range, lower reducing amounts of coagulant, lower sensitivity to water temperature, and lower residual iron concentration [6]. However, the disadvantage is that the preformed species are stable and cannot be further hydrolyzed during coagulation, and may not be efficient in removing a highly hydrophobic NOM [12,13].

The coagulation effectiveness to remove NOMs and turbidity depends on various factors, such as coagulant types and dosage, pH and particle properties, and the mechanisms through which NOMs are removed include a combination of charge neutralization, entrapment, adsorption, and complexation with coagulant metal ions into insoluble particle aggregates [6]. Among these mechanisms, the charge-driven mechanisms are believed to be the most evident when the pH is optimized [6,14–16]. However, it was noticed that metal salts alone are not effective in neutralizing the surface charges of particles in low turbidity and low temperature water [17]. Low turbidity raw water contains less negative particles and colloids, and requires less coagulant dosage to reach charge neutralization. As a result, the flocs that are formed are small and loose, and difficult to precipitate efficiently [18]. This will increase the operational cost and increase the corrosive tendency. On the other hand, the low temperature would also affect the coagulation kinetics that was reported in previous studies [19,20]. Aquantitative comparison between the coagulation kinetics at a high and that at a low temperature indicated that the low temperature exerted a significantly negative effect on the floc aggregation rate from 22 to 2℃, and the retarded perikinetic collision was found to be the cause for the low coagulation kinetics [21].

To increase the coagulation effectiveness and performance for treating water with low turbidity and low temperature, some attempts were made to use various new composite inorganic coagulants in water treatment, including polyaluminium ferric chloride (PAFC), novel composite polyaluminium ferric chloride (HPAC) and polyaluminium silicate chloride (PASC), poly-diallyldimethylammonium chloride (p-DADMAC) [30] and polyaluminium ferric silicate chloride (PAFSC). PASC combines aluminum chloride-based polymeric flocculants with polysilicate. It can enhance the bridging ability of a coagulant with the relatively large flocs formed and is more resistant to pH variations. However, the flocs are difficult to settle thus creating high turbidity values with too low dosage. PAFSC has similar properties to those of PASC with both aluminum- and ferric chloride-based polymers added. HPAC is composed of organic polymer (PDADMAC) and aluminum chloride (PACl) with such additives as active silicates. It is effective in high alkalinity or pH. p-DADMAC is a cationic polyelectrolyte and neutralization is the dominant removal mechanism. It is effective in the removal of hydrophobic NOM and produces less amount of sludge. The disadvantages of PASC and p-DADMAC are that the costs of using both these coagulants are high and may have toxic effects. Compared to novel composite coagulants, PAFC is made of prehydrolyzed aluminum and ferric chloride and thus enhances the amount of high-charged hydrolysis species. It can work at a lower temperature, a wider pH range, and helps in better NOM removal with less sludge produced than alum. Thus, PAFC has good working properties on our source water with low temperature and NOM removal. Moreover, the dominant mechanisms during the polymeric coagulants have also been suggested to be adsorption, entrapments, and complexation rather than charge neutralization [6,22].

On the other hand, as a public utility, water supply security is our priority concern. Replacement with a completely new coagulant may pose a risk and challenge, while using the combined coagulants of $FeCl_3$ and PAFC would be a good option that can be tried in Tanggu WTP. It is believed that a combination of the metal salts and polymeric coagulants could improve the coagulation performance. In this study, a series of well-designed tests were conducted to evaluate the coagulation performance of combined $FeCl_3$ and PAFC on low turbidity and low temperature source water, and to optimize the conditions of the combination coagulation.

2. Materials and methods

2.1. Source water

The source water investigated in this study was taken from the reservoir of Tanggu WTPs that was introduced from the Luan River, a local river running through northern China. The turbidity of source water before entering the plant was around 1.5–8.0 NTU in spring and winter. $FeCl_3$ was used as the coagulant in Tanggu WTPs, and the dosage of $FeCl_3$ was increased to as high as 60 mg/L to meet the national water quality standard and the internal guideline of 0.5 NTU for settled water.

However after some time, the residual turbidity in the treated water was still in the range of 0.7–1.4 NTU and the pH was lower than 7.0. A coagulant aid of activated silica was even used to enhance the flocculation. However, many difficulties were encountered in practical operation, such as the clogging of the dosing pump and the subsequent filter, difficulty in controlling the dosage, and requiring the onsite activation of activated silica. Therefore, an effective coagulation process is urgently required for water treatment plants like Tanggu WTPs, which are operated under a conventional water treatment process and using old treatment units/equipment.

During the jar tests, the source water quality used is shown in Table 1, with the turbidity content in the range of 4.03–7.32 NTU and the temperature around 0–9.8℃. It was also observed that the pH and alkalinity were high in the source water, while the UV_{254}, the indicator of NOMs was in the medium range.

2.2. Reagents

All reagents used were of analytical grade, except as mentioned otherwise. Liquid $FeCl_3$ with an Fe_2O_3 content of 38.27% (Qingyuan Water Purifying Reagent Company, Beijing) and solid PAFC with an Al_2O_3 content of 30%, Fe_2O_3 content of 2–4%, and sulfate content of 3.5–10% and 60–95% in the base saturation degree (Gongyi Yuqing Water Purification Material Co., Ltd, Henan Province), were used. PAFC is an aggregate of various dominant steady state or sub-steady-state aluminum ions/polymers, ferric ions/polymers, and aluminum-ferric co-polymers. Its chemical forms are polyhydroxy polymer and inorganic macromolecule polymer, which have inherent advantages of lower alkalinity consumption and less sludge production in particulate formation and organic removal compared to the conventional coagulants. During the production of PAFC, multivalent anions such as sulfate are added to improve the coagulation performance. Stock solutions of $FeCl_3$ and PAFC (each 10,000 mg/L) were prepared for the jar tests.

2.3. Jar tests

Coagulation jar tests were conducted in 1 L plexiglass beakers using a programmable jar testing apparatus, DC-506 Laboratory Stirrer (Huashui New Technological Development Co., Shanghai). The procedure of the batch coagulation tests was based on the practical operation and design for the Tanggu WTPs. The source water was filled in six beakers with 1 L each. Coagulant(s) were added followed by a 3-min rapid stirring at 200 rpm ($G = 102.5\ s^{-1}$) to allow complete coagulation/reaction and then followed by a 20-min slow stirring at 40 rpm ($G = 11.8\ s^{-1}$) to allow good flocculation and a 10-min settling time for floc–liquid separation. Unless specified, in combined coagulants tests the second coagulant was dosed 1 min after the first coagulant, i.e. 1 min after the rapid mixing. Supernatant samples were withdrawn at 2 cm below the water surface. Coagulation dosage was measured by a calibrated pipette.

2.4. Analytical methods

The turbidity was measured using a turbidimeter (RATIO/XR, Hach, USA). The collected water samples were filtered through a 0.45 μm pore size filtration membrane to measure the UV_{254} absorbance. The UV_{254} absorbance was measured at 254 nm with a UV-754 UV/VIS spectrophotometer (Jinghua Precision & Scientific Instrument Co. Ltd., Shanghai, China). The COD_{Mn} was determined using an acidic potassium permanganate method [23]. The residual aluminum was measured using the Chrome azurol Sphotometric method [24]. The residual iron was determined using the orthophenanthroline photometric method [24]. Morphologies of the air-dried flocs were examined and measured spectroscopically using a SEM (S-3500 N, Hitach.) under a 20 kV voltage.

Table 1
Source water quality characteristics

Parameter	Unit	Min	Max	Mean ± SD	Number of measurements
Temperature	℃	0	9.8	5.2 ± 1.1	30
pH	–	8.02	8.59	8.43 ± 0.11	30
Turbidity	NTU	4.03	7.32	5.12 ± 0.97	30
Alkalinity	mg $CaCO_3$/L	124	174	134 ± 4.8	30
NH_4^+	mg/L	0.10	0.21	0.15 ± 0.07	30
COD	mg/L	2.9	3.6	3.19 ± 0.42	30
UV_{254}	cm^{-1}	0.093	0.057	0.085 ± 0.008	30

3. Results and discussion

3.1. Effects of dosage on coagulation performance

The coagulation performance was examined with a range of dosages (5–50 mg/L) for single $FeCl_3$, single PAFC and combined PAFC/ $FeCl_3$ (1:1 by mass) by dosing PAFC followed by $FeCl_3$. It is shown in Fig. 1 that the coagulant dosage was significant on the turbidity removal efficiency. The residual turbidity in settled water decreased with increase of the coagulants' dosage from 5 to 30 mg/L. The difference using the three dosing methods was that as the dosage continued to increase after 30 mg/L, the residual turbidity continued to decrease when $FeCl_3$ was used, while it remained stable and even increased when PAFC or combined coagulants PAFC/$FeCl_3$ were used. In addition, using single $FeCl_3$ the residual turbidity was beyond 0.5 NTU (the internal guideline in Tanggu WTP), even under the high dosage of 50 mg/L. Single PAFC and combined $FeCl_3$/PAFC showed a similar turbidity removal efficiency, and below 20–30 mg/L dosage, the residual turbidity reached below 0.5 NTU. These results indicated that the composite coagulant of PAFC and the combined PAFC/$FeCl_3$ have better coagulation performance on turbidity removal and the optimal dosage was determined as 20 mg/L.

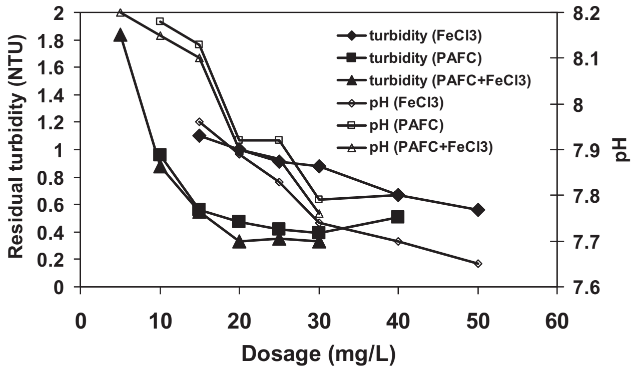

Fig. 1. The comparison of single $FeCl_3$, single PAFC, and combined PAFC/ $FeCl_3$ on turbidity removal efficiency.

It has been reported that alkalinity/pH affects the chemistry of coagulants, particularly the speciation distribution of the coagulants after dosing, and further speciation transformation [25]. The source water with high alkalinity (124–174 mg/L) would require high $FeCl_3$ dosage to adjust the pH to be favorable for coagulation. These results indicated that the combined $FeCl_3$/PAFC have better coagulation performance than single $FeCl_3$ or PAFC on turbidity removal and the optimal dosage was determined as 20 mg/L.

3.2. Effects of dosing order of the combined PAFC/$FeCl_3$ on coagulation performance

The mechanisms to explain the particle coagulation and NOMs include charge neutralization, entrapment, adsorption, and complexation [6]. Under different conditions, the different conditions or their combination may be dominant. The dominant turbidity removal mechanisms using PAFC have been suggested to be adsorption, entrapment, and sweep flocculation, while that using $FeCl_3$ has been concluded to be charge neutralization [14,15]. Thus, the dosing order representing the effect of turbidity removal mechanisms should be studied.

The coagulation performance using single $FeCl_3$, single PAFC, and different dosing order of $FeCl_3$ followed by PAFC (1:1 by mass), PAFC followed by $FeCl_3$ (1:1 by mass), and simultaneous dosing were further evaluated with the total dosage of 10 mg/L. Three tests were carried out for each dosing method. The results (Fig. 2) showed that dosing PAFC followed by $FeCl_3$ has the most effective turbidity

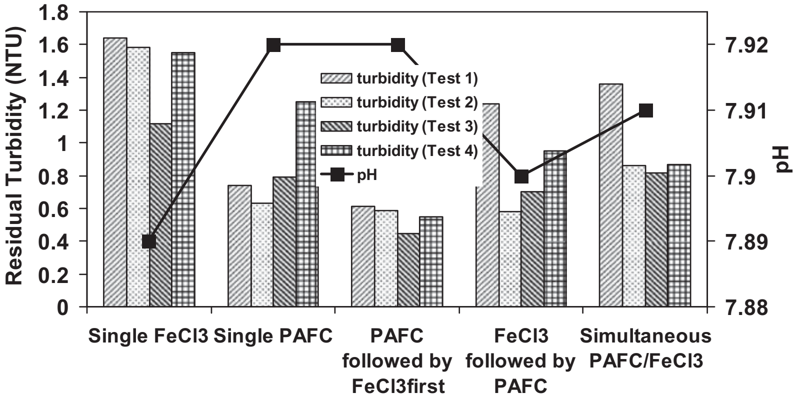

Fig. 2. Effect of dosing order of the combined $FeCl_3$/PAFC on turbidity removal efficiency.

removal efficiency with the settled turbidity of approximately 0.5 NTU on average, while using single $FeCl_3$ had the highest residual turbidity of 1.4 NTU in settled water. This is because at low dosage, the high alkalinity (i.e. high pH) in source water inhibits the degree of hydrolysis and hydrolysis products of ferric salts that affect its coagulation performance. When dosing $FeCl_3$ first into source water with the pH of 8.0, the charge neutralization effect would be very low, which strongly reduced the efficiency of $FeCl_3$ coagulation. However, coagulation using PAFC is less sensitive to pH. When dosing PAFC first, the other mechanism of entrapment, adsorption, and complexation may be dominant.

3.3. *Effects of dosing ratio of the combined $FeCl_3$/PAFC on coagulation performance*

Since the dosing order of the combined coagulant had a great effect on the turbidity removal, we hypothesized that there would be an optimal ratio of PAFC/$FeCl_3$ on the turbidity removal. PAFC can enhance the bridging ability of a coagulant, and the flocs formed are relatively large. Fig. 3 shows different dosage mass ratio of PAFC/$FeCl_3$ (6:1 to 9:1 of the two coagulants) on the residual turbidity removal. The total dosage was 10 mg/L and three different dosing orders were studied. It showed that dosing PAFC followed by $FeCl_3$ generally had low turbidity in settled water and when the ratio was 3:1, it reached the highest turbidity removal efficiency. As the ratio decreased from 3:1 to 1:9, the turbidity would increase dramatically, which was higher than those of the other two dosing methods, indicating the mechanisms other than charge neutralization on the low temperature and low turbidity source water.

3.4. *Effects of dosing interval of the combined PAFC/$FeCl_3$ on coagulation performance*

The time of rapid mixing affects the destabilization of colloids and subsequent aggregation of particles [26]. Using polymers as coagulant, Amirtharajah and Jones [27] found that rapid mixing is critical in colloid bridging. Besides, the combined coagulants showed superior efficiency. Thus, the dosing time interval of the two coagulants was measured in the study. Two tests by dosing with PAFC followed by $FeCl_3$ were conducted with the total coagulant dosage of 10 mg/L with a PAFC/$FeCl_3$ mass ratio of 3:1.

It showed that the residual turbidity in settled water decreased as the dosing interval increased from 0 to 5 s, and remained stable until 20 s, after which the turbidity started to increase until 60 s, indicating that the 5–20 s interval can bring about effective coagulation efficiency (Fig. 4). A similar result was obtained by Edzwald et al. [28,29], finding that, using aluminum as a coagulant, maximum destabilization capacity is reached within 0.1 s. The coagulation efficiency decreased continuously from 0.1 to 6 s, and then remained at the same level after 6 s. In our study, it was also noticed that the flocs formed using simultaneous dosing of PAFC and $FeCl_3$ were much smaller in size than the flocs formed using dosing of coagulants in sequence (data not shown).

3.5. *Coagulation optimization integrating combined coagulant dosage, order, ratio, and time interval*

Considering the effects of coagulant amount, order, ratio, and time interval on the coagulation performance, we integrated the optimal value for each parameter, i.e. dosing PAFC with 5 to 20 s followed by $FeCl_3$ (3:1 by mass) using different amount of dosage, for evaluating the treatment efficiency of high

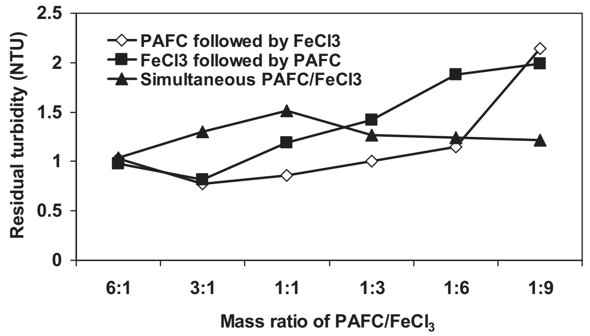

Fig. 3. Effects of dosage mass of the combined PAFC/$FeCl_3$ on the residual turbidity removal.

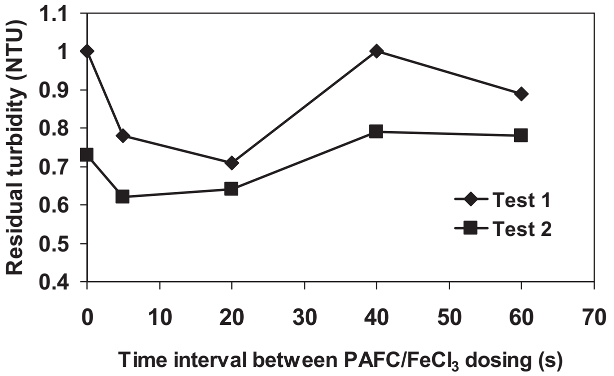

Fig. 4. Effects of dosing interval of the combined PAFC/$FeCl_3$ on turbidity removal.

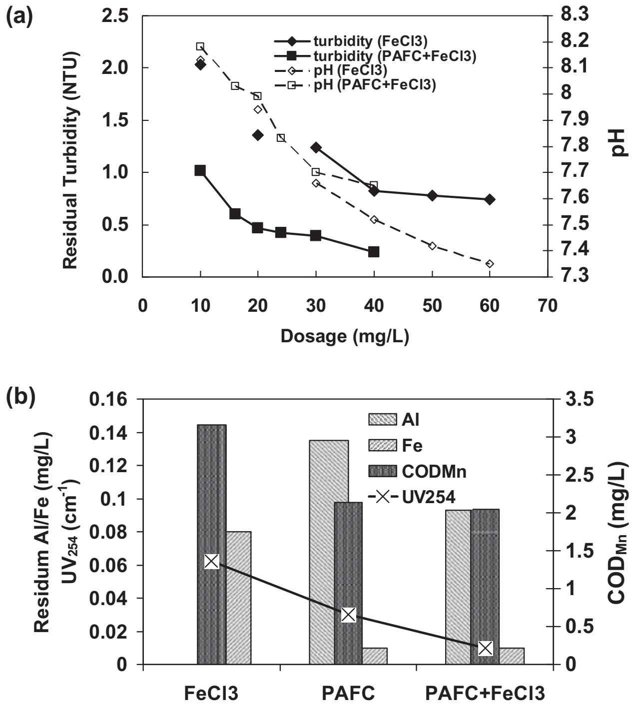

Fig. 5. Coagulation optimization by integrating combined coagulant dosage, order, ratio, and time interval using single $FeCl_3$ and combined coagulants $FeCl_3$/PAFC with the total dosage of 20 mg/L. (a) Change of residual turbidity for different amount of coagulant dosage, (b) and (c) Residual Fe and Al concentration and UV_{254}.

algae source water, and compared the results using single $FeCl_3$ dosing. The source water in this test contained a turbidity of 7.9 NTU and UV_{254} of 0.1339 cm^{-1}.

The results (Fig. 5a) showed that compared to single coagulant of $FeCl_3$, the combined coagulants (PAFC/$FeCl_3$) showed superior coagulation performance in terms of turbidity and UV_{254} removal. Using the total coagulant dose of 20 mg/L, the turbidity level can drop below 0.5 NTU.

It was also indicated that using the combined coagulant, the UV_{254} and COD_{Mn} removal efficiencies were 84 and 43%, respectively, which are much higher than using the single coagulant with the corresponding removal efficiencies of 72.4 and 11.5% (Fig. 5b). The combined coagulant can improve the removal efficiency of organic matter significantly. Besides, the use of PAFC may increase the concentration of total aluminum concentration and decrease the concentration of iron in treated water, compared to the use of single $FeCl_3$. It was suggested that aluminum in drinking water may pose a risk toward Alzheimer's disease. The residual aluminum and iron concentrations in this study were determined as 0.90 and 0.01 mg/L, respectively. A much lower iron concentration using the combined coagulant can help solve the problem of high residual iron concentration in treated water that used single $FeCl_3$ as a coagulant.

3.6. Floc structure examined by SEMs

The differences in the coagulation performance using different coagulants can also be reflected in the floc structures observed by SEMs (Fig. 6). Using single $FeCl_3$ as a coagulant, the flocs formed with porous structures, where particles and colloids (both organic and inorganic) would be adsorbed or swept afterward (Fig. 6a).The flocs formed using PAFC test tend to exist as chain-like or reticulate structures and some porous structures can be seen (Fig. 6b). It is believed that the chain-like and reticulate structures contributed to lowering the turbidity in the settled water. When using the combined PAFC/$FeCl_3$, a more compact floc structure was obtained (Fig. 6c). The mechanism of the enhanced coagulation using a combined coagulant is probably that the chain-like and reticulate structures can adsorb Fe^{3+} or contribute to a combination settling with the ferric hydrolysis species to form the soluble reticular aluminum–ferric structures. This can also be verified to some extent, by the fact that the color of the flocs/structures using the combined coagulant of a mixture was yellow with Fe^{3+} and white with PAFC. There are also some uncompleted structures in Fig. 6c, which showed the process of flocculation. The combined dosing approach enhanced the coagulation with compact structures. However, addition of $FeCl_3$ after PAFC can combine with the reticular structures, which can speed up the sedimentation of the whole flocs/structures [29].

3.7. Mechanisms of the combined coagulant using PAFC/$FeCl_3$

Similar to aluminum salt, ferric salts hydrolyze when added to water and the most effective range of pH is suggested to be 4.5–6. It has been noted to be better than alum to remove middle-size NOM fractions and not to be so sensitive to a temperature change. However, ferric-based coagulant associated optimized coagulation pH range produces treated water with less buffering capacity, and requires greater chemical addition for stabilization and corrosion control. On the other hand, PAFC may have some advantage as coagulants over monomeric forms of ferric salts. It is made by partially neutralized prehydrolyzed aluminum and ferric chloride, and only the most favorable coagulation hydrolysis and polymeric species can be produced. It

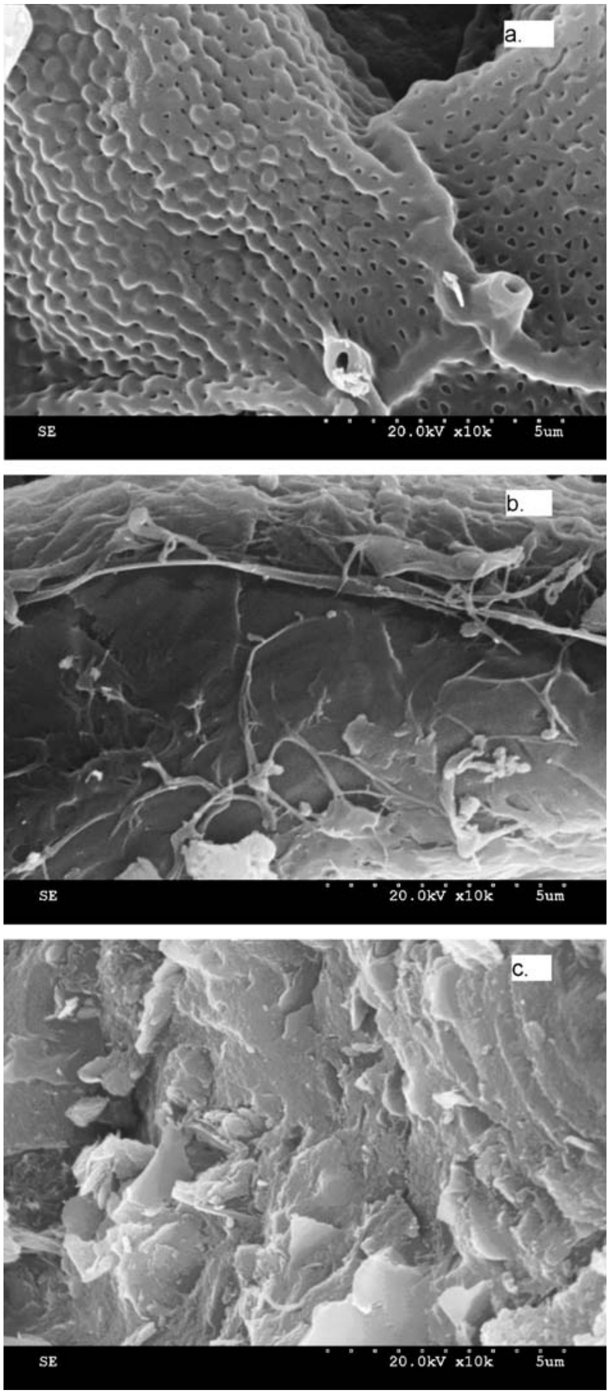

Fig. 6. SEMs for the flocs formed using different coagulants/coagulant aid, with the magnification of 10,000. (a) Single $FeCl_3$, (b) single PAFC, and (c) combined PAFC/$FeCl_3$.

enhances the amounts of high-charged and moderate-molar-mass hydrolysis species. Compared to ferric salts, PAFC has wider pH range and lower sensitivity to temperature, lower dose of coagulants, lower residual iron concentration, and is less corrosive. Rather than charge neutralization using $FeCl_3$, the possible dominant removal mechanisms during PAFC coagulation have been suggested to be adsorption, entrapment, and complexation.

The mechanism of effective coagulation using combined PAFC/$FeCl_3$ is believed to be due to the formation of the soluble reticular aluminum-ferric structures by integrating the effect of charge neutralization, adsorption, entrapment, and complexation, which can speed up the sedimentation of the whole flocs/structures. This hypothesis can be further verified by the SEMs. The mechanisms of coagulation by combined PAFC/$FeCl_3$ are more complex and will be systematically investigated in the future study.

4. Conclusion

Coagulation optimization was evaluated in this study using combination coagulants of PAFC and $FeCl_3$ for treating low turbidity and low temperature and low turbidity source water. The results showed that the efficiencies of combination coagulation are much better than using single coagulant, and the residual turbidity can reach less than 0.5 NTU. The treated water qualities such as the residual turbidity, UV_{254}, $COD_{Mn,}$ and iron in the combination coagulation are much better than those of the conventional coagulation, and the floc structures are more compact. The preliminary results made in this paper will be used as the basis for further investigations into the pilot and full-scale treatment processes of this type of source water.

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