RESEARCH ARTICLE

Experimental study on enhanced coagulation using a combined composite bioflocculant and inorganic flocculant

Yawei Zhu[1, 2], Heng Zhang[1, 2, *], Lixin Li[3], Yisu Gao[4], Ting Guo[5], Xiangqian Chen[1]

[1]School of Civil Engineering, [2]Nanyang Key Laboratory of Water Pollution Control and Solid Waste Resource, Nanyang Institute of Technology, Nanyang, Henan, China. [3]School of Environment and Chemical Engineering, Heilongjiang University of Science and Technology, Harbin, Heilongjiang, China. [4]Nanyang River and Lake Affairs Center, Nanyang, Henan, China. [5]Zhongmao Environmental Protection Technology Co., Ltd. Nanyang, Henan, China.

Received: February 11, 2026; **accepted:** June 20, 2026.

Previous research has primarily focused on the use of composite bioflocculants to remove nitrogen and phosphorus from wastewater, whereas studies regarding the removal of turbidity, color, trace organic compounds, and residual Al^{3+} and Fe^{3+} resulting from the use of inorganic coagulants in drinking water remain relatively limited. This study investigated the combined treatment of drinking water using a composite biological flocculant (CBF) with polyaluminum ferric chloride (PAFC) and a composite aluminum-iron coagulant *via* the settling method. The supernatant was analyzed to evaluate the removal effects on turbidity, color, TOC, UV_{254}, and aluminum and iron ion residuals. The floc structure before and after CBF addition was also observed. The results showed that the optimal ratio for CBF combined with PAFC was 1 mg/L:20 mg/L, demonstrating significant removal effects on turbidity, Fe^{3+}, Al^{3+}, TOC, UV_{254}, and color with purification efficiency superior to individual addition. For CBF combined with the composite aluminum-iron coagulant, the optimal ratio was 2 mg/L:20mg/L. Except for the less satisfactory removal of UV_{254}, this combination also exhibited excellent removal effects on other indicators. The addition of CBF improved floc structure, thereby enhancing sedimentation efficiency. The combination of CBF with inorganic coagulants could efficiently remove multiple pollutants, improve sludge settling performance, and reduce chemical consumption. These findings provided a theoretical basis for ensuring drinking water quality safety and facilitating the industrial application of this technology.

Keywords: composite bioflocculant; polymeric aluminum ferric chloride; composite aluminum−ferric; water pollution; environmental protection.

[*]**Corresponding author:** Heng Zhang, School of Civil Engineering, Nanyang Institute of Technology, Nanyang, Henan 473004, China. Email: 3051075@nyist.edu.cn.

Introduction

Water is the source of life for humans and the foundation for the occurrence, development, and propagation of all life on Earth [1]. With the advancement of humanity and the development of industry and agriculture, serious water environmental pollution has become increasingly prominent, posing significant challenges to national economy and people's livelihood and has emerged as a major factor constraining sustainable socio-economic development [2]. Exploring efficient and safe water pollution treatment technologies holds important practical significance.

Coagulation treatment can effectively address water pollution issues [3], and composite bio-flocculants as a new type of safe, efficient, and environmentally friendly flocculant product represent an important means for water environmental pollution control [4]. As early as the 1950s, Japanese scholars discovered bacterial cultures capable of producing flocculation effects [5]. However, it was not until 1976 that Nakamura *et al*. truly ignited the research surge on bioflocculants, achieving several landmark research results [6]. Zhu *et al*. proposed the concept of "Compound Bioflocculants (CBF)" and successfully developed the composite bioflocculant HIT-M02 [7]. This flocculant demonstrated excellent decolorization [8], turbidity removal, and organic matter removal capabilities across different water bodies, water qualities, and low-temperature, low-turbidity water with good application effects [9]. Aluminum salts are the most widely applied inorganic salt coagulants in modern water supply and wastewater treatment coagulation technology [10]. High-molecular aluminum salt coagulants exhibit superior coagulation performance compared to traditional aluminum salts [11], which feature abundant raw material sources, simple processes, and diverse production methods [12], achieving good results in both drinking water treatment and wastewater treatment [13]. Previous studies have shown that neither single chemical flocculants nor bioflocculants alone can fully meet the demands of current drinking water treatment and water pollution control engineering technology development. However, if the advantages of both can be combined, specifically, by fully leveraging the environmental safety of bioflocculants alongside the high efficiency and cost-effectiveness of chemical flocculants, it is possible to develop a novel composite flocculation treatment technology. Previous research has primarily focused on wastewater treatment, mainly targeting nitrogen and phosphorus removal. Researchers investigated the combined treatment of high-algae water, kaolin, and humic acid-kaolin simulated water samples using bioflocculants mixed with various inorganic coagulants and achieved good synergistic coagulation effects [14-16]. However, current bioflocculant research still faces issues including high preparation costs [17], low flocculating capability of flocculant-producing strains [18], unstable flocculation effects [19], short preservation shelf life of bioflocculants [20], and unclear flocculation mechanisms [21].

This research applied composite bioflocculants combined with inorganic flocculants to enhance coagulation treatment of drinking water. The study explored the optimal dosing ratio and compounding methods by systematically examining key indicators including turbidity, color, trace organic matter, Al^{3+}, and Fe^{3+} and investigating removal effects in water. This research enriched the water source treatment technology system and filled the research gap regarding turbidity, color, and coagulant residual ion removal from source water. Furthermore, this study addressed the issue of high production costs associated with bioflocculants and offered a feasible solution for practical engineering applications by reducing the use of chemical coagulants, thereby mitigating the risk of secondary pollution from residual Al^{3+} and Fe^{3+} and improving floc settling performance. Additionally, it provided a theoretical basis for ensuring drinking water safety and advancing the industrial production and large-scale application of composite microbial flocculants.

Materials and methods

Resources of research materials and sampling

Polyferric aluminum chloride (PAFC) was obtained from Zibo Yuanrun Water Purification Technology Co., Ltd. (Zibo, Shandong, China) with the main components of 30.4% aluminum oxide, 1.5% iron oxide, 77% basicity, and pH 4.4. The flocs formed in water with higher density and compact structures at low temperatures. Solid composite aluminum-iron (industrial grade) was provided by Harbin Water Supply and Drainage Group (Harbin, Heilongjiang, China), which contained 6.5% aluminum oxide, 0.2 - 0.6% iron oxide, 60 - 85% basicity, and pH 3.5 - 5.0. The water samples were obtained from the Songhua River, Harbin, Heilongjiang, China with the quality parameters of 11.4 - 15.1℃, 17.8 - 43.1 NTU turbidity, pH 6.0 - 6.2, 0.2799 - 0.4401 mg/L Al^{3+}, 0.1962 - 0.35 mg/L Fe^{3+}, color 120 - 260, 2.486 - 5.623 mg/L total organic carbon (TOC), and UV_{254} 0.168 - 0.257.

Composite bioflocculant production

The straw generated after crop harvest in rural areas was used as fermentation raw material and was collected by the State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin, Heilongjiang, China. *Xylenes denitrificans* HIT-3, a cellulose-degrading bacterium, was isolated from soil samples collected on the Harbin Institute of Technology campus, while the flocculant-producing bacterial strains, *Agrobacterium radiobacter* F2, and *Bacillus sphaericus* F6, were originated from the Key Laboratory of Environmental Biotechnology (Harbin, Heilongjiang, China). *Xylenes denitrificans* HIT-3 were seeded and fermented on 10× concentrated Hutchinson medium containing 15 g of cellulose from straw, 1.0 g of KH_2PO_4, 0.3 g of $MgSO_4{\cdot}7H_2O$, 0.1 g of NaCl, 0.01g of $FeCl_3$, 2.5 g of $NaNO_3$ in distilled water up to 1,000 mL at pH 7.2 - 7.3 and incubated for 24 h. Both *Agrobacterium radiobacter* F2, and *Bacillus sphaericus* F6 were inoculated into liquid culture media containing 10 g of glucose, 5.0 g of K_2HPO_4, 2.0 g of KH_2PO_4, 0.2 g of $MgSO_4{\cdot}7H_2O$, 0.5 g of urea, 0.1 g of NaCl, 0.5 g of yeast extract in distilled water up to 1,000 mL at pH 7.5 and incubated at 33°C, 140 rpm for 24 h. The resulting liquid bacterial culture was used as inoculum [22]. The bacterial strains were inoculated for composite bioflocculant (CBF) production with two-stage fermentation methods to achieve coupling between the cellulose saccharification stage and the flocculant-producing stage, thereby producing an efficient composite biological flocculant.

Blending experiment and indicators determination

1,000 mL of raw water was mixed with PAFC to prepare the concentrations of 5, 10, 15, 20, and 25 mg/L with 5 replicates for each concentration. CBF was added to the five water samples at the concentrations of 0, 1, 2, 3, and 4 mg/L, respectively. Subsequently, a six-laboratory stirrer was used for coagulation. The samples were rapidly stirred at 200 rpm for 30 s followed by slow stirring at 60 rpm for 120 s. After standing for 20 min, the supernatant was collected from a depth of 2 cm below the liquid surface. The turbidity was measured by using SGZ-2P microcomputer turbidimeter (Yuefeng Company, Shanghai, China). Color was measured by using HI93727 colorimeter (Hanna Instruments Co., Ltd., Beijing, China). UV_{254} was determined by using T6 UV-vis spectrophotometer (Puxi General Instrument Co., Ltd., Beijing, China). Al^{3+} and Fe^{3+} were determined by using PerkinElmer 5300DV inductively coupled plasma optical emission spectrometer (PerkinElmer, Waltham, MA, USA), and TOC concentrations were measured by using Shimadzu TOC-VCPN total organic carbon/total nitrogen analyzer (Shimadzu, Kyoto, Japan). All the measurement results were used to determine the optimal combination ratio of the two coagulants [23]. Floc morphology was observed by using Hitachi S-4700 scanning electron microscope (SEM) (Hitachi, Tokyo, Japan).

Results and discussion

The effect of CBF combination on floc morphology

The results demonstrated that, in the flocculation without CBF addition, the formed flocs were small with loose edges, poor compactness, and no clear boundaries between individual flocs. In the flocculation process with CBF participation, the formed flocs were larger, exhibiting relatively regular porous rod-like structures with distinct boundaries between flocs that were closely connected to each other. The floc structure was more compact, and the floc particle size was

A. B.

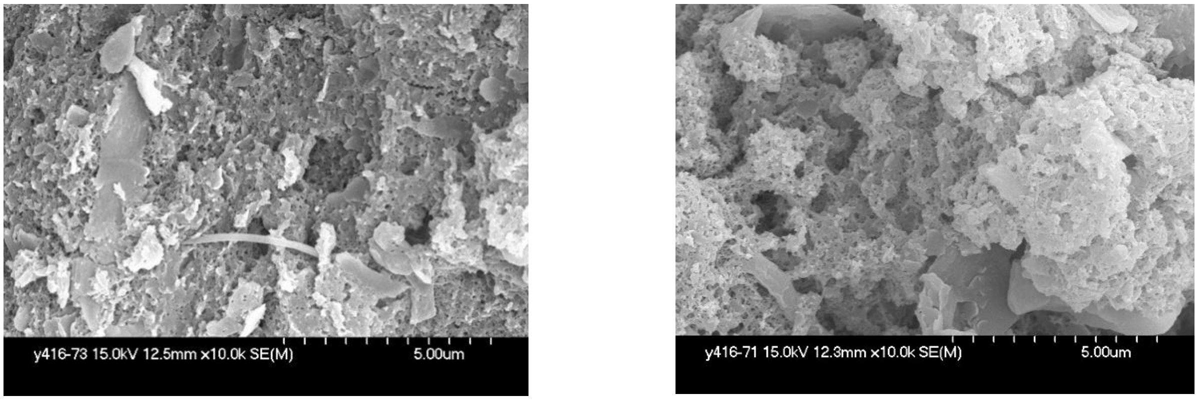


Figure 1. Flocs formed in the flocculation experiment before and after CBF combination. **A.** Flocs formed in the flocculation experiment without CBF combination. **B.** Flocs formed in the flocculation experiment with CBF combination.

larger than it was before enhanced flocculation (Figure 1). Therefore, the flocs exhibited better sedimentation performance, which effectively reduced sludge production and facilitated pollutant removal.

Turbidity removal by compound dosing of CBF and PAFC

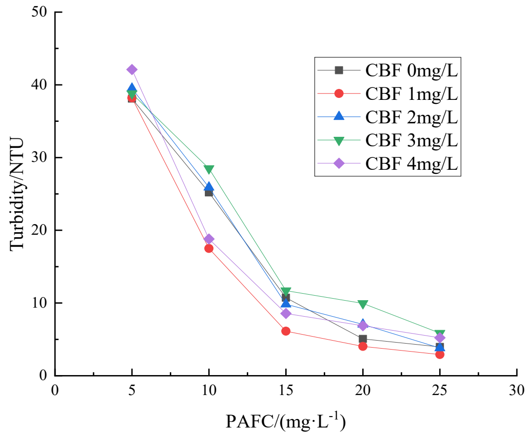
Natural waters generally exhibit a certain level of turbidity, which is mainly caused by colloidal substances dissolved from soil components, suspended particles such as silt and sand, as well as metabolic products from various animals and plants. Different dosages of the compound formulations resulted in different turbidity removal efficiencies in the water. At low dosages of PAFC, the flocculation phenomenon was insignificant with small flocs formed and a slow sedimentation rate. To achieve the same treatment performance, the dosages of both CBF and PAFC were significantly reduced in the compound dosing system, and the treatment effect of compound dosing was superior to that of PAFC dosed alone. When the dosages of CBF and PAFC were 1 mg/L and 25 mg/L, respectively, the effluent turbidity was 2.92 NTU with the removal efficiency reaching the maximum value of 90.93% among all treatments. Considering both treatment performance and economic factors comprehensively, the optimal dosages were determined as 1 mg/L for CBF and 20 mg/L for PAFC, achieving a turbidity removal efficiency of 90.59% (Figure 2a).

Residual iron concentration in the combination of CBF and PAFC

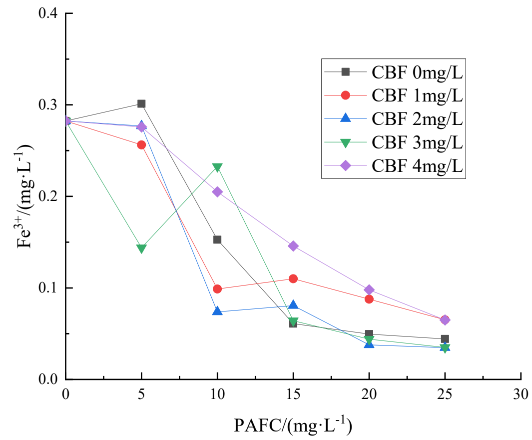
The average concentration of Fe^{3+} in raw water was 0.2994 mg/L. Although its average content was slightly lower than the national standard, most values within its fluctuation range exceeded the standard, indicating that the removal of Fe^{3+} was essential. The compound dosing of CBF and PAFC exhibited a significant effect on the removal of Fe^{3+}. When the dosages of CBF and PAFC were 2 mg/L and 25 mg/L, respectively, the residual Fe^{3+} concentration reached 0.03474 mg/L, corresponding to the maximum removal efficiency of 88.40% among all treatments. Considering the treatment effect and economic factors comprehensively, the optimal dosages of CBF and PAFC were determined to be 2 mg/L and 20 mg/L with an Fe^{3+} removal efficiency of 87.31%. At this dosage ratio, the residual Fe^{3+} concentration was 0.03799 mg/L, which met the requirement of 0.3 mg/L in the drinking water standard (Figure 2b).

Residual Al^{3+} concentration in the combination of CBF and PAFC

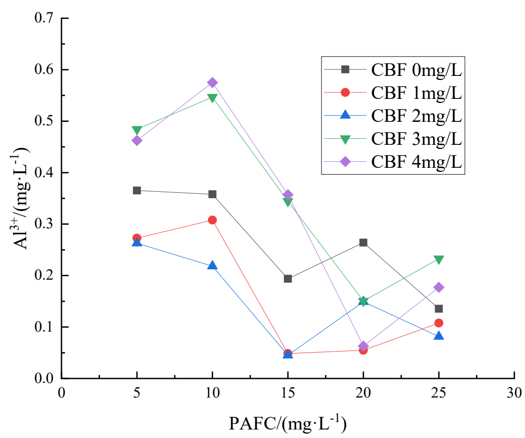
The residual Al^{3+} concentration in water after treatment with the two compounded flocculants demonstrated that an increase in PAFC dosage



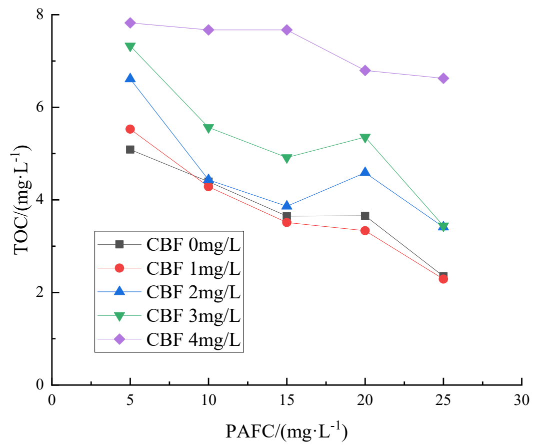
(a) Enhanced coagulation for turbidity removal by compound of CBF and PAFC



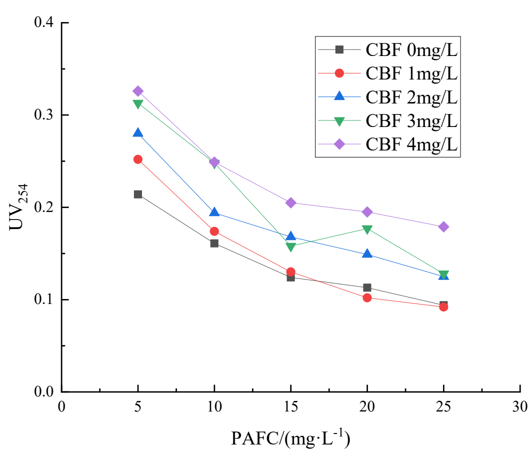
(b) Enhanced Coagulation for Residual Fe^{3+} Concentration by Compound of CBF and PAFC



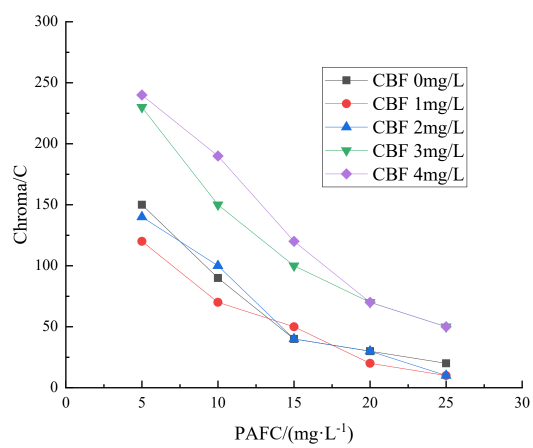
(c) Enhanced coagulation for residual Al^{3+} concentration by compound of CBF and PAFC



(d) Enhanced coagulation for TOC removal by compound of CBF and PAFC



(e) Enhanced coagulation for UV_{254} removal by compound of CBF and PAFC



(f) Enhanced coagulation for chromaticity removal by compound of CBF and PAFC

Figure 2. Removal effects of various indicators by CBF and PAFC combination.

resulted in a corresponding fluctuation in the residual Al^{3+} concentration of the water sample. When the single dosage of PAFC was 20 mg/L, the residual Al^{3+} concentration reached 0.264 mg/L. The average concentration of Al^{3+} in raw water was 0.3290 mg/L. The removal efficiency of residual aluminum was also significantly enhanced in the experiments with the compound application of the two flocculants. In addition, the presence of CBF inhibited the increase in residual aluminum concentration caused by aluminum introduced into water by PAFC. When the dosages of CBF and PAFC were 1 mg/L and 15 mg/L, respectively, the residual Al^{3+} concentration was only 0.05062 mg/L. Considering the treatment effect and economic factors comprehensively, the optimal compound ratio was determined as CBF:PAFC = 1 mg/L:15 mg/L with an Al^{3+} removal efficiency of 84.61%. At this ratio, the residual Al^{3+} concentration had met the requirement of 0.2 mg/L specified in the new drinking water standard (Figure 2c).

TOC removal by compound dosing of CBF and PAFC

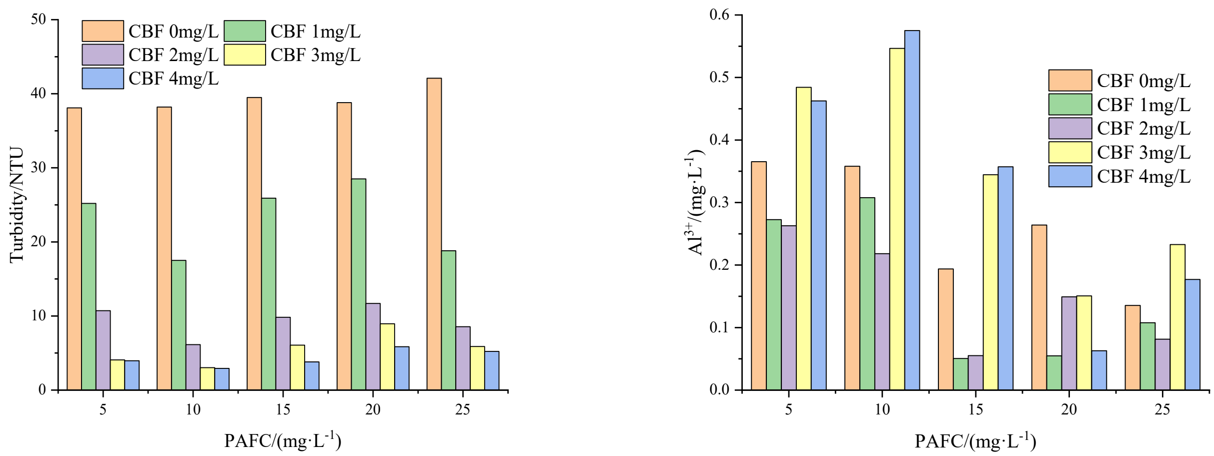
The compound dosing of CBF from 1 mg/L to 4 mg/L with PAFC all enhanced the TOC removal efficiency of raw water. When the dosages of CBF and PAFC were 1 mg/L and 25 mg/L, respectively, the TOC concentration was 2.089 mg/L with the removal efficiency reaching the maximum value of 48.91% among all treatments. However, the TOC removal efficiency started to decrease with a further increase of CBF dosage. At the CBF dosages of 2 mg/L and 3 mg/L, the residual TOC concentration was about 3.4 mg/L, corresponding to a removal efficiency of only 16.85%. Compared with the single addition of PAFC, the compound application of CBF and the corresponding PAFC exhibited an unsatisfactory TOC removal effect, which indicated that the compounding effect had become poor when the CBF dosage was 2 mg/L. With a continuous increase in CBF dosage, the TOC concentration in the effluent also rose. At a CBF dosage of 4 mg/L, the TOC removal efficiency was relatively low, and the TOC concentration was higher than that in the case of single PAFC addition, even exceeding the initial TOC concentration of the raw water. This result was caused by the organic matter residue from the flocculant itself. Nevertheless, the compounding of low concentration bioflocculant still significantly increased the TOC removal efficiency. Considering both the treatment effect and economic factors comprehensively, the optimal dosages of CBF and PAFC were determined to be 1 mg/L and 25 mg/L with a TOC removal efficiency of 48.91% and a TOC concentration of 2.089 mg/L at this dosage ratio (Figure 2d).

Ultraviolet absorbance (UV_{254}) removal by compound dosing of CBF and PAFC

The UV_{254} value of raw water ranged from 0.168 to 0.257 with the water temperature at approximately 13°C. The UV_{254} removal efficiency increased with the rise in flocculant dosage. When the dosages of CBF and PAFC were 1 mg/L and 25 mg/L, respectively, the UV_{254} value decreased to 0.092, corresponding to the maximum removal efficiency of 57.6% among all treatments. However, the UV_{254} removal efficiency started to decline with a further increase in CBF dosage. When the CBF dosage exceeded 1 mg/L, the UV_{254} removal effect became relatively weak with the removal efficiency lower than that of the experiment with PAFC alone. This phenomenon was caused by the organic matter residue from the flocculant itself. Therefore, considering both the treatment effect and economic factors comprehensively, the optimal dosages of CBF and PAFC were determined to be 1 mg/L and 20 mg/L, achieving a UV_{254} removal efficiency of 53.0% with a corresponding UV_{254} value of 0.102 (Figure 2e).

Chromaticity removal by compound dosing of CBF and PAFC

Chromaticity is one of the important water quality indices. The chromaticity exhibited by 1 mg/L platinum and 0.5 mg/L cobalt in water is defined as 1 degree, which is used as the standard chromaticity unit. Substances that induce chromaticity in water are dissolved or colloidal organic compounds with chromophoric groups such as phenols, triazenes, azo



(a) Effluent turbidity of combined dosing of PAFC and CBF

(b) Effluent Al^{3+} Concentration of combined dosing of PAFC and CBF

Figure 3. Removal effects of turbidity and Al^{3+} by PAFC-CBF combination.

compounds, and natural organic matter including humic acid, fulvic acid, and tannic acid, all of which can cause chromaticity to varying degrees. The removal of chromaticity is mainly achieved by the adsorption of color-generating organic molecules or the destruction of chromophoric groups on these organic molecules. The chromaticity of raw water ranged from 120 to 260 degrees with the water temperature at approximately 13°C. The compound dosing of PAFC exerted a significant effect on reducing the chromaticity of river water. When the dosages of CBF and PAFC were 1 mg/L, 25 mg/L and 2 mg/L, 25 mg/L, respectively, the chromaticity decreased to 10 degrees, corresponding to the maximum removal efficiency of 94.44% among all treatments. However, the chromaticity removal efficiency started to decline with a further increase in CBF dosage. When the CBF dosage reached 3 mg/L and above, the corresponding chromaticity was even higher than that in the case of PAFC dosed alone, which was attributed to the chromaticity residue induced by excessive bioflocculant. Considering both the treatment effect and economic factors comprehensively, the optimal dosages of CBF and PAFC were determined to be 1 mg/L and 20 mg/L, achieving a chromaticity removal efficiency of 88.89% with a corresponding chromaticity of 20 degrees (Figure 2f). This value had met the chromaticity requirement specified in the National Drinking Water Quality Standard (GB 5749-2022).

Selection of the optimal dosing ratio between CBF and PAFC

The results demonstrated that the compound application of the two flocculants played a crucial role in the removal of Fe^{3+} from water, and the treated water quality all met the national standards in terms of Fe^{3+}. However, PAFC was mainly composed of soluble aluminum salts, which introduced additional Al^{3+} into the water body. Therefore, the optimal dosing ratio was determined by mainly evaluating the turbidity and residual Al^{3+} concentration in water in accordance with the specifications of China's current National Drinking Water Quality Standard (GB 5749-2022). Meanwhile, the cost of doing and subsequent processes such as the secondary treatment of precipitated sludge were also taken into consideration. The removal efficiencies of turbidity and Al^{3+} in water by the compound application of PAFC and CBF showed that, in comparison with the single dosing of PAFC, the total dosage required to achieve the same level of optimal effluent performance after compound dosing was lower than that of the single PAFC dosing with the optimal dosages of CBF and PAFC being determined as 1 mg/L and 20 mg/L, respectively (Figure 3). Visual observation indicated a corresponding reduction in the total sludge production. The underlying reason for this phenomenon might be because that, during the flocculation and sedimentation process of PAFC as a single flocculant, the double electric layer structure of flocs was compressed under the influence of hydrodynamic stirring and other conditions, leading to floc destabilization and agglomeration. However, the floc could not be completely precipitated due to the limitation of their agglomeration size. In contrast, when CBF was applied in compound with PAFC, its macromolecular chain-like polysaccharide structure strengthened the sweep flocculation process during flocculation, which enabled a large number of small agglomerates to aggregate more intensively, forming flocs with larger mass and volume, which not only improved the sedimentation efficiency, but also optimized the effluent water quality. Based on a comprehensive evaluation of the removal efficiency of turbidity, UV_{254}, Al^{3+}, Fe^{3+}, TOC, and color, it was recommended that the optimal blending ratio of CBF to PAFC was 1 mg/L:20 mg/L. Meanwhile, the combined use of these two coagulants demonstrated significantly better purification performance compared to individual dosage of either coagulant alone. When comparing equivalent removal effects, the dosage of PAFC was reduced, which effectively saved on chemical consumption, minimized sludge discharge, and reduced secondary pollution.

Turbidity removal by compound dosing of CBF and CAF

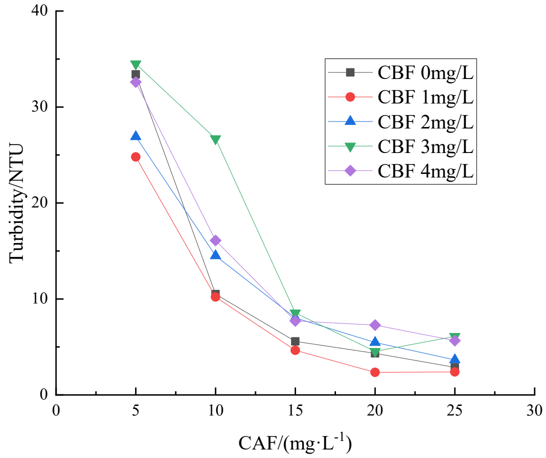
The compound application of the two coagulants significantly improved the turbidity removal performance. To achieve the same treatment effect, the total dosage of the combined CBF and CAF was lower than the dosage required when either coagulant was used alone. The optimal performance was obtained at dosages of 1 mg/L for CBF and 20 mg/L for CAF with the effluent turbidity reaching 2.35 NTU and a turbidity removal efficiency of 92.7%. Therefore, considering both treatment performance and economic factors comprehensively, the optimal dosages were determined as 1 mg/L for CBF and 20 mg/L for CAF (Figure 4a).

Residual Fe^{3+} concentration by compound dosing of CBF and CAF

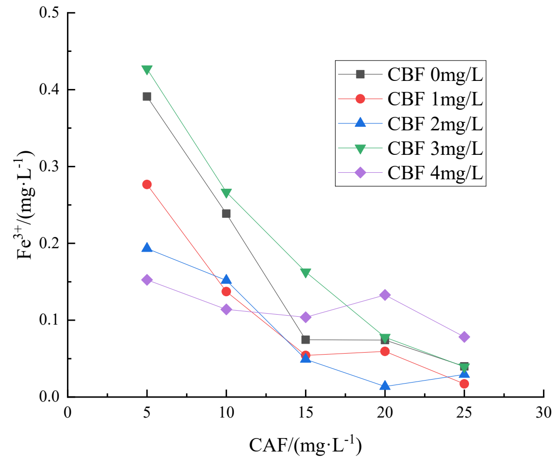
The concentration of Fe^{3+} in the raw water ranged from 0.1962 mg/L to 0.35 mg/L with the water temperature at approximately 13℃. The compound dosing of CBF and CAF coagulant exhibited a significant effect on the removal of Fe^{3+} in all cases. When the dosages of CBF and CAF coagulants were 2 mg/L and 20 mg/L, respectively, the residual Fe^{3+} concentration decreased to 0.01375 mg/L, representing an extremely low level in the treated raw water with the removal efficiency reaching the maximum value of 95.41% among all treatments. Considering both the treatment performance and economic factors comprehensively, the optimal dosages were determined as 1 mg/L for CBF and 15 mg/L for CAF, achieving an Fe^{3+} removal efficiency of 81.88% with a corresponding residual Fe^{3+} concentration of 0.05424 mg/L, which was far below the limit of 0.3 mg/L specified in the national standard (Figure 4b).

Residual Al^{3+} concentration by compound dosing of CBF and CAF

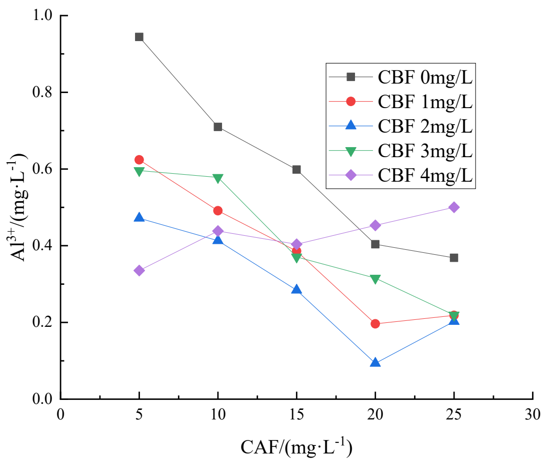
The concentration of Al^{3+} in raw water ranged from 0.2799 mg/L to 0.4401 mg/L with the water temperature at approximately 13°C. The removal efficiency of residual Al^{3+} was significantly enhanced in the experiments with the compound dosing of the two coagulants, indicating that their combination exerted a remarkable effect on Al^{3+} removal. When the dosages of CBF and CAF were 2 mg/L and 20 mg/L, respectively, the residual Al^{3+} concentration was reduced to 0.09325 mg/L with the removal efficiency reaching the maximum value of 71.66% among all treatments. The results demonstrated that the addition of CBF could effectively facilitate the removal of Al^{3+}. The presence of CBF inhibited the increase in residual Al^{3+} concentration caused by the Al^{3+} introduced into the water by the CAF. However, with the continuous addition of CAF,



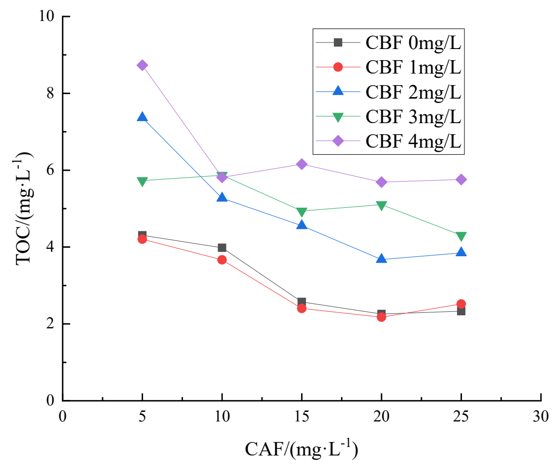
(a) Enhanced coagulation for turbidity removal by compound of CBF and CAF



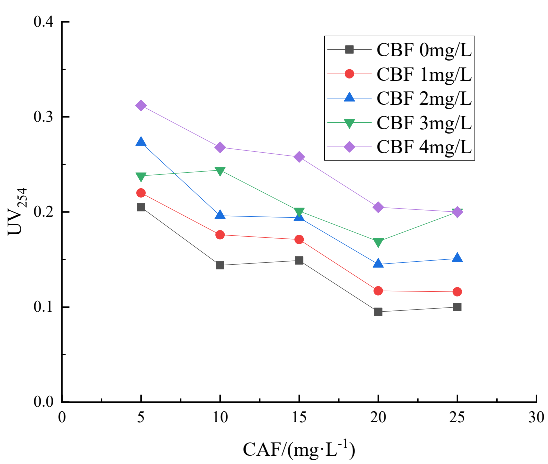
(b) Enhanced coagulation for residual Fe^{3+} concentration by compound of CBF and CAF



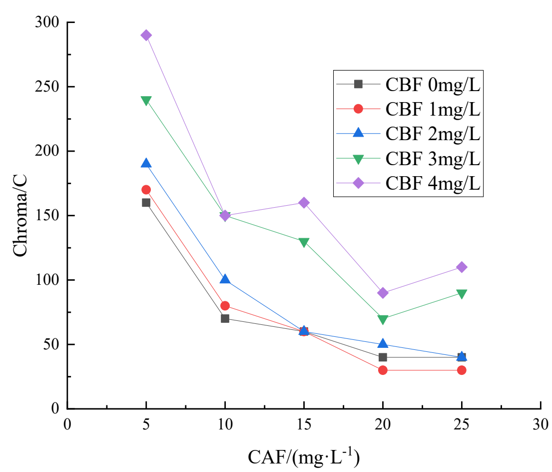
(c) Enhanced coagulation for residual Al^{3+} concentration by compound of CBF and CAF



(d) Enhanced coagulation for TOC removal by compound of CBF and CAF



(e) Enhanced coagulation for UV_{254} removal by compound of CBF and CAF



(f) Enhanced coagulation for chromaticity removal by compound of CBF and CAF

Figure 4. Removal effects of CBF and composite aluminum-iron combination on various factors.

the residual Al^{3+} concentration began to fluctuate, which might be attributed to the excessive Al^{3+} introduced into the raw water by the CAF. Therefore, considering both treatment performance and economic factors comprehensively, the optimal dosages were determined as 2 mg/L for CBF and 20 mg/L for CAF, and the effluent Al^{3+} concentration met the limit of 0.2 mg/L specified in the drinking water standard (Figure 4c).

TOC removal by compound dosing of CBF and CAF

The average TOC concentration of raw water was 4.089 mg/L. The TOC removal efficiency of raw water was improved with the increase in the dosage of CAF when 1 mg/L to 3 mg/L of CBF was combined with CAF. However, the TOC removal performance became unsatisfactory when the dosage of CBF exceeded 1 mg/L with the effluent TOC concentration even higher than that achieved by the single dosing of CAF at the corresponding dosage. When the CBF dosage reached 4 mg/L, there was almost no TOC removal effect, and the effluent TOC concentration was higher than the initial value of the raw river water, which could be attributed to the residual organic matter from the flocculant itself. Considering both treatment performance and economic factors comprehensively, the optimal dosages were determined as 1 mg/L for CBF and 20 mg/L for CAF, achieving a TOC removal efficiency of 46.81% with a corresponding TOC concentration of 2.175 mg/L (Figure 4d).

UV_{254} removal by compound dosing of CBF and CAF

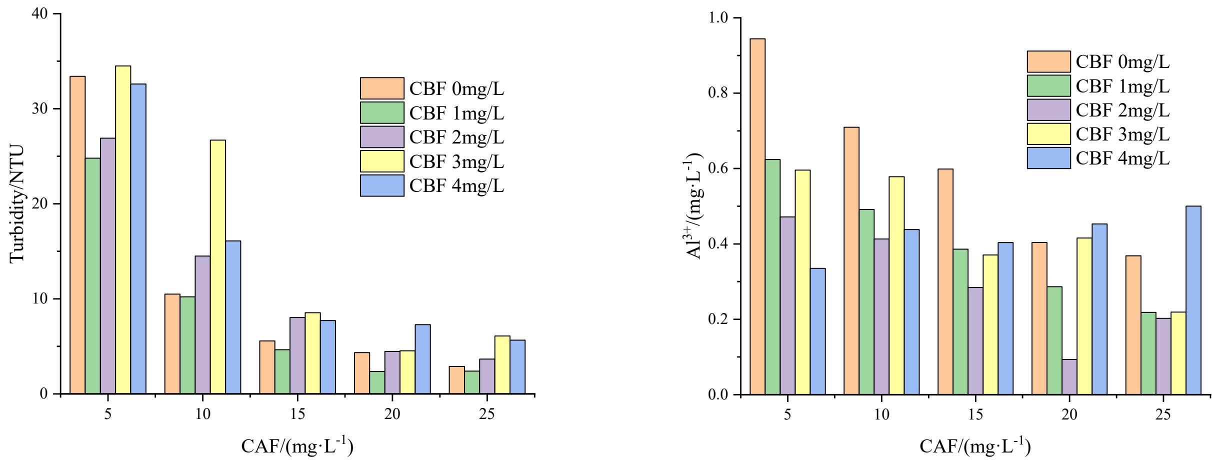
The UV_{254} value of raw water ranged from 0.168 to 0.257. The UV_{254} removal efficiency of raw water by the compound dosing of CBF and CAF was improved with the increase in the dosage of CAF. However, the UV_{254} removal performance was unsatisfactory after compounding with CBF, which was lower than that achieved by the single dosing of CAF at the corresponding dosage. With the continuous increase in CBF dosage, the removal efficiency decreased consistently, and a higher CBF dosage led to poorer treatment performance, which could be attributed to the residual organic matter from the flocculant itself. Considering both treatment performance and economic factors comprehensively, the optimal dosages were determined as 1 mg/L for CBF and 20 mg/L for CAF, achieving a UV_{254} removal efficiency of 46.08% with a corresponding UV_{254} value of 0.117 (Figure 4e).

Chromaticity removal by compound dosing of CBF and CAF

The chromaticity of raw water ranged from 120 to 260 degrees with the water temperature at approximately 13°C. The compound dosing of low concentration CBF and CAF exerted a significant effect on reducing the chromaticity of river water. When the dosages of CBF and CAF were 1 mg/L and 20 mg/L, respectively, the effluent chromaticity decreased to 30 degrees with the removal efficiency reaching the maximum value of 83.33% among all treatments. However, the chromaticity removal performance started to decline with the continuous increase in CBF dosage. When the CBF dosage was 2 mg/L or higher, the corresponding effluent chromaticity value was slightly higher than that achieved by the single dosing of CAF, which might be attributed to the chromaticity residue caused by excessive bioflocculant. Considering both treatment performance and economic factors comprehensively, the optimal dosages were determined as 1 mg/L for CBF and 20 mg/L for CAF coagulant (Figure 4f).

Selection of the optimal compound ratio between CBF and CAF

The removal effects of composite aluminum-iron and CBF combination on turbidity and Al^{3+}showed that, when the CAF was used alone as a flocculant, it achieved a satisfactory turbidity removal performance, whereas the residual Al^{3+} concentration in the effluent exceeded the standard limit. Compared with the single dosing of CAF, the total dosage required to achieve the same level of optimal effluent performance was lower for the compound dosing system. The overall purification efficiency of the compound



(a) Effluent turbidity of combined dosing of CAF and CBF

(b) Effluent Al^{3+} concentration of combined dosing of CAF and CBF

Figure 5. Removal effects of composite aluminum-iron and CBF combination on turbidity and Al^{3+}.

dosing was significantly superior to that of either coagulant used alone (Figure 5). Based on a comprehensive evaluation of the removal efficiency of turbidity, UV_{254}, Al^{3+}, Fe^{3+}, TOC, and color, it was recommended that the optimal blending ratio of CBF to CAF was 2 mg/L:20 mg/L. Compared with the combined addition of CBF and CAF, when achieving equivalent optimal effluent quality levels, the overall chemical dosage was lower than that required for the standalone addition of compound aluminum-iron coagulant alone. Furthermore, the purification performance of the blended treatment demonstrated significantly better results than either coagulant added individually.

Conclusion

For the combination of CBF and PAFC, the optimal ratio was 1 mg/L and 20 mg/L, respectively, demonstrating significant removal effects on turbidity, Fe^{3+}, Al^{3+}, TOC, UV_{254}, and color with purification efficiency superior to individual addition. For the combination of CBF and the composite aluminum-iron coagulant, the optimal ratio was 2 mg/L and 20 mg/L, respectively. Except for the less satisfactory removal of UV_{254}, this combination also exhibited excellent removal effects on other indicators. The addition of CBF improved floc structure, thereby enhancing sedimentation efficiency. The combined use of these two coagulants demonstrated significantly better purification performance compared to individual dosage of either coagulant alone, effectively removing residual Al^{3+} and Fe^{3+} ions and reducing chemical consumption. However, the TOC removal efficiency started to decline with a further increase of CBF dosage, which was caused by the organic matter residue from the flocculant itself. The combination of CBF with inorganic coagulants could efficiently remove multiple pollutants, improve sludge settling performance, and reduce chemical consumption. These findings provided a theoretical basis for ensuring drinking water quality safety and facilitating the industrial application of this technology.

Acknowledgements

This study was sponsored by the Research Project on Science and Technology of Nanyang City (Grant No. 24KJGG066, 25JCQY002) and supported by Innovative Technology Team for Water Pollution Control and Solid Waste Resource Utilization of Nanyang Institute of Technology.

References

1. Guan Y, Qiang Y, Li B, Xiao Y, Zhang NN. 2025. Environmental hazard and human health risk attributed to surface water pollution in China: City-level assessment and socioeconomic driver identification. J Hazard Mater. 501:140827.
2. Pellikainen P, Eikebrokk B, Myrmel M, Lavonen E. 2025. Investigating log removals of bacteria and viruses at a full-scale enhanced coagulation contact filtration drinking water treatment plant. J Water Process Eng. 77:108393.
3. Ge JY, Tian HY, Li L, Han YP, Liu JW. 2024. Preparation of a novel organic-inorganic composite sludge bioflocculant (SBF) from dewatered sludge as raw material: Characteristics, flocculation mechanism and application for domestic sewage. Bioresource Technol. 416:131747.
4. Qin SQ, Li Q, Wu LY, Tian HM, Man YY, Liu GF. 2025. Investigation on flocculation performance of bioflocculants for dyes in water. Sci Rep. 15:23577.
5. Chen Z, Li ZP, Liu PZ, Liu Y, Wang YP, Li QB, *et al*. 2017. Characterization of a novel bioflocculant from a marine bacterium and its application in dye wastewater treatment. BMC biotechnology. 17(1):84.
6. Jia BJ, Yu JM. 2012. The research status and development trend of microbial flocculants. Phys Procedia. 24:425-428.
7. Zhu YB, Ma F, Li DP. 2008. Study on fermentation performance of microorganisms producing compound bioflocculants. China Water Wastewater. 24(19):70-73.
8. Budi KS, Fauzul IM, Sheikh ASR, Razi OA, Abu HH. 2023. Coagulation-flocculation of aquaculture effluent using biobased flocculant: From artificial to real wastewater optimization by response surface methodology. J Water Process Eng. 53:103869.
9. Tlou SN, Tsolanku MS, Kgabo M, Peter M, Vusimuzi M. 2022. Isolation and optimisation of culture conditions for a marine bioflocculant-producing bacterium and application of its bioflocculant in wastewater treatment. Int J Env Res Pub He. 19(16):1.
10. Laschiwitz Beghetto C, Ramos RMB, Monteiro PI, De Jesus Bassetti F, Almeida Coral LA. 2025. Moringa oleifera and polyaluminum chloride: Coagulant combinations for cyanobacteria removal in drinking water. Acs Omega. 10(35):39712-39723.
11. Zarchi I, Friedler E, Rebhun M. 2013. Polyaluminium chloride as an alternative to alum for the direct filtration of drinking water. Environ Technol. 34(9-12):1199-1209.
12. Budi KS, Sheikh ASR, Fauzul IM, Mohd SNS, Ismail NI, Abu HH, *et al*. 2020. Challenges and opportunities of biocoagulant/ bioflocculant application for drinking water and wastewater treatment and its potential for sludge recovery. Int J Env Res Pub He. 17(24):93.
13. Okaiyeto K, Nwodo UU, Okoli SA, Mabinya LV, Okoh AI. 2016. Implications for public health demands alternatives to inorganic and synthetic flocculants: Bioflocculants as important candidates. Microbiol Open. 5(2):177-211.
14. Ma F, Liu JL, Li SG, Yang JX, Zhang LQ, Wu B, *et al*. 2003. Development of compound microbial flocculants. China Water Wastewater. 19(4):1-4.
15. Yang Y, Jiang CC, Wang X, Fan LJ, Xie YW, Wang DH, *et al*. 2025. Unraveling the potential of microbial flocculants: Preparation, performance, and applications in wastewater treatment. Water. 16(14):16141995.
16. Yuan S, Zhang J, Hou QJ, Zhang YW, Li WJ. 2023. Two-stage injection of polymer and microsand during ballasted flocculation for treating kaolin waters with or without humic acid: Floc evolutional characteristics, performance and mechanisms. Environ Res. 263(2):116102.
17. Liu C, Sun D, Liu JW, Zhu JR, Liu WJ. 2021. Recent advances and perspectives in efforts to reduce the production and application cost of microbial flocculants. Bioresour Bioprocess. 8:51.
18. Ding R, Luo LP, Han RX, Zhang ML, Li TT, Tang JH, *et al*. 2021. Rapid production of a novel Al (III) dependent bioflocculant isolated from *Raoultella ornithinolytica* 160-1 and its application combined with inorganic salts. Front Microbiol. 11:622365.
19. Feng Z, Yang ZZ, Cao JG, Wu ZG, Gai EX, Wu LY. 2024. Flocculation of kaolin and anion dye by cationic cellulose-based flocculant: RSM-optimized synthesis and experimental study. J Environ Chem Eng. 12(2):112309.
20. Hu M, Hei RN, Guo DJ, Luo J, Lu C, Xu WL, *et al*. 2023. Shelf-life enhancement of bio-inoculants through synergistic effects of encapsulation technology and osmotic protectants. J Environ Chem Eng. 5(11):110996.
21. Mishra B, Kallem P, Yadavalli R, Mandal SK, Reddy CN, Sumithra B, *et al*. 2025. Industrial wastewater treatment using extracellular polymer substances/bioflocculants: A review. Appl Water Sci. 15(3):1-20.
22. Li LX, Deng LH, Piao YJ, Ma F, Wang J. 2019. Effect of combined use of composite microbial flocculant and chemical flocculant on surface water treatment. J Heilongjiang Univ Sci Technol. 29(3):376-381.
23. Li LX, Xing J, Ma F, Zhan Y, Zhu YW. 2016. Removal efficiency of turbidity and color from water sources by composite bio-flocculant. J Heilongjiang Univ Sci Technol. 26(5):524-527.