Coagulation efficiency and removal mechanism for composite coagulant polyaluminium chloride/polydimethyldiallylammonium chloride in treating lightly micro-polluted raw water of Yangtze River in autumn

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The lightly micro-polluted raw water of Yangtze River (YRW) in autumn was treated via enhanced coagulation by composite coagulants composed of polyaluminium chloride (PAC) and polydimethyldiallylammonium chloride (PDMDAAC), named PAC/PDMDAAC. Coagulation mechanism and removal efficiency were investigated by assessing the water quality parameters of the resulting supernatant, i.e. turbidity, COD_{Mn}, and NH_3-N, at the same dosage and supernatant turbidity (SDST) point as using PAC only, with controlled residual turbidity of 1.0–1.50 NTU to mimic drinking water production plant supernatant condition when using lightly micro-polluted water as raw water source . In addition, the zeta potential, floc morphology, and size analysis under the condition of SDST using PAC and composite coagulants PAC/PDMDAAC for getting insight about removal mechanism were done. The results showed that, firstly, most of the composite coagulants PAC/PDMDAAC with intrinsic viscosity [η] = 0.65, 1.60, and 2.6 dL/g; and mass ratio PAC:PDMDAAC of 5:1, 10:1, and 20:1 (m:m)) could meet the requirements of controlled supernatant turbidity between 1.0 and 1.5 NTU to mimic drinking water production plant condition using YRW (Nanjing section) that comply with the new national drinking water standards. Secondly, the seven kinds of composite coagulants PAC/PDMDAAC can maintained the advantage of enhanced coagulation removal efficiency within the SDST point as using PAC only. The COD_{Mn} and ammonia nitrogen removal rates using composite coagulants at SDST points were 0–6.19 %; 0–15.62%, respectively higher than using PAC only. Finally, this study deepened and expanded the existing research knowledge about composite coagulant PAC/PDMDAAC and offered the maximum limitations in removing the water quality parameters via enhanced coagulation treatment of lightly micro-polluted surface raw water in order to meet new national drinking water standards.

INTRODUCTION

Raw water generally refers to surface water from natural water bodies or water storage bodies, such as reservoirs, rivers, lakes, groundwater, etc. It is the natural water source without any artificial treatment, and it is also the water that enters the first unit of the drinking water production plant (Sayato, 1989). Lightly micro-polluted raw water refers to raw water whose water quality index does not comply with the requirements of drinking water source standards, due to the influence of industrial, agricultural, and domestic waste; however, the level of pollution is lower than that expected for what has been termed as micro-polluted water. Over the past hundred years, conventional coagulation procedures have provided cheap and easy-to-operate technology for the production of safe drinking water from natural raw water. However, because of increasing populations and industrialization which result in increasing water pollution, some natural raw water sources have become lightly micro-polluted raw water. At the same time, stringent drinking water regulations have been introduced by national and international bodies to ensure that drinking water is safe for human consumption.

As a commonly used treatment method, the application of inorganic coagulants, such as aluminium and iron salts at controlled pH, has been used to improve the supernatant water quality, while using turbidity as the main control parameter. As a result, excess inorganic coagulant dosage has introduced the problem of not only increased treatment cost, but also residual aluminium and iron content that has exceeded maximum allowable limits set by the World Health Organization for drinking water (WHO, 2008; Yang et al., 2013; Alzahrani et al., 2020). Treatment methods for lightly micro-polluted raw water may also involve pre-oxidation and technologies such as enhanced coagulation, and advanced treatment technology or using membrane and advanced oxidation processes in combination with conventional treatment methods such as coagulation, sedimentation, filtration and disinfection (Yu and Graham, 2015; Brandt et al., 2017; Nascimento et al., 2019; Adebayo et al., 2021). Among these treatment technologies, enhanced coagulation using new coagulants, controlling the pH, or improvement of treatment facilities are the most commonly used, but the most feasible, efficient, and low-cost method is to enhance coagulation by using effective coagulants that have application advantages such as simple operation, and easy implementation which does not necessarily require changing the existing facilities (Moussas and Zouboulis, 2009; Yang et al., 2012 (Tolkou and Zouboulis, 2020; Wang et al., 2020). This is considered to be one of the most important technical options for the effective treatment of lightly micro-polluted surface raw water that is suited to China's national conditions, and is also an important development direction that has been continuously supported in many national 5-year plans of China (Shen et al., 2010a; Sun et al., 2019; Yang et al., 2021).

Usually, various inorganic aluminium and iron salt coagulants are the main coagulants used by drinking water production plants worldwide, and have performed well for many years for many countries (Nowacka et al., 2014; Bratby, 2016; Cui et al., 2020; Adebayo et al., 2021). However, because of the challenge of meeting stringent water regulations and the demand for continuous improvement of water quality, the role those inorganic coagulants or their complexes can play is becoming increasingly limited, given the practical constraints of effectiveness and cost.

Polydimethyldiallylammonium chloride (PDMDAAC) is a water-soluble cationic polymer with unique functional groups enabling interactions favouring coagulation, and is approved for drinking water treatment (Edzwald, 2011). When compared with other linear polymers, it has a high positive charge density with the advantage of good water solubility, easy control of molecular weight (MW) measured as intrinsic viscosity, enhanced removal efficiency at reduced dosage, and is non-toxic (Bolto et al., 1999; Zhang et al., 2009; Zhang et al., 2010; Li et al., 2015; Olukowi et al., 2022). Haarhoff and Cleasby (1989) found that PDMDAAC had a good removal effect for algal extracellular organic matter (EOM) and algal cells during the treatment of algae-polluted raw water. Since then, many researchers have carried out studies on the application of PDMDAAC for many aspects, such as turbidity removal, humic acid removal, total organic carbon removal, floc formation and floc strength (Wang et al., 2008; Liu et al., 2019; Cui et al., 2020). In China, composite coagulants of polyaluminium chloride (PAC) and PDMMAAC with different intrinsic viscosity to PDMDAAC (0.50–3.90 dL/g) and a mass ratio of PAC: PDMDAAC of 5:1–20:1, have been used for the past 2 decades for the improvement of water quality parameter removal, with the advantage of enhanced coagulation, including limited residual aluminium at reduced dosage. The investigations of composite inorganic salt/PDMDAAC coagulant for the enhanced coagulation treatment of high temperature, high algae and low temperature, low and high turbidity, lightly micro-polluted surface raw water showed that enhanced coagulation by composite inorganic-PDMDAAC coagulant has a better sedimentation effect, with stronger, compacted, and easily settled floc compared to that achieved by corresponding inorganic coagulants (Yang et al., 2012; Sun et al., 2019). Furthermore, it greatly improved the removal rate of water quality parameters at lower and optimum dosage ranges, and can replace the pre-chlorination oxidation process, helping to reduce the cost of treating surface raw water. Therefore, research on PDMDAAC is developing rapidly, and significant progress has been made (Zhang et al., 2010; Shen et al., 2017; Adebayo et al., 2021).

In recent decades, researchers have focused on improving the performance and removal efficiency by composite coagulants composed of inorganic salt/PDMDAAC, reporting the effect of dosing method (single or dual dosing), coagulation/application mode, intrinsic viscosity of PDMDAAC and mass ratio (Bolto et al., 2001; Sillanpää et al., 2018; Liu et al., 2019). For instance, some researchers have reported the use of composite coagulants composed of inorganic salt/PDMDAAC to enhance coagulation in the treatment of lightly micro-polluted and highly polluted surface raw waters, focusing on two major coagulation/application modes for the composite coagulants and corresponding inorganic coagulants (Moussas and Zouboulis, 2009; Li et al., 2015; Adebayo et al., 2021), namely: (i) same supernatant turbidity (SST) using different dosages for the composite coagulants and corresponding inorganic coagulant, calculated as Al_3O_2 and Fe^{+n}. In this application mode, the dosage of aluminium or iron salts can be significantly reduced, which can reduce the cost of water treatment and help to reduce the residual aluminium content in the resulting supernatant (Zhang et al., 2010; Adebayo et al., 2021); (ii) same dosage mode (SD) for all coagulants – when this

is applied, it can significantly improve the water quality of the resulting supernatants after coagulation treatment, withstand the impact of a sudden increase in the quantity of surface raw water while ensuring the quality of the supernatant, and replace the pre-oxidation step for high-temperature algae-containing water, leading to a reduction in generated disinfection byproducts, etc. However, these studies were carried out under the requirement of limiting the residual turbidity of the supernatant water to between 2.0 and 4.0 NTU (Yang et al., 2012; Adebayo et al., 2021). Furthermore, some literature reviews have found that the composite inorganic salt/PDMDAAC coagulant can enhance the charge neutralization, polymer-bridging, and net capturing of suspended colloidal particles in the treatment of lightly micro-polluted surface raw water, which strengthens the flocculation/precipitation and adsorption processes resulting in improved water quality parameters (Wei et al., 2009; Bolto et al., 1999; Shen et al., 2017; Olukowi et al., 2022). This enhanced the overall removal efficiency for colloidal particles and other water quality indicators (Yilun, 2019; Adebayo et al., 2021).

Generally, the water quality parameter removal efficiencies for enhanced coagulation by composite inorganic salt/PDMDAAC coagulants were observed to depend on factors such as initial surface raw water quality parameters, the coagulant type, the intrinsic viscosity of PDMDAAC and mass ratio of inorganic salt to PDMDAAC in the composite coagulant mixture. These studies have opened up a new field of application of composite inorganic salt/PDMDAAC coagulants to enhance removal efficiency; however, due to new national drinking water requirements, new studies are needed, as previous studies only addressed the coagulation condition of same dosage (SD) and same supernatant turbidity (SST), as inorganic coagulant at controlled supernatant turbidity of 2.0 NTU or above using lightly micro-polluted and highly polluted surface raw water sources. For instance, in the developed areas of eastern China, including the middle and lower reaches of the Yangtze River, the supernatant turbidity is now required to be controlled between 1.0 and 1.5 NTU to ensure that the turbidity of drinking water is lower than 0.5 NTU and even 0.1 NTU, and residual aluminium must be below the allowable limit of 0.2 mg/L and meet WHO and national drinking water regulations for safe drinking water. In addition, the study of the removal efficiency of composite inorganic salt/PDMDAAC coagulants until now was chiefly for micro-polluted and highly polluted surface raw water sources; however, it has been limited for lightly micro-polluted surface raw water sources for water quality parameters at controlled supernatant turbidity between 1.0 and 1.5 NTU. This study is needed to maximize removal efficiency at a reduced and practical cost, as well as to meet international and national drinking water standards when using lightly micro-polluted raw water as source water.

In view of this, for this study the Yangtze River water in autumn was selected as the lightly micro-polluted raw surface water source. The dosage–residual turbidity tests were performed using jar tests, and curves were obtained to determine the SDST point which was also the intersection point for the composite coagulant PAC/PDMDAAC and PAC coagulants at controlled residual turbidity between 1.0 NTU and 1.5 NTU, while the basic water quality parameters, such as turbidity, COD_{Mn}, and ammonia-nitrogen, of the resulting supernatant samples at these intersection points were measured. Then, based on the measured parameters under the limiting condition of intrinsic viscosity of PDMDAAC, the mass ratios of PAC to PDMDAAC in the various composite coagulants of PAC/PDMDAAC, and the dosages used, insight into maximum removal efficiency points at which the composite coagulants PAC/PDMDAAC can exhibit the most effective water quality parameter removal in comparison to using PAC coagulant alone, under a low supernatant turbidity of less than

1.5 NTU, was obtained. In addition, the determination of zeta potential of raw and resulting supernatant water samples, and the morphology and size of precipitated flocs, aimed to understand the removal mechanism of enhanced coagulation by composite coagulant PAC/PDMDAAC in order to improve the removal rate of water quality parameters. Therefore, the obtained results are expected to give new insights into the application and mechanism of composite coagulants, as their superior enhanced coagulation effect fades away beyond certain dosage limits, in comparison to using inorganic coagulant in the treatment of lightly micro-polluted surface raw water. However, the enhanced coagulation using composite coagulant PAC/PDMDAAC at optimum dosage and new controlled supernatant turbidity will be useful for water professionals and water treatment plants using lightly micro-polluted water as raw water source in meeting national and international drinking water standards.

MATERIALS AND METHODS

Materials

The polyaluminium chloride (PAC) was an industrial product with an Al_2O_3 content (w/w) of 30% (Gonyi Fuyuan Water Purification Materials Co. Ltd.; Henan Province, China). Polydimethyldiallylammonium chloride (PDMDAAC) coagulants were synthesized in our laboratory with measured molecular weights represented as intrinsic viscosity of 0.65, 1.60, and 2.60 dL/g, having solid content of 68.46, 76.38, and 71.30%, respectively. Two program-controlled jar test apparatuses (TA6-2, Wuhan Henling Technology Co. Ltd., China) were used.

Preparation of composite coagulant PAC/PDMDAAC

The composite coagulant PAC/PDMDAAC mixtures with varying intrinsic viscosities of PDMDAAC at 0.65, 1.60, and 2.60 dL/g, and mass ratios of PAC to PDMDAAC at 5:1, 10:1 and 20:1 (PAC calculated as Al_2O_2, $m{:}m$) were prepared (9 mixtures) according to the method in literature (Zhang et al., 2010, 2015; Adebayo et al., 2021). Firstly, a stock solution of PAC coagulant containing 10% Al_2O_3 and equivalent to 20 g/L was prepared by weighing 66.68 g of PAC salt into a beaker and dissolving it in deionized water to make a 200 g solution. Secondly, according to selected intrinsic viscosity, the solid content of PDMDAAC, and the selected mass ratio of composite coagulants, an appropriate amount of PDMDAAC was accurately weighed and dissolved in each previously prepared 10% PAC stock solution. The composite coagulants were homogenized by stirring and allowed to stand for 24 h.

Raw water quality parameters

The investigations were done with two batches of surface raw water collected from a water reservoir located along the Yangtze River in the autumn; the water quality parameters during the collection period are presented in Table 1. The first batch of raw water samples was used for selection of coagulation/application mode of SDST point while the second batch was used to verify the SDST point and removal efficiency for all coagulants.

Water quality parameter measurement

Water quality parameters such as turbidity, COD_{Mn}, NH_3-N, and zeta potential were measured for surface raw water and supernatant samples according to APHA standard methods for water and wastewater and Chinese national drinking water and wastewater standards (Clesceri et al., 1999). The method of Yinlong et al. (2006) was used for the evaluation of coagulation removal efficiency as an indicator of improved water quality parameters. The turbidity of each water sample was measured using a light-scattering method (formazin standard), the COD_{Mn} measurement was the acidic potassium permanganate titration method; the ammonia nitrogen measurement method was Nessler reagent spectrophotometry. The zeta potential measurement method was by microelectrophoresis while the morphology of settled flocs was assessed by direct photography and microphotography (Chakraborti et al., 2000).

Experimental procedures

Selection of coagulation stirring conditions and jar test procedures

The selection of the coagulation stirring condition for the jar tests aimed to mimic the actual coagulation conditions at the drinking water production plant in Nanjing at the time of this study, to meet the residual turbidity requirement of 1.0–1.5 NTU for the supernatant from the sedimentation tank . The stirring conditions were determined as follows: rapid stirring (300 r/min) for 15 s, before adding a certain dosage of coagulant, rapid stirring at 300 r/min for 1 min for dispersion, then stirring at medium speed of 100 r/min for 3 min, slow speed at 30 r/min for 3 min and, lastly, sedimentation for 30 min at 0 r/min.

Coagulant dosage–residual turbidity test

The dosage–residual turbidity test was performed as described in Adebayo et al. (2021). By adding 1 000 mL of raw water sample into each of 12 jar beakers placed into two jar machines for coagulation, a series of prepared and increasing amounts of each coagulant was added to each jar beaker, one by one, to complete the coagulation and sedimentation operation. Finally, each 100 mL supernatant sample was taken at about 2 cm below the liquid surface of each beaker after sedimentation. The residual turbidity of the resulting supernatants was measured, and the dosage–residual turbidity curves were drawn for all coagulants in order to determine the point of same dosage and supernatant turbidity (SDST) for all coagulants, which is also the point of intersection for using PAC and composite coagulants PAC/PDMDAAC. The coagulant dosages at this point were used for bulk coagulation operations in order to control supernatant turbidity between 1.0 and 1.5 NTU to mimic drinking water production plant condition during this study.

Removal efficiency for water quality parameters

The bulk coagulation operation using PAC and the 9 kinds of composite coagulants PAC/PDMDAAC was synchronously repeated under the coagulation/application mode of SDST point, which was also the interception point selected for using PAC and composite coagulants PAC/PDMDAAC to meet drinking water production plant condition and maximize removal efficiency of water quality parameters using jar testing. After the sedimentation step was completed, 100 mL of the resulting supernatant samples were used to measure the parameters of residual turbidity, COD_{Mn}, ammonia nitrogen (NH_3-N) and zeta potential. The removal efficiencies for water quality parameters were evaluated while

Table 1. The basic water quality conditions of the Yangtze River in autumn

Serial number	Turbidity (NTU)	COD (mg·L⁻¹)	NH₃-N (mg·L⁻¹)	Zeta potential (mV)	Water temperature (°C)
1	22.40–45.00	2.80–3.30	0.32–0.37	−24.20 to −22.34	19–24

the zeta potential values provided insight into the coagulation mechanism.

Floc size and morphology characterization at varying composite coagulant PAC/PDMDAAC dosages

Based on the dosage–residual turbidity experimental procedures, one group of results obtained using the composite coagulants PAC/PDMDAAC was selected for floc size and morphology characterization under varying dosages, in order to assess the effect of composite coagulant dosage on floc strength and compactness. Photographs were taken of suspended particles in the beakers during jar testing and the particle size estimated. Furthermore, after the flocculation and sedimentation steps were completed, some flocs were carefully withdrawn by a disposable plastic pipette from the bottom of each of the 12 beakers, and placed one by one on the micro-graduated glass slide under the microscope. The flocs were observed and photographed to complete the characterization of the morphology of the flocs, which was then used to analyse the influence of varying coagulant dosage on floc strength and ability to settle easily.

Floc size and morphology characterization at the same optimum dosage for all coagulants

Based on the dosage–residual turbidity test procedures, assessment of floc size and morphology characterization under the same optimum dosage of PAC and 9 kinds of composite coagulants PAC/PDMDAAC were done. The maximum optimum dosage at SDST point was selected as the common dosage for all coagulants and then, following the coagulation jar test operation, the 10 kinds of coagulants were added into the jar beakers to perform the coagulation operation. After flocculation and sedimentation were completed, a disposable plastic pipette was used to carefully withdrawn some flocs from the bottom of each jar beaker, and the flocs were placed on a micro-scaled glass slide under the microscope to complete their characterization. The morphological characterization of the flocs from using the coagulants were used to analyse the effect of coagulant performance parameters, such as intrinsic viscosity of PDMDAAC, and mass ratio of PAC to PDMDAAC, on floc strength, compactness, and ability to settle easily.

RESULTS AND DISCUSSION

Relationship of residual turbidity to coagulant dosage and removal mechanism

Using the coagulation jar test procedure, firstly, the dosage–residual turbidity test was undertaken using varying dosages of PAC and 9 kinds of composite coagulants of PAC/PDMDAAC to obtain the corresponding residual supernatant turbidity in the treatment of the first batch of Yangtze River lightly micro-polluted raw water in autumn; the results are shown in Fig. 1. It was observed that residual turbidity of the supernatant water generally decreases with increasing dosage when using PAC only and also the various composite coagulants. Moreover, the coagulation and turbidity removal efficiencies of the composite coagulants under the coagulation mode of same dosage (SD) as PAC coagulant or same supernatant turbidity (SST) usually required a dosage lower than that when using inorganic salt coagulant only, a result which has been previously reported by other researchers (Adebayo et al., 2021; Cui et al., 2020). The reason for this observation is related to the multiple strengthening effects of the cationic PDMDAAC coagulant in the composite coagulants, including charge neutralization, compression of the electric double layer, polymer bridging, and net capturing. For instance, in Table 2 and Fig. 1, when using composite coagulants with 1.60 dL/g, 5:1 and 2.60 dL/g, 5:1, with higher mass ratio at coagulation mode using the SDST point, it was observed that the turbidity removal effect was better than when using PAC only at residual turbidity near 1.50 NTU. But when the residual supernatant turbidity was lower than 1.50 NTU, these two kinds of composite coagulants had lost their superior turbidity removal performance as the coagulant dosage exceeded their optimum range. This effect may occur when the molecular weight of PDMDAAC is higher, i.e., with higher intrinsic viscosity, resulting in a poor dispersion effect; this appears after a certain dosage limit is exceeded. The excessive effects of partial charge neutralization make the suspended particles re-disperse after flocculation causing the phenomenon of particle re-stabilization leading to an increase in residual turbidity (Yang et al., 2012; Leia et al., 2012; Katrivesis et al., 2019; Adebayo et al., 2021).
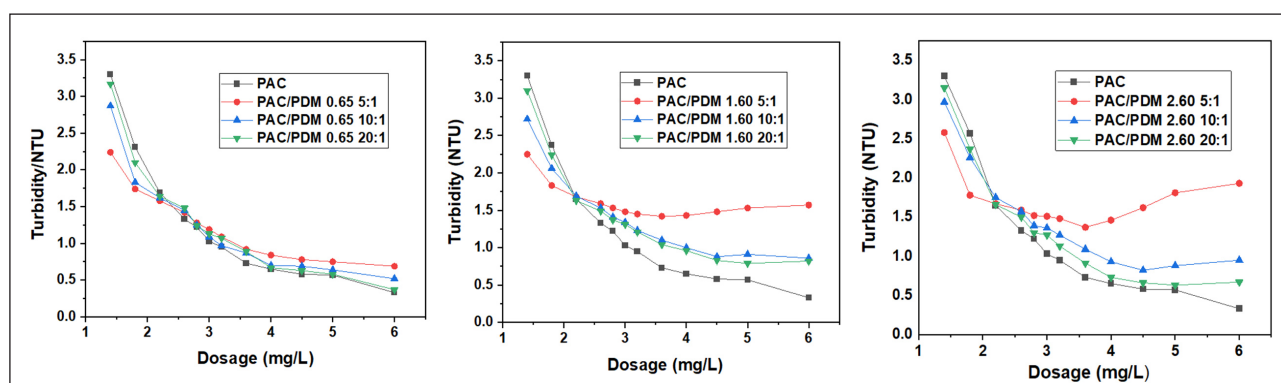

Figure 1. Dosage–residual turbidity curve for PAC and 9 kinds of composite coagulants

Table 2. Residual turbidity and dosage for supernatant at interception point for 9 kinds of composite and PAC (first batch)

Coagulant type	[η] / m(PAC): m(PDMDAAC)											
	PAC	PAC	PAC	0.65/ 5:1	0.65/ 10:1	0.65/ 20:1	1.60/ 5:1	1.60/ 10:1	1.60/ 20:1	2.60/ 5:1	2.60/ 10:1	2.60/ 20:1
Dosing amount (mg·L^{-1})	2.6	2.8	3.0	3.0	2.8	2.8	2.6	2.8	2.8	2.6	2.8	3.0
Intersection turbidity (NTU)	1.45	1.30	1.00	1.03	1.30	1.30	1.48	1.29	1.26	1.50	1.35	1.02

In addition, the various composite coagulants that can meet the requirement of the residual turbidity of the supernatant water below 1.5 NTU at coagulation mode of SDST are, namely, composite PAC/PDMDAAC at 0.65 dL/g, 5:1, 20:1); 1.60 dL/g, 10:1, 20:1; and 2.60 dL/g, 10:1, 20:1. Only these 6 kinds of composite coagulants can still maintain better coagulation and turbidity removal efficiency than using PAC for the treatment of lightly micro-polluted surface raw water of the Yangtze River when it is required to control the residual turbidity of the supernatant water below 1.5 NTU. The reason for this phenomenon may be that composite coagulants with lower and medium molecular weight (MW), or intrinsic viscosity of PDMDAAC having lower or medium mass ratio of PAC to PDMDAAC, are linear molecules and used within the dosage range within which they can be easily dispersed. The residual turbidity of the supernatant water can be acceptable when composite coagulants are well dispersed and diluted. However, the inability of composite coagulant with 2.60 dL/g, 5:1 to meet the controlled turbidity requirement may possibly be due to its poor dispersion effect (Shen et al., 2010b). It appears the composite coagulant can achieve a residual turbidity lower than 1.5 NTU at a lower dosage of the higher intrinsic viscosity of PDMDAAC and higher mass ratio of PAC to PDMDAAC (lower PDMDAAC content) when it can be effectively dispersed during the coagulation process (Adebayo et al., 2021). In summary, it was observed that the residual turbidity reached by using composite coagulants was lower than when using PAC only before the SDST point. Beyond this point, as the dosage exceeds the optimum limit, the superior turbidity removal effect disappears. Two kinds of composite coagulants with higher intrinsic viscosity and lower mass ratio did not meet requirement to control supernatant turbidity between 1.0 and 1.50 NTU, while the other 7 kinds of composite coagulants achieved this condition.

Water quality parameter removal rate at same dosage and supernatant turbidity (SDST)

The repeat coagulation operations were done using the SDST point which was also the intersection point for using PAC and 9 kinds of composite PAC/PDMDAAC coagulants with the second batch of Yangtze River lightly micro-polluted raw water for removal of water quality parameters. The water quality parameter removal rates for supernatant turbidity, COD_{Mn}, ammonia-nitrogen (NH_3N), and zeta potential, respectively, are shown in Figs 2–5.

Residual turbidity-removal rate at SDST point

Figure 2 shows the water quality parameters from the coagulation experimental procedure. It can be seen from Fig. 2, which represents the residual turbidity-removal rate at SDST point at controlled residual turbidity of 1.0– 1.5 NTU, that when the mass ratio of PAC to PDMDAAC was 5:1, with the increasing intrinsic viscosity of PDMDAAC, the turbidity removal rate using the composite coagulants at the SDST point was lower. For example, when treating lightly micro-polluted Yangtze River raw water using composite coagulant with 1.60 dL/g (5:1) and 2.60 dL/g (5:1), the lowest point of residual turbidity was only 1.63 NTU and 1.65 NTU with removal rates of 95.43% and 95.38%, respectively. In contrast, the composite coagulant at 0.65 dL/g (5:1) has the lowest residual turbidity of 1.03 NTU, achieving 97.11% removal, which was the best performance among the composite coagulants with mass ratio of 5:1. However, when the mass ratio was 10:1 with moderate content of PDMDAAC, with increasing intrinsic viscosity of PDMDAAC in the composite coagulant, the turbidity removal rates at the SDST point were relatively uniform, usually between 1.0 and 1.2 NTU, which meets the requirements for the supernatant water residual turbidity to be below 1.5 NTU. It appears that composite coagulants with mass ratio of 10:1 are better dispersed and effectively diluted. When the mass ratio was 20:1 with the lowest PDMDAAC content, with increasing intrinsic viscosity, the turbidity removal rates for the composite coagulant were slightly improved. But when choosing this composite coagulant, it should be noted that the coagulant dosage may increase (see Table 3). These observations were in agreement with those reported for other studies (Zhang et al., 2009; Shen et al., 2017; Adebayo et al., 2021). It can be inferred that composite coagulants with lower intrinsic viscosity and higher mass ratio or higher intrinsic viscosity and lower mass ratio were effective in meeting the requirement of controlling supernatant turbidity below 1.5 NTU, with maximum turbidity removal rate due to effective dispersion and dilution within the coagulant dosage used.

Residual COD_{Mn} removal rate at SDST point

Figure 3 presents the results of the coagulation experimental procedure. This represents the residual COD_{Mn} removal rate at SDST points and optimum coagulant dosage of 2.0, 2.4, and 2.80 mg/L, respectively. When using composite coagulants with 0.65 dL/g (5:1, 10:1, 20:1); 1.6 dL/g (10:1, 20:1); 2.60 dL/g (10:1),

Table 3. Residual turbidity and dosage for supernatant at interception point for 9 kinds of composite and PAC (second batch)

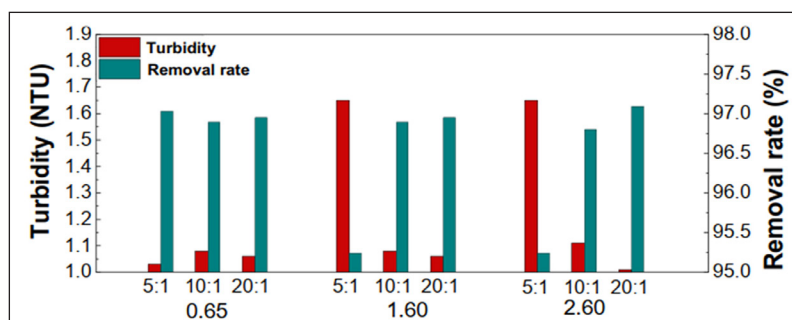| Coagulant type	[η] / m(PAC): m(PDMDAAC)											
	PAC	PAC	PAC	0.65			1.60			2.60		
				5:1	10:1	20:1	5:1	10:1	20:1	5:1	10:1	20:1
Dosage (mg·L⁻¹)	2.8	2.4	2.0	2.4	2.8	2.8	2.0	2.8	2.8	2.0	2.8	2.4
Supernatant turbidity (NTU)	1.08	1.05	1.63	1.03	1.08	1.06	1.65	1.08	1.06	1.65	1.11	1.01

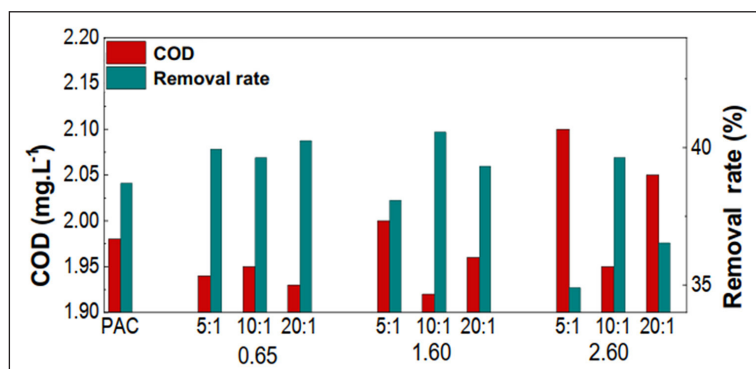**Figure 2.** Residual turbidity-removal rate results at SDT points

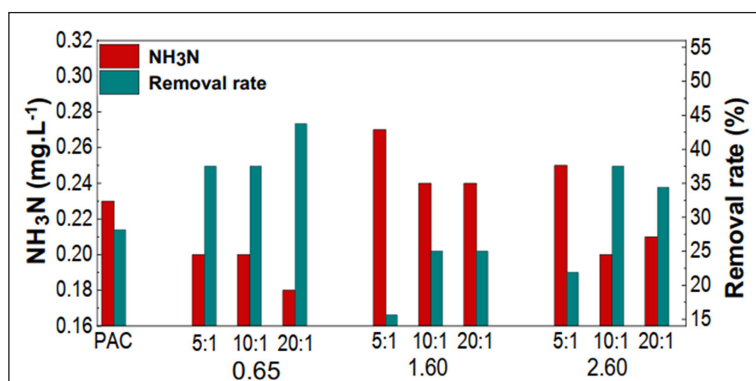**Figure 3**. COD removal rate at same dosage and turbidity points

Figure 4. NH₃-N removal rate at same dosage and turbidity points
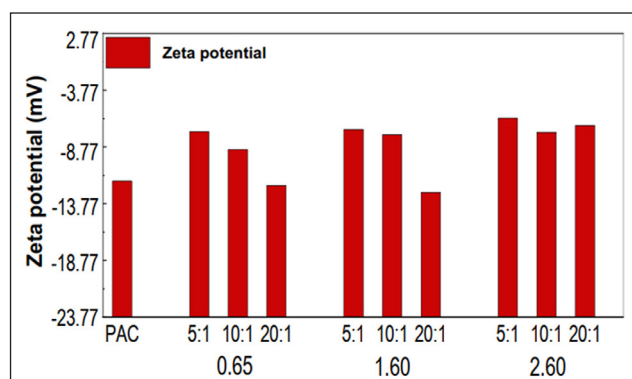

Figure 5. Zeta potential at same dosage and turbidity points

the resulting supernatants obtained have COD_{Mn} removal rates higher than when using PAC only at the maximum optimum dosage of 2.80 mg/L. The COD_{Mn} removal efficiency improved by 0–6.19% using these composite coagulants compared with using PAC only. It appears that when the residual turbidities of the supernatant waters are similar, the removal rate of these composite coagulants for COD_{Mn} are also higher than when using PAC only. A possible reason for this is that the mechanism of turbidity and COD_{Mn} removal may be related, resulting from effective charge neutralization and adsorption-bridging of medium and larger organic molecules on suspended particles, the metal hydroxide, and the polymer, leading to simultaneous and enhanced removal (Moussas and Zouboulis, 2009; Saxena et al., 2020).

Residual NH₃ N removal rate at SDST point

Figures 2 and 4 show the results of the coagulation experimental procedure, and the residual NH₃-N and turbidity removal rates at SDST using PAC only and composite coagulants PAC/

PDMDAAC at optimum coagulant dosages of 2.0, 2.40, and 2.80 mg/L. However, only composite coagulants PAC/PDMDAAC with 0.65 dL/g (5:1, 10:1, 20:1), and 2.60 dL/g (10:1, 20:1) had NH₃-N removal rates higher than those achieved using PAC only at optimum dosage of 2.80 mg/L. The NH₃-N removal rate increase was in the range of 0–15.62%. This observation may be related to the ability of the composite PAC/PDMDAAC, with effective dispersion and dilution within the dosage used, to bridge smaller polar molecules between metal hydroxide and the polymer, thereby enhancing their removal compared to the charge neutralization mechanism which occurs when using PAC only (Lee and Westerhoff, 2006; Adebayo et al., 2021).

Zeta potential values at SDST point

Figures 2 and 5 show the results for residual turbidity removal and zeta potential at the SDST point for PAC and 9 kinds of composite coagulants using the coagulation experimental procedure. The zeta potential values were used to obtain an understanding of

the effectiveness of charge neutralization after coagulation. It was observed that composite coagulants with higher intrinsic viscosity of PDMDAAC and higher mass ratio of PAC to PDMDAAC have a higher charge neutralization effect based on zeta potential values. The resulting supernatant water samples obtained using composite coagulants with 0.65 dL/g (5:1, 10:1); 1.60 dL/g (5:1, 10:1); 2.60 dL/g (5:1, 10:1, 20:1) were observed to have zeta potential values with fewer negative values in the range of −5 mV to −7.84 mV, compared to −11.55 mV when using PAC. The zeta potential values of the resulting supernatants give an indication of coagulants having positive charges which enable them to remove negatively charged suspended particles in raw surface water, predominantly by charge neutralization, depending on the coagulant dosage used. Generally, the negative potential of the residual particles in the supernatant water after using the composite coagulant is lower than when using PAC only without regard to supernatant turbidity, suggesting enhanced charge neutralization within the dosage range used (López-Maldonado et al., 2014; Olukowi et al., 2022). The destabilization of colloid particles during coagulation, and their ability to agglomerate and form flocs, can be monitored by zeta potential values – particles with a less negative value after treatment have a smaller repulsive force and can easily settle (Wei et al., 2009; Adebayo et al., 2021).

Floc size and morphology

Particle size and morphology of flocs at varying dosages of the same composite coagulant

Figure 6 shows the suspended flocs in the slow stirring stage of coagulation for all 12 beakers having varying dosages of a composite PAC/PDMDAAC coagulant in the range of 1.6–3.8 mg/L (average dosage interval of 0.2 mg/L). Figures 1 to 7 show photomicrographs of the composite coagulant (1.60 dL/g, 10:1) at dosage range of 1.6–3.8 mg/L (average dosage interval of 0.2 mg /L) for the settled flocs. It can be seen from Figs 6–7 that, as the dosage increased, using the same composite coagulant, the average particle size of the suspended flocs gradually increased. Also, the sizes of the settled flocs increased as coagulant dosage increased; however, it was observed that the compactness of the flocs reached a maximum at the interception dosage point and no significant increase was seen thereafter. For the suspended flocs in the slow stirring stage in Fig. 6, the average particle size changes
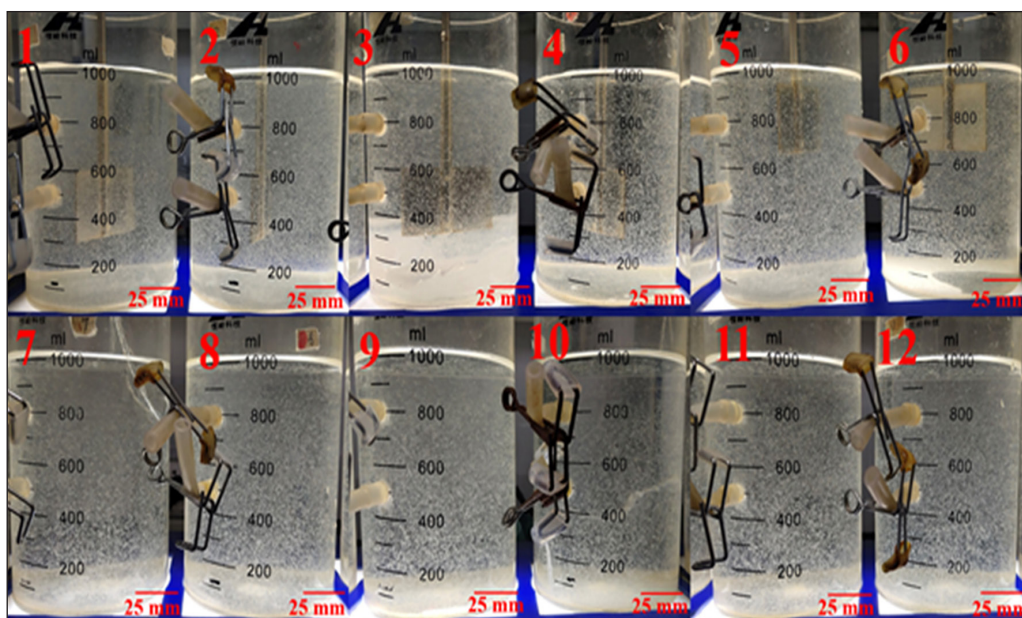

Figure 6. Photos of suspended flocs in the slow stirring stage of coagulation with different dosages of the same composite PAC/PDMDAAC (1.60 dL/g, 10:1)
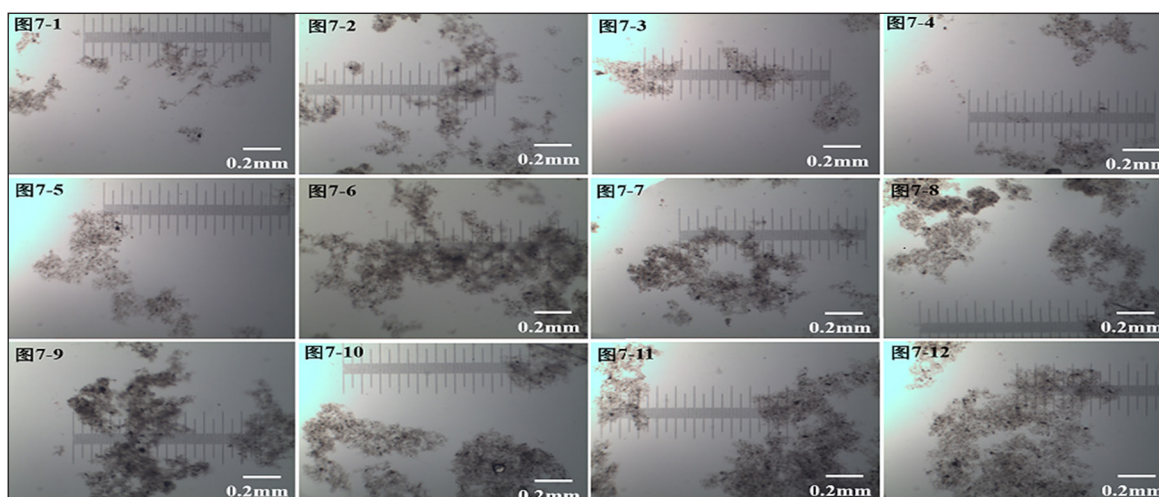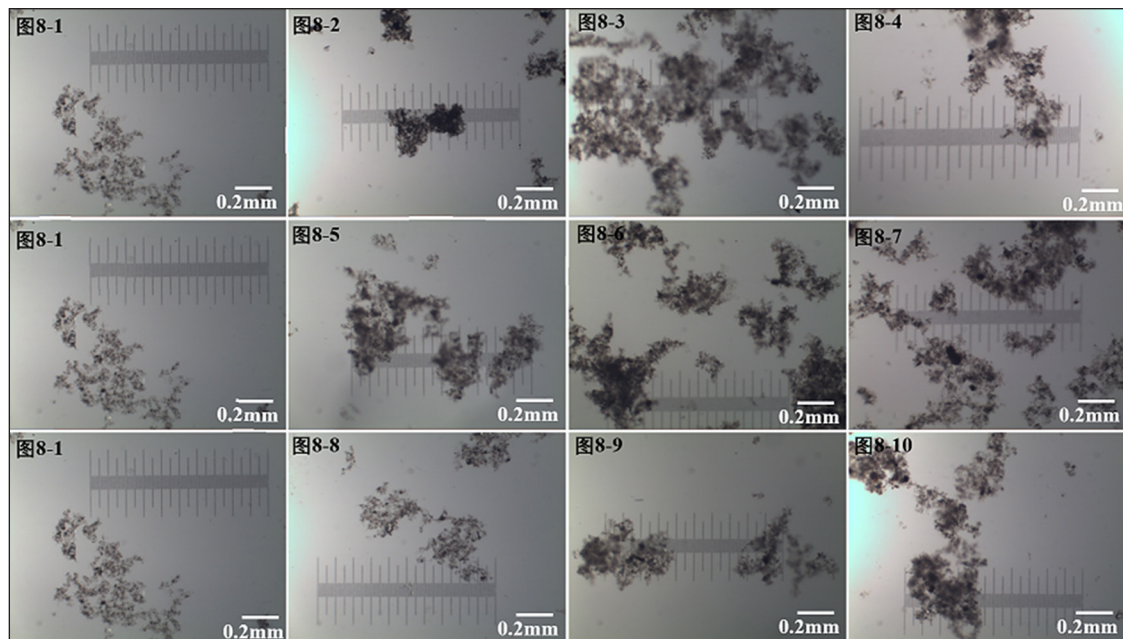

Figure 7. Photomicrographs of the settled flocs for PAC/PDMDAAC coagulant at different dosages

Table 4. Particle size of suspended flocs at different dosages

Dosage (mg·L⁻¹)	1.6	1.8	2.0	2.2	2.4	2.6	2.8	3.0	3.2	3.4	3.6	3.8
Floc size (mm)	0.85	1.10	1.35	1.55	1.60	1.85	2.05	2.05	2.00	2.00	2.00	2.00

Table 5. Analysis of floc density at the same dosage of 9 coagulants and PAC

Coagulant type	PAC	(0.65 dL·g⁻¹)			m(PAC): m(PDMDAAC) (1.60 dL·g⁻¹)			(2.60 dL·g⁻¹)		
		5:1	10:1	20:1	5:1	10:1	20:1	5:1	10:1	20:1
Dosage (mg·L⁻¹)	2.8	2.8	2.8	2.8	2.8	2.8	2.8	2.8	2.8	2.8
Density (observation)	Loose	High density	High density	Dense	High density	High density	High density	Loose	Dense	High density

Figure 8. Micrographs of the settled flocs with the same dosage of different PAC/PDMDAAC coagulants

were measured, and the results are shown in Table 4. It was observed that the average particle size of the flocs in the suspension varies. This trend is basically consistent with the variation of the residual turbidity of the corresponding composite coagulants with dosage seen in Fig. 1. In addition, as for the particle size of suspended particles, the resulting floc size reaches a maximum at the interception dosage point, and remains basically unchanged or slightly reduced thereafter. This is consistent with the trend of change in residual turbidity of the corresponding composite coagulant with dosage seen in Fig. 1.

Morphology characterization of flocs with different coagulants at same dosage

According to the coagulation experimental procedure and the measurement results of the same turbidity and other dosages, the maximum optimum dosage of PAC and composite coagulants under the condition of using the second batch of Yangtze River raw water at 2.80 mg/L in the jar test experiments was obtained. The flocs were sampled and photographed after sedimentation, and the results are shown in Fig. 8, while Table 5 presents the settled floc density at same dosage for all coagulants. The size and compactness of the flocs formed by the composite coagulants with different intrinsic viscosities are different, even at the same coagulant dosage. The outstanding bridging and net-capturing ability of the composite coagulants compared with PAC only was

fully demonstrated by the compactness of the flocs, which was in agreement with literature (Wang et al., 2017). The coagulants with high density have strong bridging and net capture ability. It was observed that composite coagulants with different intrinsic viscosities of PDMDAAC and mass ratios of PAC to PDMDAAC produced high density flocs when the intrinsic viscosity of PDMDAAC was high (i.e. high molecular weight) and the mass ratio of PAC:PDMDAAC was low (PDMDAAC content is higher) or when the intrinsic viscosity of PDMDAAC was low and the mass ratio of PAC:PDMDAAC was high (PDMDAAC content is low). This was in agreement with other published studies which reported that the presence of PDMDAAC in the composite coagulant resulted in more compact and denser flocs (Shen et al., 2017). The floc morphologies give insight into the polymer bridging and net capture ability of composite coagulants of inorganic salt/PDMDAAC and explain their enhanced coagulation efficiency compared to using the corresponding inorganic coagulant only, at reduced dosage.

Enhanced coagulation shown by zeta potential and floc characterization

As mentioned earlier, the mechanism of turbidity removal at varying dosages of composite coagulants were by charge neutralization, polymer-bridging, and net catching, depending on coagulant composition as well as controlled residual supernatant

turbidity during the coagulation process. For example, the composite coagulant contains Al^{3+} ions and highly charged cationic PDMDAAC, rich in positive surface charges for effective charge neutralization and destabilization of particles of different sizes in raw surface water which are usually negatively charged; however, the zeta potential measurements and the floc characterization can also provide supporting evidence with insight into the mechanism of coagulation. The zeta potential values of supernatants obtained using composite coagulants are usually less negative compared to those obtained using PAC only, hence it can be inferred that the dominant coagulation mechanism is charge neutralization, within the coagulant dosage used. However, there appears to be a lack of strong correlation between the zeta potential values and water quality parameter removal rates, suggesting that there are other coagulation mechanisms and factors that affect coagulation efficiency. It was observed that composite coagulants with less negative zeta potential values did not always lead to improved water qualities. In addition, floc characterization can give insights about agglomeration of particles to form flocs of varying size, strength, compactness, and ease of settling when using composite coagulants. The floc size and image analysis showed that the presence of PDMDAAC, depending on the intrinsic viscosity of PDMDAAC and mass ratio PAC to PDMDDAC, can further enhance coagulation removal efficiency through adsorption-bridging and net catching mechanism, via surface charges and functional group interactions between the polymer and particles with vacant sites (Bolto and Gregory, 2007; Adebayo et al., 2021)). In summary, the zeta potential measurements provide supporting evidence of the charge neutralization mechanism while the floc size and image analysis support the adsorption-bridging mechanism, which helps to explain the enhanced coagulation achieved by the composite coagulant within the optimum dosage range.

CONCLUSIONS

For lightly micro-polluted raw water of Yangtze River in the autumn (Nanjing section), with turbidity in the range of 25–45 NTU and temperature in the range of 19–24°C, when the residual turbidity of the supernatant is controlled in the range of 1.0–1.5 NTU for the 9 kinds of composite coagulants of PAC/PDMDAAC with intrinsic viscosity $[\eta]$ = 0.65–2.60 dL/g; and mass ratio PAC:PDMDAAC = 5:1, 10:1, 20:1, the composite coagulants still maintain the advantage of enhanced coagulation efficiency before and on the intersection point of SDST as using PAC coagulant, and at the same time meet the requirement of keeping the supernatant turbidity in the range of 1.0–1.2 NTU, except for PAC/PDMDAAC 1.60 (5:1) and PAC/PDMDAAC 2.60 (5:1).

The removal efficiency for water quality parameters via the coagulation/application mode of SDST as using PAC and the composite coagulants showed that the average maximum turbidity removal rate for two kinds of composite coagulants that did not meet the controlled turbidity requirement of 1.0–1.5 NTU was 95.48%, while for 7 kinds of composite coagulant that met the requirement, the average maximum turbidity removal rate was 97.6%. In addition, the COD_{Mn} and NH_3-N removal rate increases were 0–6.19% and 0–15.62%, respectively, compared to using PAC only at same maximum optimum dosage of 2.80 mg/L.

The composite coagulants that met the requirement of supernatant residual turbidity of less than 1.5 NTU after coagulation at SDST point using lightly micro-polluted raw water of Yangtze River in autumn, and still maintained the advantages of removing other water quality parameters are as follows: composite coagulants with 0.65 dL/g (5:1,10:1,20:1); 1.60 dL/g (10:1, 20:1); and 2.60 dL/g (10:1–20:1).

In view of the influence of the intrinsic viscosity of PDMDAAC and amount (or mass ratio) of PDMDAAC in the composite coagulants applied on the resulting water quality parameters, including the residual turbidity at the SDST point, the composite coagulants, with both high and medium intrinsic viscosity values having low and medium amount of PDMDAAC, and with low intrinsic viscosity values with high, medium, and low amounts of PDMDAAC, exhibit the advantages of improved supernatant water qualities when treating lightly micro-polluted raw water of Yangtze River in autumn. Furthermore, the zeta potential values of the resulting supernatant water and the floc characterization give supporting evidence for the enhancement effects of charge neutralization, bridging and net capture on the performance of PDMDAAC, as well as its dispersion in water. These coagulation mechanisms provide an explanation for the coagulation performance outlined above.

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REFERENCES

ADEBAYO IO, OLUKOWI OO, ZHIYUAN Z and ZHANG Y (2021) Comparisons of coagulation efficiency of conventional aluminium sulfate and enhanced composite aluminium sulfate/polydimethyldiallylammonium chloride coagulants coupled with rapid sand filtration. *J. Water Process Eng.* **44** (June) 102322. https://doi.org/10.1016/j.jwpe.2021.102322

ALZAHRANI F, COLLINS AR and ERFANIAN E (2020) Drinking water quality impacts on health care expenditures in the United States. *Water Resour. Econ.* **32** (June) 100162. https://doi.org/10.1016/j.wre.2020.100162

BOLTO B, ABBT-BRAUN G, DIXON D, ELDRIDGE R, FRIMMEL F, HESSE S, KING S and TOIFL M (1999) Experimental evaluation of cationic polyelectrolytes for removing natural organic matter from water. *Water Sci. Technol.* **40** (9) 71–79. https://doi.org/10.1016/S0273-1223(99)00642-3

BOLTO B, DIXON D, ELDRIDGE R and KING S (2001) Cationic polymer and clay or metal oxide combinations for natural organic matter removal. *Water Res.* **35** (11) 2669–2676. https://doi.org/10.1016/S0043-1354(00)00552-2

BOLTO B and GREGORY J (2007) Organic polyelectrolytes in water treatment. *Water Res.* **41** (11) 2301–2324. https://doi.org/10.1016/j.watres.2007.03.012

BRANDT MJ, JOHNSON KM, ELPHINSTON AJ and RATNAYAKA DD (2017) *Storage, Clarification and Chemical Treatment.* https://doi.org/10.1016/b978-0-08-100025-0.00008-9

BRATBY J (2016) *Coagulation and Flocculation in Water and Wastewater Treatment* (3rd edn). IWA, London. https://doi.org/10.2166/9781780407500

CHAKRABORTI RK, ATKINSON JF and VAN BENSCHOTEN JE (2000) Characterization of alum floc by image analysis. *Environ. Sci. Technol.* **34** (18) 3969–3976. https://doi.org/10.1021/es990818o

CLESCERI LS, GREENBERG AE and EATON AD (1999) *Standard Methods for the Examination of Water and Wastewater.* (20th edn). APHA, Washington DC. 1027–1033.

CUI H, HUANG X, YU Z, CHEN P and CAO X (2020) Application progress of enhanced coagulation in water treatment. *RSC Adv.* **10** (34) 20231–20244. https://doi.org/10.1039/d0ra02979c

HAARHOFF BJ and CLEASBY JL (1989) Direct filtration of *Chlorella* with cationic polymer. *J. Environ. Eng.* **115** (2) 348–366. https://doi.org/10.1061/(ASCE)0733-9372(1989)115:2(348)

KATRIVESIS FK, KARELA AD, PAPADAKIS VG and PARASKEVA CA (2019) Revisiting of coagulation-flocculation processes in the production of potable water. *J. Water Process Eng.* **27** (September 2018) 193–204. https://doi.org/10.1016/j.jwpe.2018.12.007

LEE W and WESTERHOFF P (2006) Dissolved organic nitrogen removal during water treatment by aluminum sulfate and cationic polymer coagulation. *Water Res.* **40** (20) 3767–3774. https://doi.org/10.1016/j.watres.2006.08.008

LEIA CN, LOU IC, SONG HU and SUN P (2012) Turbidity removal improvement for Yangtze River raw water. *Desalin. Water Treat.* **45** (1–3) 215–221. https://doi.org/10.1080/19443994.2012.692060

LI X, ZHANG Y, ZHAO X, GAO N and FU T (2015) The characteristics of sludge from enhanced coagulation processes using PAC/PDMDAAC composite coagulants in treatment of tiny-polluted raw water. https://doi.org/10.1016/j.seppur.2015.04.015

LIU Z, WEI H, LI A and YANG H (2019) Enhanced coagulation of low-turbidity tiny-polluted surface water: Properties and optimization. *J. Environ. Manage.* **233** (March 2018) 739–747. https://doi.org/10.1016/j.jenvman.2018.08.101

LÓPEZ-MALDONADO EA, OROPEZA-GUZMÁN MT and OCHOA-TERÁN A (2014) Improving the efficiency of a coagulation-flocculation wastewater treatment of the semiconductor industry through zeta potential measurements. *J. Chem.* **2014** 6–9. https://doi.org/10.1155/2014/969720

MOUSSAS PA and ZOUBOULIS AI (2009) A new inorganic-organic composite coagulant, consisting of polyferric sulphate (PFS) and polyacrylamide (PAA). *Water Res.* **43** (14) 3511–3524. https://doi.org/10.1016/j.watres.2009.05.015

NASCIMENTO COC, VEIT MT, PALÁCIO SM, GONÇALVES GC and FAGUNDES-KLEN MR (2019) Combined application of coagulation/flocculation/sedimentation and membrane separation for the treatment of laundry wastewater. *Int. J. Chem. Eng.* **2019**. https://doi.org/10.1155/2019/8324710

NOWACKA A, WŁODARCZYK-MAKUŁA M and MACHERZYŃSKI B (2014) Comparison of effectiveness of coagulation with aluminum sulfate and pre-hydrolyzed aluminum coagulants. *Desalin. Water Treat.* **52** (19–21) 3843–3851. https://doi.org/10.1080/19443994.2014.888129

OLUKOWI OM, XIE Y, ZHOU Z, ADEBAYO IO and ZHANG Y (2022) Performance improvement and mechanism of composite PAC/PDMDAAC coagulant via enhanced coagulation coupled with rapid sand filtration in the treatment of tiny-polluted surface water. *J. Environ. Chem. Eng.* **10** (5). https://doi.org/10.1016/j.jece.2022.108450

SAXENA K, BRIGHU U and CHOUDHARY A (2020) Pilot-scale coagulation of organic and inorganic impurities: Mechanisms, role of particle concentration and scale effects. *J. Environ. Chem. Eng.* **8** (4) 103990. https://doi.org/10.1016/j.jece.2020.103990

SAYATO Y (1989) WHO Guidelines for Drinking-Water Quality. *Eisei Kagaku* **35** (5) 307–312. https://doi.org/10.1248/jhs1956.35.307

SHEN L, CHENG L, WANG H, LIAN X and QI J (2010a) Pilot-scale study of efficient coagulation sedimentation of tiny-polluted water. *ICBEE 2010 – 2010 2nd International Conference on Chemical, Biological and Environmental Engineering, Proceedings.* 124–127. https://doi.org/10.1109/ICBEE.2010.5653560

SHEN L, CHENG L, WANG H, LIAN X and QI J (2010b) Pilot-scale study of efficient coagulation sedimentation of tiny-polluted water. *ICBEE 2010 – 2010 2nd International Conference on Chemical, Biological and Environmental Engineering, Proceedings* (ICBEE) 124–127. https://doi.org/10.1109/ICBEE.2010.5653560

SHEN X, GAO B, HUANG X, BU F, YUE Q, LI R and JIN B (2017) Effect of the dosage ratio and the viscosity of PAC/PDMDAAC on coagulation performance and membrane fouling in a hybrid coagulation-ultrafiltration process. *Chemosphere* **173** 288–298. https://doi.org/10.1016/j.chemosphere.2017.01.074

SILLANPÄÄ M, NCIBI MC, MATILAINEN A and VEPSÄLÄINEN M (2018) Removal of natural organic matter in drinking water treatment by coagulation: A comprehensive review. *Chemosphere* **190** 54–71. https://doi.org/10.1016/j.chemosphere.2017.09.113

SUN Y, ZHOU S, CHIANG P-C and SHAH KJ (2019) Evaluation and optimization of enhanced coagulation process: Water and energy nexus. *Water-Energy Nexus* **2** (1) 25–36. https://doi.org/10.1016/j.wen.2020.01.001

TOLKOU AK and ZOUBOULIS AI (2020) Application of composite pre-polymerized coagulants for the treatment of high-strength industrial wastewaters. *Water* **12** (5). https://doi.org/10.3390/W12051258

WANG B, SHUI Y, HE M and LIU P (2017) Comparison of flocs characteristics using before and after composite coagulants under different coagulation mechanisms. *Biochem. Eng. J.* **121** 107–117. https://doi.org/10.1016/j.bej.2017.01.020

WANG D, GREGORY J and TANG H (2008) Mechanistic difference of coagulation of kaolin between PACl and cationic polyelectrolytes: A comparative study on zone 2 coagulation. *Drying Technol.* **26** (8) 1060–1067. https://doi.org/10.1080/07373930802179327

WANG S, LI E, LI J, DU Z and CHENG F (2020) Preparation and coagulation-flocculation performance of covalently bound organic hybrid coagulant with excellent stability. *Colloids Surf. A: Physicochem. Eng. Aspects* **600** (May) 124966. https://doi.org/10.1016/j.colsurfa.2020.124966

WEI JC, GAO BY, YUE QY, WANG Y and LU L (2009) Performance and mechanism of polyferric-quaternary ammonium salt composite flocculants in treating high organic matter and high alkalinity surface water. *J. Hazardous Mater.* **165** 789–795. https://doi.org/10.1016/j.jhazmat.2008.10.069

WHO (2008) *Guidelines for Drinking-water Quality. Volume 1* (3rd edn). WHO Library, Geneva.

XI YILUN (2019) 硕士学位论文 强化混凝耦合过滤. Nanjiang University of Science and Technology.

YANG Z, ZHAO L, HU M, CAI D, TIAN Z, BAEYENS J, DEWIL R, QIN P, YANG W and GRAHAM NJD (2021) Hydrophobic-modified metal-hydroxide nanoflocculants enable one-step removal of multi-contaminants for drinking water production. *iScience* **24** (5) 102491. https://doi.org/10.1016/J.ISCI.2021.102491

YANG Z, GAO B, WANG Y, ZHANG X and YUE Q (2012) The effect of additional poly-diallyl dimethyl ammonium-chloride on the speciation distribution of residual aluminum (Al) in a low DOC and high alkalinity reservoir water treatment. *Chem. Eng. J.* **197** 56–66. https://doi.org/10.1016/j.cej.2012.05.002

YANG Z, GAO B, WANG Y, ZHANG X and YUE Q (2013) Relationship between residual Al species, floc operational parameters and coagulation performance during reservoir water treatment by PAC-PDMDAAC. *Sep. Purif. Technol.* **102** 147–156. https://doi.org/10.1016/j.seppur.2012.09.032

YINLONG J, XUELI E, CHANGJIE C, XIPING C, ZHANG L, YAYAN C, ZUGEN C, RIHUA G, TUHANG S, CHANGYI G and co-authors (2006) National Standard of the People's Republic of China Standards for Drinking Water Quality (2nd edn). Ministry of Health of China, Standardization of China. 18 pp.

YU W and GRAHAM NJD (2015) Performance of an integrated granular media - Ultrafiltration membrane process for drinking water treatment. *J. Membr. Sci.* **492** 164–172. https://doi.org/10.1016/j.memsci.2015.05.032

ZHANG YJ, LI XX, ZHAO XL, SUN B and YIN X (2009) Coagulation aid effect of PDM in treatment of winter Yangtze River water combined with PFS (in Chinese). *Ind. Water Wastewater* **40** (2) 19–22.

ZHANG YJ, ZHAO XL, LI XX, LIU C and ZHU LL (2010) Performance of PAC/PDM composite coagulants for removal of algae from Lake Taihu waters in summer. *Water Sci. Technol.* **62** (2) 330–339. https://doi.org/10.2166/wst.2010.292

YINLONG J, XUELI E, CHANGJIE C, XIPING C, ZHANG L, YAYAN C, ZUGEN C, RIHUA G, TUHANG S, CHANGYI G and co-authors (2006) National Standard of the People's Republic of China Standards for Drinking Water Quality (2nd edn). Ministry of Health of China, Standardization of China. 18 pp.