Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2020.26252

204 (2020) 204–212
November

The influence of coagulation mechanism on coagulation effects under different Al-based coagulants conditions

Yonglei Wang[a,*], Ya'nan Li[a], Wenhao Wang[b], Liping Tian[c], Ruibao Jia[d], Wuchang Song[d], Ling Ju[a]

[a]*College of Environmental and Municipal Engineering, Shandong Jianzhu University, Jinan, China, Tel. +8618865928865; email: wyl1016@sdjzu.edu.cn (Y. Wang), Tel. +8617865161430; email: 654396630@qq.com (Y. Li), Tel. +8615610172413; email: 1558376349@qq.com (L. Ju)*
[b]*Jinan Municipal Engineering Design and Research Institute, Jinan, China, Tel. +8618366185082; email: 841396516@qq.com (W. Wang)*
[c]*Weifang Municipal Public Utilities Management Office, Weifang, China, Tel. +8613905366156; email: 13905366156@139.com (L. Tian)*
[d]*Shandong Province City Water Supply and Drainage Water Quality Monitoring Center, Jinan, China, Tel. +8613505317293; email: jiaruibao68@126.com (R. Jia)*

Received 28 January 2020; Accepted 13 June 2020

ABSTRACT

As the first step in a conventional water treatment process, coagulation often directly affects the conditions of the subsequent process and affects the overall operating cost and water quality of the effluent. In this paper, several Al-based coagulants were used to conduct coagulation tests on the water from the diversion reservoir (Queshan reservoir), and the removal effects of different pollutants were systematically analyzed. At the same time, the mechanism of different Al-coagulants in the coagulation process was compared. The results showed that the coagulant dominated by charge neutralization (such as $AlCl_3$) was effective in removing UV_{254}, total organic carbon, and humic acid in water. When the coagulant was dominated by adsorption bridging (such as composite coagulant), it was more beneficial to remove turbidity, chemical oxygen demand, and soluble microbial products in water. In addition, when adsorption bridging was the dominant mechanism, the coagulant was conducive to the growth of flocs. When the flocs were broken and regenerated, the charge neutralization was more favorable.

Keywords: Coagulation mechanism; Floc characteristics; Removal efficiency; Al-based coagulants

1. Introduction

As the core process of conventional water treatment, the treatment effect of coagulation often directly affects the operating conditions of the subsequent process and affects the overall operating cost and water quality of the effluent. In recent years, domestic and foreign researchers have conducted many fundamental studies on the coagulation effect of different coagulants. Ke et al. [1] prepared polymeric aluminum with different alkalinities by a microtitration method and determined its aluminum morphological distribution. Based on floc parameters, such as zeta potential and turbidity, the hydrolyzed morphology, and floc characteristics of polymeric aluminum were comprehensively evaluated. Wang et al. [2] selected three coagulants, $AlCl_3$, $FeCl_3$, and $Fe_2(SO_4)_3$, for compounding. The results showed that when the composite coagulants were added with a ratio of $m(AlCl_3):m(FeCl_3):m[Fe_2(SO_4)_3] = 5:6:7$, the removal effect of pollutants was the best. As the research field has advanced, many researchers have found that

---

[*] Corresponding author.

different coagulation mechanisms also play a very important role in the coagulation process [3]. Currently, it is generally recognized by researchers that the four mechanisms of charge neutralization, sweep flocculation, adsorption bridging, and electric double layer compression plays the main roles [4–7]. Shen et al. [8] used polyaluminum chloride–poly dimethyl diallyl ammonium chloride (PAC-PDMDAAC) dual coagulation system to form bulky and dense flocs, which effectively removed the natural organic matter (NOM) in the water, and found that the former mainly played the role of charge neutralization in the coagulation process, while the latter mainly played the role of sweep flocculation, adsorption bridging. However, in the practical application of water treatment, the above mechanisms are related to each other, and factors such as coagulant characteristics and coagulation conditions determine which mechanism plays a leading role. The removal effect corresponding to different coagulation mechanisms is quite different. Due to the complex composition of water sources and the large influence of water temperature and alkalinity, the leading mechanism of coagulation has not been clearly explained. There are relatively few studies on the effects of different coagulation mechanisms on the coagulation effect.

The water samples used in this experiment were taken from the diversion reservoir (Queshan reservoir) (the main water source in Jinan). The water quality is susceptible to seasonal changes. Because of the high algae content in the water in summer and the low temperature and low turbidity in winter, the difficulty of water treatment is increased. At present, most of the water plants in this area adopt coagulation–sedimentation–filtration as the main water treatment process, and the main coagulant used is Al-based coagulants. In order to improve the quality of the effluent water without changing the existing treatment process, it is necessary to strengthen the coagulation effect. At present, there are few studies on the coagulation mechanism of different Al-based coagulants. Therefore, in this paper, several common Al-based coagulants were used to conduct coagulation tests on the water of the diversion reservoir (Queshan reservoir). The coagulation effects were evaluated by comparing the basic water quality index (turbidity, COD, UV_{254}, etc.), floc characteristics and floc structure changes. At the same time, the corresponding removal mechanism was analyzed by zeta potential changes and other aspects. It was expected to be helpful for the use and improvement of coagulant, and provide reference for the study of coagulation mechanism in the future.

2. Materials and methods

2.1. Raw water

This experiment was based on the water of a diversion reservoir (Queshan reservoir) in Jinan. The water quality indexes during the test are shown in Table 1.

2.2. Jar testing

Jar testing was performed using a programmable six-paddle stirrer (ZR4-6, CN). Six identical coagulation beakers were selected, and 1 L of the sample water was added into them. The hydraulic conditions and added time of the coagulant settings are shown in Table 2. After the completion of coagulation, it was allowed to stand for 30 min, and at 2 cm below the liquid surface, water samples were measured for changes in turbidity, COD, UV_{254}, and other indicators (the detection methods were carried out in accordance with the method described in the "methods for the analysis of water and wastewater" (4th ed. Supplement)) [9]. The coagulants used in this test were aluminum chloride ($AlCl_3$), polyaluminum chloride (PAC), polyaluminum ferric chloride (PAFC), and composite coagulants (adding chitosan (CTS) to PAC). The concentration of coagulant added in the test was measured by Al^{3+}. The pH was measured using a METTLER TOLEDO pH tester (FiveEasy, CN). During the test, the pH of the solution was maintained at about 8.25. Turbidity was measured using a HACH 2100N turbidimeter (HACH 2100N, USA). TOC was measured using a total organic carbon analyzer (SHIMADZU CPH, JP). UV_{254} was measured using a Tu-1810 UV-vis spectrophotometer.

2.3. Floc characterization

2.3.1. Floc particle size measurement

The jar testing process was used to improve the device (Fig. 1). The formation of flocs was detected by an on-line monitoring device (Mastersizer 3000E, UK). At the same time, the floc size and the variation along the path were detected. The values were recorded every 3–5 s.

2.3.2. Zeta potential measurement

The zeta potential of the effluent was measured using a Marvin zeta potential analyzer (Nano Z, UK).

Scanning electron microscopy (SEM) detection: The generated flocs were pretreated by a freeze-drying method and then analyzed by SEM (Zeiss Sigma 300, GE).

2.3.3. 3D fluorescence detection

Excitation-emission matrix (EEM) spectra were detected using a fluorometer (Hitachi F-2700, JP). The scanning ranges of EX and EM were 220–450 and 220–550 nm, respectively. The scanning speed was 12,000 nm/min, and the interval was 5 nm. EEM data were redrawn using Origin graphics software and characterized in the form of contours and histograms. Under the same conditions, pure water was used as a blank to eliminate Raman scattering and background noise.

Table 1
Main characteristics of raw water

Sample	Reservoir water
T (°C)	20–25
Turbidity (NTU)	2.0–5.0
pH	8.10–8.40
COD_{Mn} (mg/L)	4.0–5.5
UV_{254} (cm^{-1})	0.054–0.105
Alkalinity (mg/L)	90–110
TOC (mg/L)	2.9–3.25

Table 2
Jar test procedure

Speed (rpm)	*T* (s)	Dosing	G/S-1
200	30	0	100.1
200	30	1	100.1
50	900	0	15.9
0	900	0	0

"1" in the dosing column means dosing occurred, and "0" means no dosing occurred.

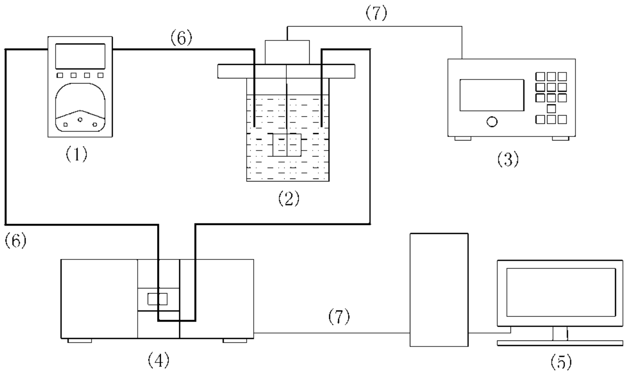

Fig. 1. Schematic diagram of the online monitoring device: (1) peristaltic pump, (2) coagulation beaker, (3) program-controlled device, (4) online monitoring equipment, (5) computer, (6) water pipe, and (7) equipment connection line.

3. Results and discussion

3.1. Turbidity removal comparison and mechanism analysis

Fig. 2 shows the removal of water turbidity and the corresponding effluent zeta potential of several different types of aluminum salt coagulants at different dosages. It can be seen from Fig. 2, that the turbidity removal effect of water by using different coagulants was quite different, and the turbidity removal efficiency increased gradually with increasing dosage. The turbidity removal effect of some coagulants (such as $AlCl_3$) had a basically linear relationship with the changes in the trend of the zeta potential, and the turbidity removal effect was the best when the potential was near the isoelectric point (zero potential point), with the removal efficiency reaching 80.54%. According to the change in potential during the coagulation process, the removal of turbidity by $AlCl_3$ was dominated by charge neutralization, and the removal of turbidity was better under this mechanism. As the dosage continued to increase, it produced a large amount of amorphous aluminum salt hydroxide. Under this condition, the dominant mechanism gradually turned to sweep flocculation [10]. For the composite coagulants, the turbidity removal effect was very significant at a low dosage (1 mg/L), which increased by approximately 17% compared with that of $AlCl_3$. Compared with PAC and PAFC, it increased by more than 40%. However, with a continuous increase in the coagulant dosage, the removal effect of turbidity was not obvious, it basically remained in a stable state, and the removal effect was not as good as that of the other coagulants alone under the same conditions (decreased by approximately 10% in the same period). Due to the low turbidity of the raw water used in the test, the good coagulation effect of the composite coagulant at low dosage (2 mg/L) should be attributed to the good adsorption bridging effect of the long-chain structure of CTS macromolecules [11,12]. In addition, according to the change of zeta potential in Fig. 2, with the increase of the dosage of the composite coagulant, the potential increased, but the turbidity removal efficiency did not increase. These results indicated that the composite coagulant was not dominated by charge neutralization during the coagulation process, and the increase in potential was caused by the large amount of positive charge carried by the CTS contained in the composite coagulant.

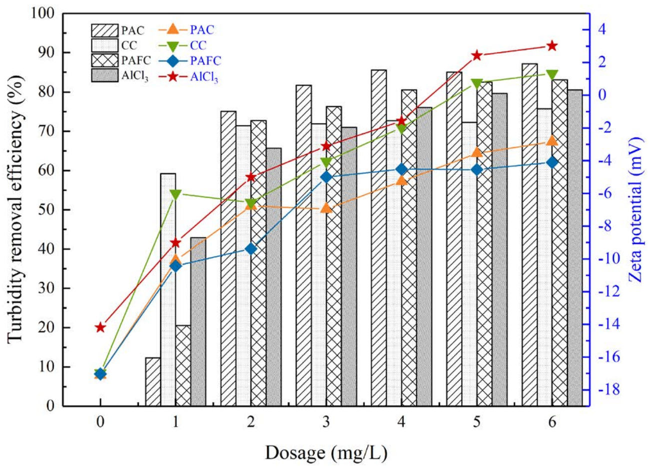

Fig. 2. Turbidity removal effect and zeta potential change under different coagulant conditions (CC: composite coagulant).

In addition, it can be seen from Fig. 2, that when PAC was used, the removal effect of turbidity was significantly improved with an increasing dosage, and the best removal efficiency reached 87.16%. According to the change of zeta potential, charge neutralization was the dominant mechanism of PAC in turbidity removal [8]. At the same time, recent studies have shown that the coagulation performance of PAC is related to the morphological distribution of aluminum after being added to water. Al_b plays the role of charge neutralization, and Al_c has a strong adsorption bridging ability [1,13]. Therefore, in addition to charge neutralization, adsorption bridging also played a positive role. This was similar to the conclusion of Zhao et al. [14].

It can be seen from the above results that under a low dosage, the effect of the coagulant dominated by charge neutralization on turbidity removal was less than that of adsorption bridging. However, as the amount of coagulant used increased, the gap narrowed.

3.2. Organic removal comparison and mechanism analysis

Experiments were carried out to compare the removal effects of COD, UV_{254}, and TOC under different coagulation conditions. At the same time, a mechanism analysis was carried out in combination with the zeta potential changes, and the effects of different dominant mechanisms were studied based on the above indicators.

Fig. 3 shows the removal of various organic indicators by several different coagulants under different dosages. Additionally, the figure compares the removal effects of various organic substances when the dosages of several coagulants were the same, but the mechanism of action was different. It can be clearly seen from Fig. 3 that for the same coagulant, the removal effect of most coagulants on various organic indicators in the study showed a gradually increasing trend with an increased dosage of coagulant, and some coagulants showed a trend of first increasing and then slightly decreasing. The composite coagulant produced a good coagulation effect at a low dosage (1–2 mg/L), the removal effect of COD can reach 44.8%. With an increasing dosage, the removal effect of composite coagulants on various indicators did not significantly increase, while the removal effect of some indicators, such as TOC, became worse. This may be because the added chitosan was in itself a kind of polymer organic matter. When the dosage was increased, it was not completely combined with the impurities in the water and resulted in a surplus, which led to the increased content of indicators, such as TOC, in the water. The removal effect of $AlCl_3$ on various organic substances was relatively stable. It had a better removal effect on UV_{254} than other coagulants at the same dosage, the best removal efficiency reached 55.56%. For the removal of TOC, the removal effect of PAC and $AlCl_3$ at the same dosage was ideal, the highest removal efficiency was 24.51% and 26.05%.

Through the study of the removal of water turbidity and the change in zeta potential, it can be known that the composite coagulant was mainly dominated by adsorption bridging when removing organic matter, and $AlCl_3$ was mainly dominated by charge neutralization. For both PAC and PAFC, the above two mechanisms of action played a positive role. Combined with the results of the study in Fig. 3, it can be seen that the removal effect of UV_{254} and TOC was better than that of adsorption bridging when the coagulant was dominated by charge neutralization. When the coagulant was dominated by adsorption bridging, its removal effect on COD seemed to be better.

In addition, due to recent attention for the removal of organic pollutants, especially dissolved organic matter (such as humic acid and fulvic acid, etc.) [15–19], this study also examined the removal effect of some dissolved organic matter in water. Fig. 4 shows the corresponding 3D fluorescence spectrum of raw water before and after the addition of the coagulant. The map was divided into five regions, representing five different types of dissolved organic matter. The five regions and their representative organic matter were tyrosine (aromatic amino acid): EX 200–250 nm, EM 280–320 nm; tryptophan (aromatic amino acid): EX 200–250 nm, EM 320–380 nm; fulvic acid: EX 200–250 nm, EM 380–540 nm; soluble microbial products (SMP): EX 250–340 nm, EM 280–380 nm; and humic acid: EX 250–400 nm, EM 380–540 nm. Because the content of microbial products in the sample water was too high and the display ratio was set to be large, the changes in other organic substances were not intuitive and difficult to compare. Therefore, the four dissolved organic matter profiles, except SMP, were listed separately. They were drawn at a smaller display scale and placed around the main image so that they could be observed more clearly and intuitively.

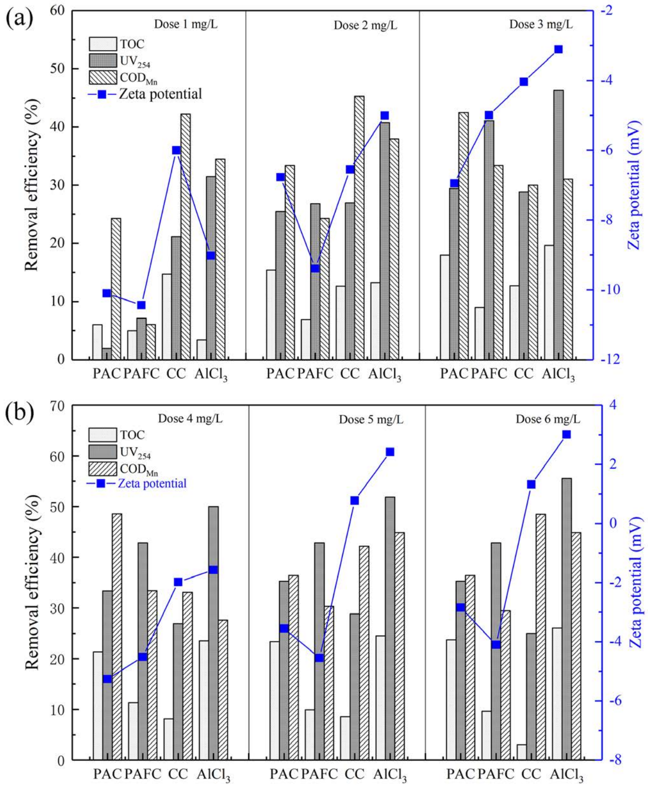

Fig. 3. (a and b) Removal of organic matter under different dosages of several coagulants (1–6 mg/L) (CC: composite coagulant).

From the distribution of the material in Fig. 4a, it was found that SMP was the main component in the target water, and its fluorescent intensity was much higher than that of other substances, followed by tyrosine, humic acid, and fulvic acid. It can be seen from the comparison between Figs. 4b and c and Fig. 4a that as the dosage was increased, the fluorescent substances in each part were reduced to a certain extent. The removal of SMP was more obvious. Figs. 4d and e show that the composite coagulant had a better removal effect on SMP and tyrosine than $AlCl_3$, and this phenomenon became increasingly obvious with an increased dosage. This may be due to the increase in the amount of Al_b in the composite coagulant, which increased the charge neutralization and enhanced the removal effect of SMP [20]. Previous studies have shown that SMP is the main component of COD [21]. Therefore, after the composite coagulant enhanced the removal of SMP, it can have a positive effect on the removal of COD, which also conformed to the relevant discussion in section 3.2 (Organic removal comparison and mechanism analysis).

By sorting out the results, it was found that the removal effects of the $AlCl_3$ and composite coagulant on humic acid were 30.06% and 26.85%, respectively. In contrast, the removal effect of humic acid was better when using $AlCl_3$ and indicated that charge neutralization was more favorable

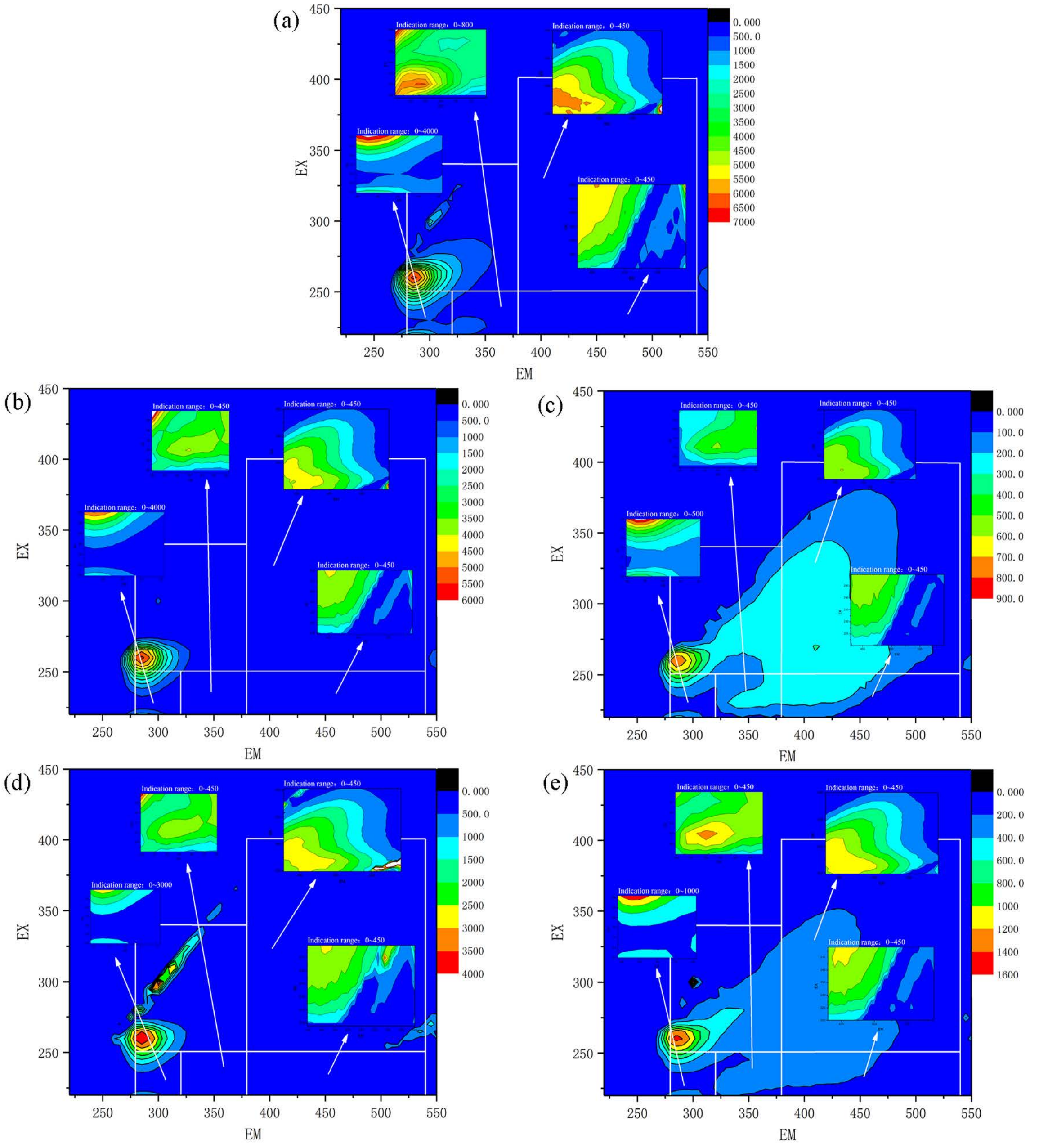

Fig. 4. 3D fluorescence spectrum of the effluent under different coagulant conditions (a) raw water, (b) adding 1 mg/L $AlCl_3$ to the raw water, (c) adding 2 mg/L $AlCl_3$ to the raw water, (d) adding 1 mg/L composite coagulant to the raw water, and (e) adding 2 mg/L composite coagulant to the raw water.

for the removal of humic acid, which was similar to the research results of Cheng [22] and Kam and Gregory [23]. The poor removal effect of compound coagulant may be attributed to the following two reasons: the dosage of the coagulant was less, and the charge neutralization mechanism worked in both ways, but the effect was more obvious when $AlCl_3$ was used. In addition, the removal efficiency of fulvic acid by composite coagulant and PAC was less than that of humic acid, which were 26.79% and 26.89%, respectively. This also confirmed the research results of Wang and Ma [24] and Zhou et al. [25]. It may be that humic acid provided more bonding points with Al^{3+}, which changed its particle size and zeta potential to a greater extent and promoted the formation of flocs.

3.3. Floc characteristics comparison and mechanism analysis

The particle size of the flocs and their growth characteristics directly determined the subsequent solid–liquid separation effect [26]. This study used a Mastersizer 3000E and other equipment to conduct related research on the particle size and structure of flocs to explore the formation of flocs, the role of different dominant mechanisms on characteristics, and methods of influencing the dominant mechanism.

Floc size is an important index to evaluate coagulation performance. The settling speed of larger flocs is faster than that of smaller flocs, which leads to higher removal efficiency [27]. Fig. 5 shows the comparison of the floc size produced by several coagulants at different dosages and the corresponding zeta potential changes. It can be clearly seen from the figure that with increased dosage, the floc particle size showed a gradual increase or a slight decrease after the increase, which was similar to the research results of Zhao et al [28]. It can be seen from Fig. 5 that the flocs formed by $AlCl_3$ in the whole dosage range were smaller, while the flocs formed by composite coagulants were significantly larger than other coagulants with the increase of dosage. Combined with the changes of zeta potential data in the figure and the results of the previous sections, it was known that the particle size of the floc formed by the composite coagulant dominated by adsorption bridging was larger than that of $AlCl_3$ dominated by charge neutralization, and a good bridging effect contributed to the growth of the particle size. When PAC and PAFC were involved in the formation of flocs, both mechanisms played a positive role, but the particle size of the flocs was still significantly smaller than that formed by the composite coagulant. It may be because the CTS particles in the composite coagulant had a larger particle size than other coagulants (the average particle size of the CTS particles used in this experiment was 10 μm after detection by a laser particle size analyzer, while that of other coagulants were less than 1 μm), and with the increase of the dosage, more chitosan participated in the formation of flocs. Therefore, it had a certain influence on the floc size.

Fig. 6 shows the change in floc size and the regrowth of the floc under a partial dosage of different coagulants. Because the mechanism of PAC and PAFC in the coagulation process was similar, and it is known from Fig. 5 that the floc particle size generated by PAC and PAFC under the same dosage was also similar, so in order to express more clearly, PAC was selected as a representative and compared with other two coagulants. In general, after adding coagulant, the growth of floc particle size showed two stages: rapid growth and stability. In the stable stage, the growth and breakage of the floc reached an appropriate balance [4]. The changes in the floc in these two stages can be clearly seen in the first half of Fig. 6. In addition, different coagulants, dosages, and leading coagulation mechanisms directly affected the size of the floc when it reached stability and the time it took to reach that stability. It can be seen from Fig. 6 that the floc growth formed by the charge neutralization was relatively slow, and the floc particle size was significantly smaller than that of the floc formed under the action of adsorption bridging. This also showed that the flocculation ability of the flocs produced by $AlCl_3$ was worse than that of the composite coagulant. Comparing the floc growth of the PAC and composite coagulant, it was found that CTS can fully exert an adsorption bridging effect, and the growth of flocs and the median particle size were significantly improved relative to PAC.

In addition, the flocs produced were broken in the study. The breakage and regrowth of the flocs are shown in the second half of Fig. 6. It can be seen from the figure that the flocs had limited ability to regrow after being broken and exhibited incomplete reversibility. However, it can be seen from the figure that the flocs formed by $AlCl_3$ (dominated by charge neutralization) had stronger regrowth ability after being broken than composite coagulants (dominated by adsorption bridging), which was similar to the research results of Yu et al. [29] Yu et al.'s [29] research showed that the kaolin flocs formed by charge neutralization regrew after being broken. Although the reasons for the irreversible breakage of flocs are not fully understood, it has been found that the complete regrowth of flocs can be achieved by adding very small dosages of coagulants at the breaking stage. This means that the presence of new precipitates was of some importance.

3.4. Floc structure comparison and mechanism

Due to the different formation mechanisms of flocs, the corresponding microstructures also showed certain

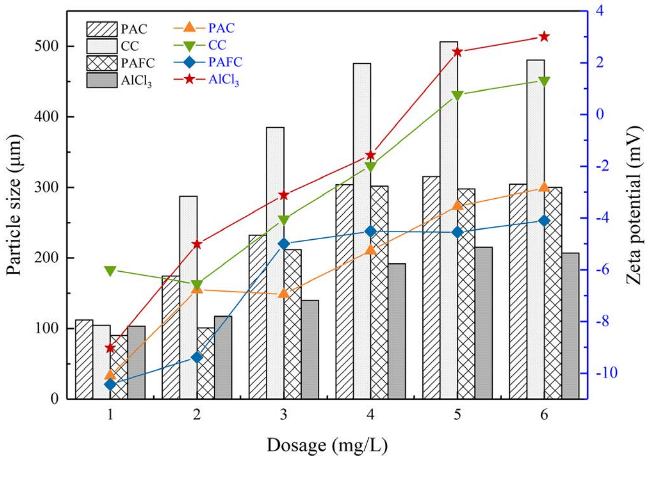

Fig. 5. Changes in particle size under different coagulant conditions (CC: composite coagulant).

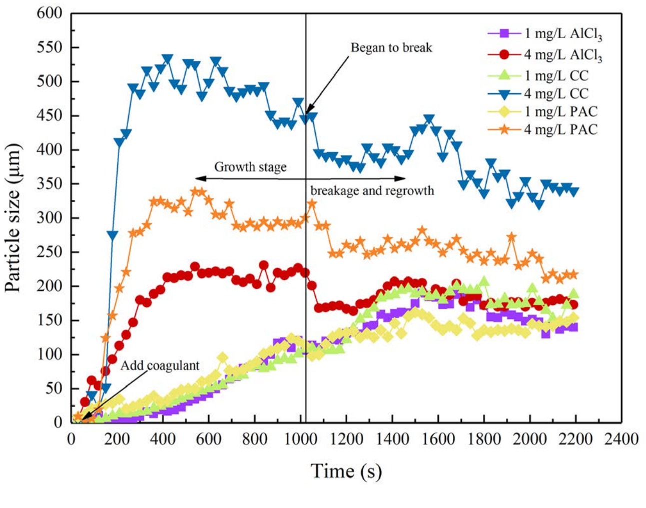

Fig. 6. Changes in flocs under different coagulant conditions (CC: composite coagulant).

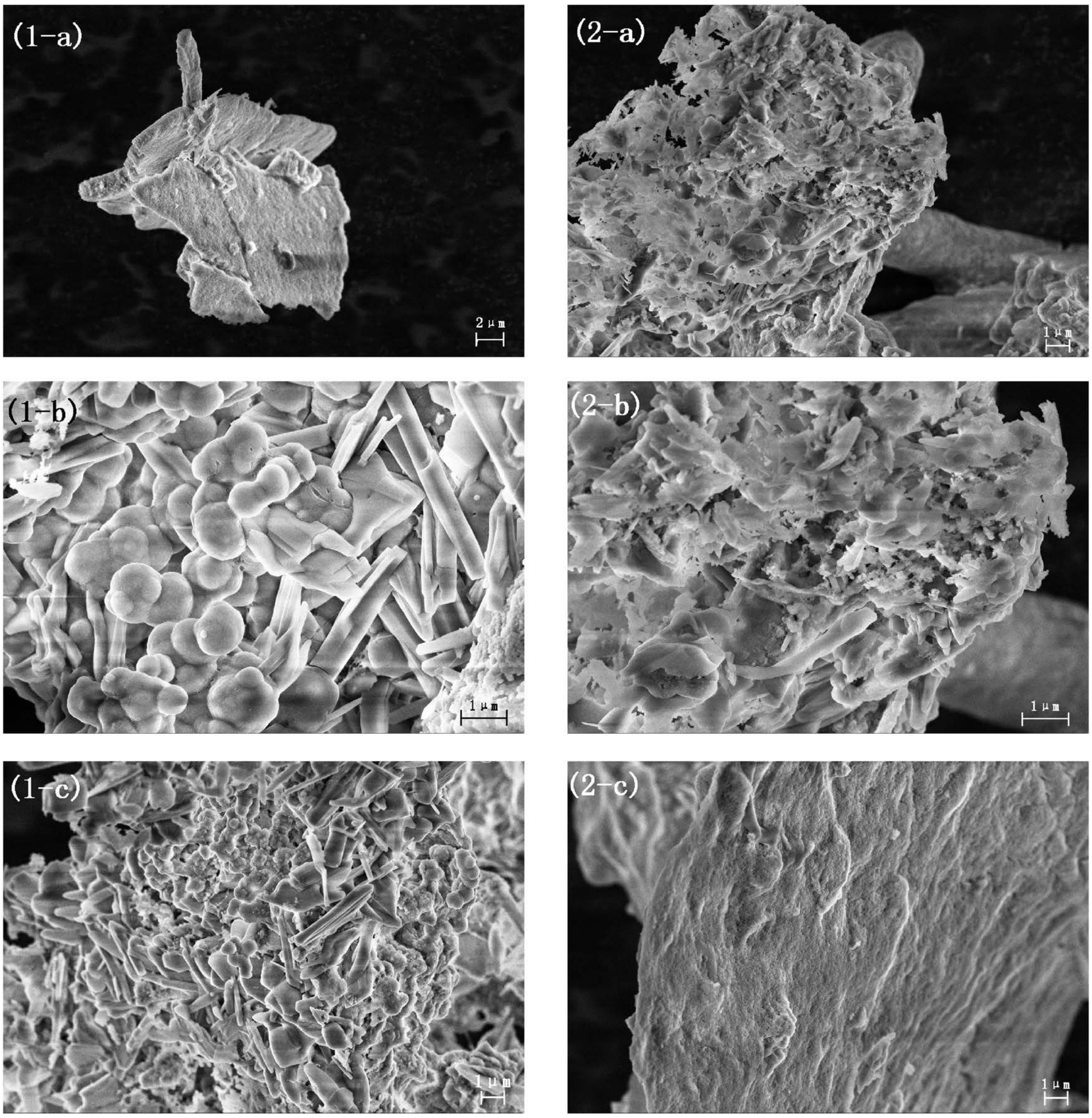

Fig. 7. SEM of flocs produced under different coagulant conditions: (1a–c) the surface of the floc produced by using $AlCl_3$ and (2a–c) the surface of the floc produced by the composite coagulant.

differences. In this study, the flocs formed under different dominant mechanisms were analyzed by SEM. The magnification varied from 1.00 to 10.00 KX, and the difference in the surface structure of the flocs was compared. The SEM images of the flocs produced by different coagulants under the same conditions after freeze-drying are shown in Fig. 7.

In the absence of long-chain bridging, the formation of the floc mainly depended on the addition of an aluminum salt coagulant. The aluminum ion or polymer neutralized the charge on the surface of the colloid and destroyed its stability so that the colloid broke through the energy barrier by contact collision. Therefore, this situation was a colloidal condensation produced by charge neutralization alone [30]. It can be found from Figs. 7(1-b) and (1-c) that when $AlCl_3$ was used (charge neutralization was the dominant mechanism), the flocs formed were mostly lamellar or blocky with uneven surfaces. At the same time, a rod-like structure was also exhibited, the structural shape was slightly uneven, and the gap between the structures was large. When the composite coagulant was used (adsorption bridging was the dominant mechanism), the floc surface was smooth (Fig. 7(2-c)), and there was a small amount of pore structure. When composite coagulants were used (adsorption bridging was the dominant mechanism), the floc surface formed was relatively smooth (Fig. 7(2-c)), with a small amount of pore structure and adhesion between particles (Figs. 7(2-a) and (2-b)). The floc structure was relatively dense, and the small flocs were wrapped, which had a stronger adsorption capacity. These observations explained why the composite coagulant had a better coagulation effect and formed larger floc sizes than the other coagulants.

In general, the bridging effect of polymers played a very important role in the formation of flocs. It effectively concentrated cell particles in flocs [31]. At the same time, particles with poor regrowth ability or inactivity were adsorbed on the surface of the flocs. This process effectively removed particles that cannot be effectively removed when there was little to no bridging effect.

4. Conclusion

This paper evaluated several different Al-based coagulants by comparing basic water quality index, floc characteristics, and floc structure changes. In addition, the influence of different dominant coagulation mechanisms on the coagulation process and their effects were also studied. Conclusions were drawn as follows:

- When the coagulant was dominated by charge neutralization, under a low dosage, the removal effect of organic matter was not as good as that of adsorption bridging. However, with an increased coagulant dosage, this gap gradually narrowed. At the same time, the charge neutralization effect was more beneficial for removing UV_{254} and TOC in water, and the removal effect on humic acid was also better. When adsorption bridging was dominant, the removal effect of the coagulant on turbidity, COD, and SMP was better.
- In the growth stage of flocs, the flocs formed by adsorption bridging were superior to the flocs formed by charge neutralization in terms of growth rate, particle size, and strength. The effect can be improved to some extent by enhancing the bridging. However, after the flocs were broken, the regrowth ability of the flocs formed by the charge neutralization was better. At the same time, the SEM analysis of the flocs under the same conditions also confirmed that the bridging effect of long chain molecules played an important role in the formation of flocs. The flocs formed by this effect were more compact than those formed by charge neutralization.

Acknowledgments

This work was financially supported by the Science and Technology Development Plan of Weifang (2019ZJ1088), the Natural Science Foundation of Shandong Province (ZR2016EEM32). There was no conflict of interest in this study.

References

[1] S. Ke, J. Tu, J. Zhu, W. Wei, Effect of PAC hydrolysis species on coagulation efficiency and floc characteristics, Chin. J. Environ. Eng., 11 (2017) 733–738.

[2] L. Wang, Y. Chen, S. Du, X. Ma, M. Cheng, Z. Tan, Experimental study on the composite coagulant for the enhanced treatment of domestic sewage, Ind. Water Treat., 2 (2017).

[3] L. Liu, C. Wu, Y. Chen, H. Wang, Preparation, characterization and coagulation behaviour of polyferric magnesium silicate (PFMSi) coagulant, Water Sci. Technol., 75 (2017) 1961–1970.

[4] Y.X. Zhao, B.Y. Gao, B.C. Cao, Z.L. Yang, Q.Y. Yue, H.K. Shon, J.H. Kim, Comparison of coagulation behavior and floc characteristics of titanium tetrachloride ($TiCl_4$) and polyaluminum chloride (PACl) with surface water treatment, J. Environ. Eng., 166 (2011) 544–550.

[5] W.U. Yan, Y.L. Yang, L.I. Xing, Z.W. Zhou, W.Q. Wang, S.U. Zhao-Yang, Study on flocs characteristics under three common dominant coagulation mechanisms, China Environ. Sci., 34 (2014) 150–155.

[6] W. Chen, H. Zheng, J. Zhai, Y. Wang, W. Xue, X. Tang, Z. Zhang, Y. Sun, Characterization and coagulation–flocculation performance of a composite coagulant: poly-ferric-aluminum-silicate-sulfate, Desal. Water Treat., 56 (2015) 1776–1786.

[7] R. Jiao, H. Xu, W. Xu, X. Yang, D. Wang, Influence of coagulation mechanisms on the residual aluminum – the roles of coagulant species and MW of organic matter, J. Hazard. Mater., 290 (2015) 16–25.

[8] X. Shen, B. Gao, K. Guo, Q. Yue, Characterization and influence of floc under different coagulation systems on ultrafiltration membrane fouling, Chemosphere, 238 (2020), doi: 10.1016/j.chemosphere.2019.124659.

[9] Editorial Board of Water and Wastewater Monitoring and Analysis Method of State Environmental Protection Administration, Methods for the Analysis of Water and Wastewater, 4th ed., Supplement, China Environmental Science Press, China, 2002.

[10] R.F. Packham, Some studies of the coagulation of dispersed clays with hydrolyzing salts, J. Colloid Sci., 20 (1965) 81–92.

[11] P. Loganathan, M. Gradzielski, H. Bustamante, S. Vigneswaran, Progress, challenges, and opportunities in enhancing NOM flocculation using chemically modified chitosan: a review towards future development, Environ. Sci. Water Res. Technol., 6 (2020) 45–61.

[12] S. Gao, Study on the Application of Multiple Flocculants in the Treatment of Low Temperature and Low Turbidity Water from Hongfeng Lake, Wuhan Institute of Technology, China, 2018.

[13] Y. Luo, B. Gao, J. Wang, Q. Yue, Synchronous removal of CuO nanoparticles and Cu^{2+} by polyaluminum chloride-Enteromorpha polysaccharides: effect of Al species and pH, J. Environ. Sci., 88 (2020) 1–11.

[14] Y.X. Zhao, S. Phuntsho, B.Y. Gao, Y.Z. Yang, J.-H. Kim, H.K. Shon, Comparison of a novel polytitanium chloride coagulant with polyaluminium chloride: coagulation performance and floc characteristics, J. Environ. Manage., 147 (2015) 194–202.

[15] Y. Wang, W. Wang, R. Jia, M. Li, B. Liu, K. Zhang, W. Song, J. Jia, Research on treating algae-polluted reservoir water by the process of pre-oxidation/dissolved air flotation/carbon sand filter, Water Supply, 19 (2018) 823–830.

[16] Y.L. Wang, X. Xu, R. Jia, B. Liu, W. Song, J. Jia, Experimental study on the removal of organic pollutants and NH_3–N from surface water via an integrated copolymerization air flotation-carbon sand filtration process, J. Water Supply Res. Technol. AQUA, 67 (2018) 506–516.

[17] W. Huang, H. Chu, B. Dong, M. Hu, Y. Yao, A membrane combined process to cope with algae blooms in water, Desalination, 355 (2015) 99–109.

[18] H.K. Hudnell, The state of US freshwater harmful algal blooms assessments, policy and legislation, Toxicon, 55 (2010) 1024–1034.

[19] D.N. Kothawala, C.A. Stedmon, R.A. Müller, G.A. Weyhenmeyer, S.J. Köhler, L.J. Tranvik, Controls of dissolved organic matter quality: evidence from a large-scale boreal lake survey, GCB Bioenergy, 20 (2014) 1101–1114.

[20] R. Yu, R. Liu, D. Zhang, X. Hui, N.W. Coltd, Investigation of the coagulation process in the Ning Dong Water Treatment Plant during abnormal water quality in spring, Water Technol., 11 (2017) 11–16.

[21] S. Yu, X. Huang, Photodegradation of soluble microbial products (SMPs) from membrane bioreactor by GO-COOH/TiO_2/Ag, J. Environ. Sci., 88 (2020) 292–300.

[22] W.P. Cheng, Comparison of hydrolysis/coagulation behavior of polymeric and monomeric iron coagulants in humic acid solution, Chemosphere, 47 (2002) 963–969.

[23] S.K. Kam, J. Gregory, The interaction of humic substances with cationic polyelectrolytes, Water Res., 35 (2001) 3557–3566.

[24] N. Wang, J. Ma, Property of Humic Substances in Coagulation and Pre-Oxidation Processes and the Impact on Water Treatment Effectiveness, Harbin Institute of Technology, China, 2012.

[25] L.L. Zhou, Y.J. Zhang, Y.E. He-Xiu, Y.Q. Zhang, Comparison study of enhanced coagulation on humic acid and fulvic acid removal, Environ. Sci., 33 (2012) 2680–2684.

[26] R. Jiao, R. Fabris, C.W.K. Chow, M. Drikas, J. van Leeuwen, D. Wang, Z. Xu, Influence of coagulation mechanisms and floc formation on filterability, J. Environ. Sci., 57 (2017) 338–345.

[27] J. Galloux, L. Chekli, S. Phuntsho, L.D. Tijing, S. Jeong, Y.X. Zhao, B.Y. Gao, S.H. Park, H.K. Shon, Coagulation performance and floc characteristics of polytitanium tetrachloride and titanium tetrachloride compared with ferric chloride for coal mining wastewater treatment, Sep. Purif. Technol., 152 (2015) 94–100.

[28] J. Zhao, S. Wang, J. Wang, Y. Zhang, Seawater coagulation process based on floc properties and stability kinetics, Chin. J. Environ. Eng., 9 (2015) 2840–2846.

[29] W. Yu, J. Gregory, L. Campos, The effect of additional coagulant on the re-growth of alum–kaolin flocs, Sep. Purif. Technol., 74 (2010) 305–309.

[30] W. Wei, M. Du, X. Li, Experimental Research on the Performance of Enhanced Flocculation Based on Bottom Sludge Addition, Harbin Institute of Technology, China, 2015.

[31] W. Zhiyuan, W. Chao, W. Peifang, Q. Jin, H. Jun, A. Yanhui, W. Baohai, The performance of chitosan/montmorillonite nanocomposite during the flocculation and floc storage processes of *Microcystis aeruginosa* cells, Environ. Sci. Pollut. Res., 22 (2015) 11148–11161.