Performance of chitosan/PAC enhanced coagulation for low-temperature and low-turbidity Pi River source water

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Abstract

This work aims to investigate the effect of combined polyaluminum chloride (PAC) and chitosan (CTS) on improving coagulation performance in treating special water, such as low-temperature and low-turbidity water. Furthermore, the study aims to elucidate the mechanism of action involved in applying PAC and CTS in the removal of turbidity, organic matter, and other pollutants. A coagulant of chitosan (CTS)/polyaluminum chloride (PAC) was formulated for the treatment of low-temperature and low-turbidity Pi River source water, located in the southwest of Anhui Province. PAC was used as the main coagulant, and CTS was used as the coagulant aid. The effects of the compounding ratio of CTS/PAC, agitation intensity during coagulation, and sedimentation time on turbidity reduction, decolorization, and organic matter removal were investigated through coagulation beaker tests. Finally, the electric potential after coagulation was analyzed, and the structure of CTS and PAC was characterized. Results showed that the highest removal efficiency of turbidity, chromaticity, and ultraviolet absorbance at 254 nm (UV 254) was obtained when the volume ratio of CTS/PAC was 0.4:1, reaching 86.32%, 16.98%, and 32.79%, respectively, which was 17.9%, 3.77%, and 8.2% higher than those of PAC alone. The optimal agitation intensity was attained at a mixing speed of 300 rpm and a flocculation speed of 150 and 100 rpm. Besides, sedimentation time showed the most significant impact on turbidity. Moreover, scanning electron microscopy and zeta potential results confirmed that PAC coagulation mainly relies on electrical neutralization, while CTS coagulation is highly attributed to adsorption and bridging. The results suggest the formation of a new structure via the compounding of CTS and PAC, which further strengthened the electrical neutralization and bridging and netting capabilities, consequently improving the coagulation effect.

Keywords: Pi River; Low-temperature and low-turbidity; Polyaluminum chloride; Chitosan; Enhanced coagulation

1. Introduction

Lu’an City is located in the western part of Anhui Province, China, between the Yangtze River and the Huai River, at the northern foot of the Dabie Mountains. It serves as an essential drinking water source in the western region of Anhui Province. The Pi River main canal basin provides drinking water not only for the urban area of Lu’an City but also for Hefei City and Huainan City, ensuring the life, health, and safety of the people in those cities. However, the Pi River source water is a typical low-temperature and low-turbidity water [1] with an average turbidity of less than 10 NTU and a temperature of 10°C in winter. Consequently, the requirements for influent water quality in winter are challenging to be met using conventional processes, reducing the drinking water quality [2]. Therefore, more effective methods are required for treating low-temperature and low-turbidity water, compared to conventional processes, such as enhanced coagulation, membrane treatment, sludge reflux, dissolved air flotation, pre-oxidation, and...
Li and Zhou et al. analyzed the effects of PAC, old alum reaction and a higher sedimentation efficiency, respectively, indicating a better cost performance than that of the pre-filtration water decreased by 51.5% and 38%, respectively. The study showed that the turbidity of effluent water and temperature and low-turbidity micro-polluted melting water from the Qinghai-Tibet Plateau in northwest China and investigated coagulants based on conventional processes or the enhancement of coagulants, mixing processes or formulas, coagulation, and flocculation in conventional processes, further improving the purification effect. Polyaluminum chloride (PAC) coagulation is the most commonly used coagulation method due to its low cost and strong sensitivity to low temperatures. For instance, Xu et al. and Peng et al. used PAC as the coagulant and polyacrylamide (PAM), sodium alginate, and hydroxypropyl methylcellulose as the coagulant aids to effectively remove turbidity and chromaticity; the study revealed that larger flocs have a fast sedimentation speed because the compounds ratio of coagulants affects the treatment of low-temperature and low-turbidity water, thus forming impurity flocs with large density and volume. Zhang and Li [10] and Wang et al. [14] optimized the coagulants and coagulant aids commonly used in water plants for the treatment of low-temperature and low-turbidity water. The results showed that the combination of PAC and poly aluminum ferric chloride (PAFC) with activated silicic acid exhibit the best effect on turbidity and pollution removal of low-temperature and low-turbidity water. The combination of PAC and activated silicic acid can be more suitable for use in water plants due to the relatively low cost. Barkács et al. [11] and Zhong [13] studied the effect of PAM and PAC on the removal efficacy of turbidity and dissolved organic matter (DOM) in drinking water. The study found that PAC is more cost-effective than PAM, and it can maintain the effluent quality while enhancing the stability of the flocculation process. Compared to inorganic small-molecule flocculants, PAC is more economical and has fewer residues. Yuan et al. [12] and Zhong [13] used several flocculants to treat low-temperature and low-turbidity micro-polluted melting water from the Qinghai-Tibet Plateau in northwest China and investigated the removal ability of various flocculants on turbidity and organic matter in micro-polluted water samples. When PAC is used as the main coagulant in practice, coagulant aids are usually added to further improve the coagulation performance and ensure a low concentration of residual Al in water. Studies have shown that compound coagulants, which are formed by adding PAC to a polyelectrolyte, are more efficient in removing colloidal suspensions than PAC alone, as the positive aspects of their components are integrated, resulting in an optimized coagulation efficiency flocc density, and accelerated sedimentation speed. Peng et al. [9] and Yan [21] treated low-temperature and low-turbidity water samples with a new compound high-efficiency PAC water purification agent. The study showed that the turbidity of effluent water and pre-filtration water decreased by 51.5% and 38%, respectively, indicating a better cost performance than that of the old alum reaction and a higher sedimentation efficiency. Li [22] and Zhou et al. [23] analyzed the effects of PAC, PAFC, PAC + PAM, PAC + modified activated silicic acid, PAFC + PAM, and PAFC + modified activated silicic acid on Songhua River water in low-temperature and low-turbidity period. The test results indicated that PAC + PAM and PAC + modified activated silica had significantly better coagulation effects than other coagulants, reducing the initial turbidity from 7.78 to 0.21 NTU. Zhang et al. [24] found that the integration of PAC with a high basicity (90.3%) and chitosan (CTS) with a high viscosity (500 mPa.s) effectively removed turbidity and natural organic matter (NOM), with removal efficiencies of approximately 87%, 63%, and 82% for turbidity, DOM, and ultraviolet absorbance at 254 nm (UV254), respectively. Among the numerous polymer coagulants, CTS, a natural biopolymer, is a safe and non-toxic coagulant with excellent performance. Chitosan has electrical neutralization ability and strong adsorption and bridging ability, showing good adsorption and coagulation on metal ions [25], dyes [26], and DOM [27] in water, which has received great interest in the recent decades. Previous studies have also shown that CTS combined with aluminum-based salts/polymer effectively removes turbidity. Additionally, the residual Al concentration is found to be much lower than that obtained when PAC is used alone as a coagulant [24]. However, investigating the effect of combined PAC and CTS on improving coagulation performance in treating special water, such as low-temperature and low-turbidity water, has not been sufficiently studied. This work aims to investigate the effects of different CTS/PAC ratios, hydraulic conditions, and sedimentation time on coagulation performance and treating low-temperature and low-turbidity water. In addition, the study aims to elucidate the mechanism of action involved in applying PAC and CTS to remove turbidity, organic matter, and other pollutants.

2. Materials and methods
2.1. Materials
2.1.1. Main instruments
The following instruments were used: T6 UV-Vis spectrophotometer (Beijing Persee General Instrument Co., Ltd., China), Nano ZS90 nanoparticle size and zeta potential analyzer (MALVERN Instruments, UK), Sigma 300 field-emission scanning electron microscope (Zeiss, Germany), 2100Q Portable Turbidimeter Hach Water Quality Analysis Instrument (Shanghai) Co., Ltd., China, ZR4-6 coagulation test mixer (Guangzhou Ruifeng Equipment Co., Ltd., China), PHS-3D pH tester (Hangzhou Qiwei Instrument Co., Ltd., China), SD9012A colorimeter (Shanghai Xinrui Apparatus & Instrument Co., Ltd., China), FA2204N analytical balance (Suzhou Keruisi Instrument Co., Ltd., China), and magnetic agitator (Qiwei Instrument Co., Ltd., China).

2.1.2. Main reagents
The main reagents included: CTS (degree of deacetylation ≥95%, viscosity 100–200 mPa.s) produced by Shanghai McLean Co., Ltd., PAC (Al₂O₃ content 28%, basicity 88.3%) produced by Botao Water Purification Materials Co., Ltd., and acetic acid (AR) and concentrated sulfuric acid produced by Xilong Scientific Co., Ltd.
2.2. Water samples for analysis

The raw water was taken from the Pi River main channel of the Gaocheng Road Bridge in Lu’an, Anhui. The samples were obtained between December 20, 2022, and January 25, 2023. The raw water quality conditions are shown in Table 1.

2.3. Preparation of flocculants

In brief, 5 g of PAC were accurately weighed and added into a beaker containing 200 mL distilled water and agitated with a magnetic agitator for about 30 min. Finally, the obtained solution was diluted with distilled water to 500 mL solution volume to prepare a PAC solution with a mass concentration of 10 g/L. 1 g of CTS was accurately weighed, added into a beaker with 200 mL of AR solution (with a volume fraction of 1%), and agitated for about 30 min with a magnetic agitator. After complete dissolution, the obtained solution was diluted to 500 mL with the AR solution to obtain CTS AR solution with a final concentration of 2 g/L [13].

2.4. Analytical methods

The test indicators were determined by referring to the detection methods in Standard Examination Methods for Drinking Water (GB/T5750-2023). In brief, the pH value was measured by a pH meter, turbidity was determined by a turbidimeter, UV254 was determined via UV spectrophotometry, the chromaticity was determined by a colorimeter, zeta potential was measured on a NanoZS-90 nanoparticle size, and zeta potential analyzer and the microstructure of the coagulant was characterized by the Sigma 300 field emission scanning electron microscope [30].

2.5. Coagulation test

The programmable ZR4-6 intelligent coagulation equipment was utilized for coagulation. Specifically, 1 L of raw water from the Pi River was first poured into a 1-L circular beaker and added with the coagulant, followed by mixing with an agitator at a speed of 300–550 rpm for 30 s, 150–250 rpm for flocculation for 4 min, and 100 rpm for flocculation for 4 min. The obtained liquid was then settled still for 5–50 min. Finally, samples were taken and determined at 3 cm below the water surface in 1-L circular beakers. The procedures of coagulation and sedimentation test are shown in Table 2.

3. Results and discussion

3.1. Influence of single flocculant addition on flocculation performance

As can be seen from Fig. 1, when the dosage of PAC increased from 0.5 to 1.0 mL, the removal rates of turbidity increased from 18.38% to 81.57%, chromaticity rose from 2.47% to 16.67%, and UV254 increased from 3.28% to 26.23%, increasing by 63.19%, 14.20%, and 22.95%, respectively. When the dosage of PAC increased from 1.0 to 1.5 mL, the three indicators increased by 3.89%, 1.85% and 8%, respectively. At the dosage of 1.5 mL, the removal rate of chromaticity remained unchanged, while the increase in removal size, and zeta potential analyzer and the microstructure of the coagulant was characterized by the Sigma 300 field emission scanning electron microscope [30].

Table 1

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Table 2

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rates of turbidity and UV$_{254}$ was less than 2%. When further increasing the dosage of PAC to 2.5 mL, the removal efficiency of turbidity and UV$_{254}$ began to decline, and that of chromaticity remained unchanged, indicating that a careful dose is required for an optimum coagulation effect. The main reason is that a small dosage of flocculant results in a small number of positive ions provided by PAC, which cannot fully generate electrical neutralization with the negatively charged colloidal particles in the water. In addition, the formed flocculant has a small particle size and is suspended in water, which cannot overcome the buoyancy and thus settle. On the contrary, excessive dosage results in full coverage of the colloidal particles, which keeps the particles deflocculated, resulting in the phenomenon of colloidal protection [13,31]. Furthermore, according to the adsorption principle, the flocculation effect is best when the polymer coverage rate on the surface of the colloidal particles is 1/2 [13,32]. Therefore, excessive addition is not conducive to the flocculation effect. Taking into account the removal effect and cost, the best dosage of PAC should be about 15 mg/L.

It can be observed from Fig. 2 that when increasing the dosage of CTS from 0.5 to 2 mL, the turbidity, chromaticity, and UV$_{254}$ increase to a certain limit and then decrease. The removal rates of turbidity, chromaticity, and UV$_{254}$ are best at a dosage of 1.7 mL, with a percentage increase of 27.09%, 15.31%, and 19.64%, respectively, showing that the removal rates are not ideal when at very low or very high CTS dosages. The main reason is that the flocculation mechanism of the CTS flocculant is mainly based on adsorption, bridging and group reaction, followed by electrical neutralization. Moreover, the flocculation mechanism of the CTS flocculant varies for different treatment samples. For example, the flocculation mechanisms of the CTS flocculant on reservoir water and river water are different [32,35]. At very low dosages, it is difficult to form flocs or fewer flocs are formed. Conversely, when excessive flocculants are added, all active groups on the particle surface in water are completely covered by CTS molecules, preventing the flocculation of the particles through bridging. Simultaneously, the flocculation efficiency will be reduced [33,34,36].

3.2. Effect of different CTS/PAC ratios on flocculation performance

In order to maintain a total dosage of 1.5 and 1 mL of PAC with a mass concentration of 10 g/L and 0–0.5 mL of CTS with a mass concentration of 2 g/L were added. CTS/PAC flocculants with volume ratios of 0:1, 0.1:1, 0.2:1, 0.3:1, 0.4:1, and 0.5:1 were added under agitation using a six-link agitator. After 30 min of sedimentation, the corresponding indicators were tested to compare the effect of flocculation. As can be observed from Fig. 3, When increasing the CTS dosage, the removal rates of turbidity and UV$_{254}$ by different volume ratios of CTS/PAC showed a gradual increase behavior where the removal rates of turbidity, UV$_{254}$ and chromaticity were increased by 17.9%, 8%, and 3.77%, respectively, at volume ratios of 0:1–0.1:1. When the volume ratio was 0.5:1, the removal rates of turbidity and UV$_{254}$ decreased slowly, while the chromaticity remained unchanged. Besides, flocs formation was observed when increasing CTS, reducing sedimentation time. This behavior could be attributed to two main reasons. First, the positive charge on the CTS chain is superimposed with the positive charge of PAC, which enhances the electric neutralization ability of polyalumina. Second, the bridging of molecular chains of organic polymers between the destabilized particles is conducive to the formation of larger flocs [13], and the ability to remove tiny particles in water is enhanced through the sweeping effect of flocs [33]. It was also found that when increasing the proportion of CTS to a certain extent, the flocs became relatively sparse, so a greater proportion of CTS did not result in a better removal effect. This could be due to the Al-NH$_2$ chemical bonds between CTS and PAC, where their interaction requires that the CTS and PAC should be mixed according to a certain proportion to achieve a better removal effect [36]. Considering the removal efficacy and cost, the optimal volume ratio of CTS/PAC should be 0.4:1.
3.3. Influence of different agitating intensities on flocculation performance

CTS/PAC with a volume ratio of 0.4:1 and a dosage of 1.4 mL, that is, CTS of 0.4 mL and PAC of 1 mL, was tested. The coagulation test was conducted using Procedures 1–6. Finally, the coagulation effect was evaluated by superposing the removal rates of turbidity, chromaticity, and UV$_{254}$ as shown in Fig. 4.

Fig. 4 and Table 2 show that, under similar mixing and flocculation time, different coagulation effects could be observed at different speeds at the mixing and flocculation stages in the agitation procedure. Procedure 2 resulted in the best coagulation effect, followed by Procedures 4, 3, 1, 5, and 6. In Procedure 6, a rotation speed of 550 rpm in the mixing stage and 250 rpm in the first stage of flocculation were both the highest; however, the coagulation effect was poor, being the lowest. Differently, in Procedure 2, the rotation speed was 300 rpm in the mixing stage and 150 rpm in the first stage of flocculation, which was relatively moderate, achieving the best coagulation effect. Moreover, the coagulation effect of other procedures was closely related to the rotation speed of the mixing stage. At high mixing speed, the coagulation was worse, and vice versa. This is mainly attributed to the high charge, small volume, small deformability, and weak adhesive strength between aluminium ions and the colloidal particles after the addition of PAC. Thus, a moderate rotation speed is required because a moderate rotation speed is more conducive to the collision and integration of particles, thus forming flocs that are gradually becoming larger with a good coagulation effect. In the case of low turbidity, there are few impurities and colloidal particles in the water. The formed micro-flocculation particles might be disrupted to a certain extent when the water is strongly agitated during the mixing stage and thus affecting the subsequent flocculation and ultimately exhibiting a moderate or even decreasing effect.

3.4. Influence of static sedimentation time on flocculation performance

CTS/PAC with a volume ratio of 0.4:1 and a dosage of 1.4 mL was selected for coagulation tests with Procedure 2. The turbidity, chromaticity, and UV$_{254}$ were determined at a sedimentation time from 5 to 50 min in a 5-min interval. The results are shown in Fig. 5.

Fig. 5 shows that the sedimentation time had a great influence on the turbidity and UV$_{254}$ of the low-temperature and low-turbidity Pi River raw water but had little effect on its chromaticity. After 5 min of sedimentation, the removal rates of turbidity, UV$_{254}$, and chromaticity were very low, with only 52.27%, 22.22%, and 11.18%, respectively. After 30 min of sedimentation, the removal rates of turbidity,
UV$_{254}$ and chromaticity were increased, with the removal efficiency of turbidity being the best, of not more than 80%. Although enhancing the removal rates of turbidity, the removal efficiency of chromaticity decreased. After 40 min of sedimentation, the removal rate of turbidity exceeded 80% for the first time, reaching 81.71%. However, the removal rates of UV$_{254}$ and chromaticity remained very low, at only 35.19% and 13.82%, respectively. After 50 min of sedimentation, the removal rate of turbidity increased slowly to 84.30%, but those of UV$_{254}$ and chromaticity remained stable. In the sedimentation process, when a sedimentation time is less than 30 min, the removal rates of turbidity and UV$_{254}$ slowly increased with the increasing time, however with a slow increase rate. After 40 min of sedimentation, the removal rate of turbidity slowly increased while those of the chromaticity and UV$_{254}$ remained unchanged. The increase in sedimentation time was mainly ascribed to the fact that the hydrolysis rate of the coagulant is reduced under low-temperature and low-turbidity conditions, the thermal movement of particles in the water is weakened, the number of collisions between particles is also reduced, and the formed flocs are relatively light, all of which require more time to complete. Therefore, to achieve a better flocculation effect, it is recommended to control the sedimentation time of low-temperature and low-turbidity water at around 40 min.

3.5. Mechanism of enhanced CTS/PAC coagulation treatment of low-temperature and low-turbidity Pi River water

3.5.1. Zeta potential

The destabilization state of colloidal particles is closely related to the charging state of the coagulant added, and the electrophoretic properties of the coagulant can be well characterized by zeta potential [30]. The zeta potential values after adding PAC and CTS/PAC to the raw water are shown in Figs. 6–9.

From Figs. 6–8, it can be seen that when increasing the dosage of PAC from 0.5 to 1 mL, a significant increase in the electric potential of the raw water of the Pi River was observed, with an increase from −14.57 to −3.9 mV. The hydrolysis of the coagulant PAC exhibited strong electrical neutralization ability. This is mainly due to the hydrolysis of AL$_3^+$ into mononuclear complexes such as AL(OH)$_2^+$, AL(OH)$_3^+$, and ALO$_2^-$ when PAC is dispersed in water. Then the mononuclear complexes undergo further condensation through collision, forming a series of polynuclear complexes AL$_{n}$ (OH)$_{m}$ $^{(3-3n)}$ ($n > 1$, m ≤ 3n) [37], of which AL$_{13}$(OH)$_{34}^{5+}$ plays a major role in the flocculation process. Polynuclear complexes often have high positive charges and large specific surface areas, which can rapidly adsorb negatively charged impurities in water and thus neutralize colloidal charges, compress the electric double layer, reduce the colloidal electric potential, and promote the rapid destabilization, agglomeration and sedimentation of colloids and suspended solids, resulting in enhanced effects [36–38].
As can be seen from Fig. 9, the addition of compounding coagulant of CTS/PAC with a volume ratio of 0.4:1 increases the electric potential in raw water compared to that obtained from the addition of a single coagulant of PAC, confirming that the presence of CTS increases the cation content in the system and enhances the electrical neutralization ability of PAC [13]. This finding is in good agreement with Renault et al.’s view that the cation concentration generated by CTS hydrolysis gives PAC a strong flocculation ability that may be manifested through electrical neutralization and other effects at lower doses [13,39].

### 3.5.2. Scanning electron microscopy

Scanning electron microscopy (SEM) analyses of CTS and PAC solid powders are shown in Figs. 10 and 11.

Fig. 10 shows that the microstructure of PAC was relatively simple, with no specific shape structure on the surface and a small amount of cracking distribution, showing a layer-like superimposed structure. Besides, it exhibited a relatively tight connection between the layers due to the active components generated, such as hydroxyl aluminum chloride polynuclear complexes. Moreover, it indicated that hydroxyl aluminum complexes with high coagulation and adsorption activity are generated on the surface of the PAC coagulant, with a large specific surface area [40], which plays a role in electric neutralization during coagulation.

This conclusion was also verified by the zeta potential measurements. It can be seen from Fig. 11 that the surface morphology of CTS particles was obviously different compared to
to PAC, showing a porous floculent structure and relatively loose and rough surface, with many folds and bulges, which facilitated the penetration of the solution into the floculent structure, coinciding with its strong adsorption and bridging ability [40]. It was speculated that the electric neutralization and adsorption and bridging of both might exist under the condition of low temperature and low turbidity, thereby improving the coagulation effect.

4. Conclusions

When treating low-temperature and low-turbidity Pi River raw water, the compounding of CTS and PAC with a specific viscosity can effectively change its coagulation performance under certain conditions. The following conclusions were drawn:

- When the turbidity of the raw water is less than 10 NTU, the temperature is less than 10°C, and the pH value is 7.1~7.3, the removal efficiency of CTS/PAC with different volume ratios is different under the same dosage. When the volume ratio is 0.4:1, the highest removal efficiency of turbidity, chromaticity and UV$_{254}$ can be achieved when compared with the addition of PAC solely.
- For low-temperature and low-turbidity Pi River water, a higher agitation intensity does not lead to a better treatment effect during the mixing and flocculation stages. The optimal agitation intensity is a mixing speed of 300 rpm, and a flocculation speed is 150 and 100 rpm, respectively.
- Under the conditions of low temperature and low turbidity, sedimentation time poses the greatest impact on turbidity. Within the first 5 min, turbidity, chromaticity, and UV$_{254}$ hardly changed. After 40 min of sedimentation, turbidity decreases significantly while the chromaticity and UV$_{254}$ remain unchanged. Therefore, it is more appropriate to control the sedimentation time at 40 min.
- Based on SEM and zeta analyses, it is found that the coagulation of PAC is mainly based on electrical neutralization, and that of CTS is mainly based on adsorption and bridging. It is speculated that both electrical neutralization ability and the ability to bridge and net may exist after mixing and adding CTS and PAC in a certain proportion, which improves the coagulation effect.

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