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Using potassium ferrate as advanced treatment for municipal wastewater

Hongbo Wang[a,b,*], Haining Li[a], Ning Ding[a], Mei Li[a], Ning Wang[a,*]

[a]*School of Municipal and Environmental Engineering, Shandong Jianzhu University, Jinan 250101, Shandong, China, email: wanghongbo@sdjzu.edu.cn (H. Wang), 986272112@qq.com (H. Li), 1223478347@qq.com (N. Ding), limei@sdjzu.edu.cn (M. Li), wangning@sdjzu.edu.cn (N. Wang)*
[b]*Shandong Co-Innovation Center of Green Building, Jinan, China*

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ABSTRACT

Potassium ferrate is a potential high efficient chemical for wastewater treatment as a combination of oxidant and flocculent. In this research, potassium ferrate was used to treat simulated wastewater and secondary effluent of a wastewater treatment and the removal performance was evaluated base on the treatment of chemical oxygen demand (COD), total organic carbon (TOC), suspended solid (SS) and total phosphorous (TP). The results present that the organic matters (indexed by COD and TOC) could be effectively eliminated during both the oxidation stage and flocculation stage while SS and TP were majorly got rid of during flocculation stage. The effects of pH, reaction time and potassium ferrate dosing were also investigated and the optimum conditions were found as: The potassium ferrate concentration of 40 mg/L, oxidation time of 20 min (pH of 10.31), and flocculation time of 30 min (pH of 3.28) except TP and SS removals; For secondary effluent, potassium ferrate of 40–50 mg/L, 25 min of oxidation, and 20 min of flocculation.

Keywords: Potassium ferrate; Municipal wastewater; Advanced treatment

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1. Introduction

Treated municipal wastewater is usually discharged into the surrounding water bodies, and to a certain extent, influences the quality of drinking water indirectly. Previous research has identified the effluent of municipal sewage as one of the main source of micro pollutants (pollutants at trace concentrations) into natural water cycle, such as hormones, drugs, etc. [1]. Currently, it is still a problem that, in traditional waste water treatment plants (WWTP), the lack of capacity to eliminate residual pollutants from the secondary effluent after biochemical treatment because of various sewage sources and complicate compositions. For instance, some recalcitrant organic matters were suggested to be removed with subsequent treatment processes via improving biodegradability [2]. In addition to chemical oxygen demand (COD), total phosphorus (TP), suspended solid (SS) and other conventional pollution indicators, total organic carbon (TOC) and UV_{254} are also used to characterize secondary effluents [2]. As an environment-friendly agent, potassium ferrate (K_2FeO_4) can effectively remove some remaining pollutants, such as bacteria [3,4], algae [3,4], organic matter [5–7], inorganic matter [8], heavy metals [9–11], and other pollutants [3,4], since it could serve as oxidant, adsorbent, flocculant, coagulant, sterilant, and deodorant simultaneously [12–14]. The final product of potassium ferrate is $Fe(OH)_3$, which is an excellent inorganic flocculant removing suspended matters, phosphorus, and other unexpected substances in the sewage without causing secondary pollution [15]. Potassium ferrate efficiently decreases turbidity and dephosphorization at a wide pH range [8,16]. The valence of iron as potassium ferrate is 6+, implying the strong oxidation potential. When oxidizing organic matter, some intermediate states, such as Fe^{5+} or Fe^{4+}, hydrolyze and form a large network structure which can compress and neutralize the colloidal diffusion layer with negative charges, resulting in flocculation and capturing pollutants [17]. Wu [18] proposed that K_2FeO_4 at

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*Corresponding author.

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a dose of 500 mg/L could effectively oxidize and decompose sludge to improve the dewatering and biodegradability of sludge. Moreover, previous studies [14] have shown that, compared with other iron and aluminum salts, potassium ferrate had higher removal rate of SS in surface water and shorter time to precipitation, implying the potential as flocculant.

It is more cost-effective to use potassium ferrate to treat secondary effluent than other advanced treatment of wastewater [8]. Previous studies have shown that the promising potential of potassium ferrate as both coagulant and disinfectant for the treatment of secondary effluent evaluated by some relevant parameters (SS, orthophosphate, ammonia, total bacteria, and coliform reduction) [19]. In urban secondary wastewater treatment, lower doses of ferrate could decrease the indigenous bacteria by 99.9% and be effective to chlorine-resistant bacteria, compared with other traditional disinfectants [20]. Gombos [21] found that Fe (VI) could also reduce the total organic carbon (TOC) in secondary treatment of wastewater. Yang [22] reported the effectiveness of Fe(VI) treatment as a tertiary treatment technology for micro pollutants (endocrine disrupting chemicals, pharmaceuticals, and personal care products) in second effluent. However, besides chemical oxygen demand (COD), total phosphorus (TP), and suspended solid (SS), total organic carbon (TOC) and UV_{254} are also used to characterize secondary effluent [2], on which little research focused systemically.

In the present study, to evaluate the oxidation and flocculation of potassium ferrateas advanced treatment for secondary effluent of municipal wastewater, artificial wastewater, simulating the secondary effluent,was used to investigate the effect of operational parameters including ferrate concentration, pH, time, and other factors on removing COD, TOC, TP, SS, and UV_{254}. Additionally, potassium ferrate was also employed to treat actual secondary effluent to verify its feasibility as advanced treatment for municipal wastewater.

2. Materials and methods

2.1. Chemicals and water samples

Used regents were potassium ferrate (purchased from the Municipal Laboratory of Zhejiang University, China); potassium dichromate, potassium dihydrogen phosphate, glucose, and kaolin (AR, Sinopharm Chemical Reagents Co., Ltd., China); ultra-pure water(resistivity = 18.6 Momega, pH = 2.5). The purity of potassium ferrate was above 95%, measured by spectrophotometry according to the method of Lee [23].

To minimize unexpected interference, artificial wastewater used in the experiments was prepared with ultrapure water and appropriate concentrations of glucose (about 47 mg//L), potassium dihydrogen phosphate (about 1.6 mg/L), and kaolin (about 15 mg/L) by referring to the water quality of the secondary effluent of Ever bright WWTP. The pH of artificial wastewater was adjusted with 1% hydrochloric acid and 1 mol/L sodium hydroxide. The quality of the artificial wastewater is shown in Table 1.

The influent, secondary effluent, and effluent samples were fully loaded into vials from Ever bright WWTP and stored at 4°C. The water quality of water samples of Ever bright WWTP was measured and is shown in Table 2.

Table1
Characteristic parameters of artificial wastewater

COD (mg/L)	49.3–54.4
TOC (mg/L)	15.2– 20.8
TP (mg/L)	0.35–0.40
SS (mg/L)	11–18
pH	6.73–6.98

Table 2
Characteristic parameters of influent, secondary effluent and effluent measured in the Ever bright WWTP

Index	Influent	Secondary effluent	Effluent
COD (mg/L)	519–621	49.4–56.2	24.6–31.9
TOC (mg/L)	147–175	15.6–20.5	7.2–9.3
TP (mg/L)	5.21–8.41	0.36–0.40	0.34–0.41
UV_{254} (cm^{-1})		0.12–0.15	0.12–0.21
SS (mg/L)	216–371	12–17	12–17
pH	6.34–8.26	6.76–7.03	6.51–7.84

2.2. Methodology

2.2.1. Oxidation and flocculation experiments

For oxidation experiments, certain mass of potassium ferrate was added in 800 mL of simulated water with pH and reaction time adjusted to expected values. The timing stirring instrument (MY3000-6, Wu Han Meiyuyiqi Co., Ltd., China) was adjusted to 300 rpm at room temperature. After the reaction, the sample was added with Na_2SO_3 solution to stop the oxidation and stood still for 20 min. The indicators of quality of supernatant water samples, such as TOC, COD, TP, SS, etc., were monitored before and after treatment to obtain suitable oxidation conditions.

Flocculation experiment was conducted under the optimized oxidation conditions. The stirring instrument was set to 300 rpm at room temperature after the Na_2SO_3 solution was added to end the oxidation process. Then, the stirring speed was adjusted to 50 rpm after the pH and time were changed in water sample. Certain amount of supernatant was sampled after 20-min standing at the end of the flocculation to obtain optimum conditions.

2.2.2. Performance of potassium ferrate for secondary effluent from sewage treatment plant

Secondary effluent of Ever bright Water No.1 sewage treatment plant was used for oxidation and flocculation experiments with monitored indicators (COD, TOC, TP, SS, and SUVA). SUVA was defined as SUVA = 100 × UV_{254}/TOC, which reflected humification of organic matters in sewage, molecule size, and the number of organic matters with unsaturated carbon bonds or aromatic rings [24,25].

3. Results and discussion

3.1. Oxidation experiments of potassium ferrate

3.1.1. Effect of pH

The forms of potassium ferrate ions vary at different pH values, indicating that the stability is greatly affected by pH [24,25]. The results of pH effect are shown in Fig. 1. The removal COD and TOC favored alkaline conditions and the removal efficiency increased with the pH while TP and SS decreased. When pH was 10.31, the removal of COD and TOC achieved the highest.

It can be seen that the removal of organic compounds, TP, and SS by potassium ferrate changed with pH in the oxidation stage; and compared to COD and TOC, the less effects of TP and SS were presented. This is because that the stability and oxidation of potassium ferrate solution is affected by the pH of the solution. It was reported [26] that Fe (VI) had four forms: $H_3FeO_4^+$, H_2FeO_4, $HFeO_4^-$, and FeO_4^{2-}, which had different redox capacity and redox potential. In acidic solutions, more Fe (VI) could quickly trig redox reaction because of the high oxidation potential (+2.20V) [13,27] due to the instantaneous decomposition of potassium ferrate, while Fe (VI) reduced to Fe (III), like a flocculent $Fe(OH)_3$ with high adsorption activity, when part of TP and SS were removed due to flocculation in the oxidation stage. But the reaction ended before all the organic compounds having contacted with potassium ferrate. Wronska [28] found that the self-decomposition of FeO_4^{2-} decreased with the increase of OH^- in a certain alkalinity range, due to less collision of FeO_4^{2-} molecules. Therefore, not all the potassium ferrate decomposed at pH 7–10, resulting in remaining strong oxidization potential. Under alkaline conditions, both firm stability of potassium ferrate and long contact time with organic compounds benefited the oxidation, regardless of the negative effect of lower redox potential until the pH was greater than 10, when the oxidation capacity was harmed and the removal rates of COD and TOC were slightly reduced.

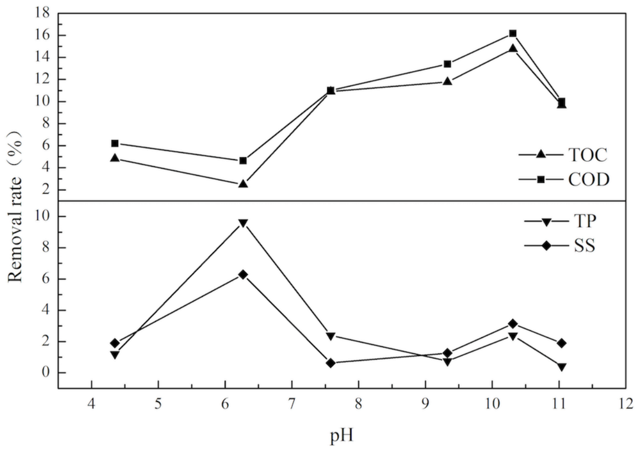

Fig. 1. Removal rate of TOC, COD, TP, SS at different pH (ferrate (VI) dosing of 20 mg/L and oxidation time of 10 min).

3.1.2. Effect of ferrate (VI) dosing and oxidation time

The removal of TOC, COD, TP, and SS was improved with increasing dosing and oxidation time as shown in Fig. 2. With concentrations of potassium ferrate increasing, the water quality indicators gradually decreased. The removal of COD and TOC was faster than SS and TP. When the lowest COD and TOC occurred when the concentration was 40 mg/L, the lowest concentrations of COD and TOC were 39.12 mg/L and 13.32 mg/L respectively, corresponding to removal efficiencies of 20.25% and 20.45% respectively. As shown in Fig. 2b, in the beginning 25 min, all four indexes decreased with time. In the following five minutes, the concentrations of COD and TOC became stable. Compared these two indexes, the removal of TP and SS were not obvious.

Previous study found that the oxidation of potassium ferrate removed the pollutants through the electron transfer, such as Fe(VI)→Fe(V)→Fe(III) [26]. During the transfer process, $HO^\bullet$ and intermediate valence iron compounds were formed, both of which were strong oxidizing regents, resulting in the removal of organic pollutants during the oxidation stage, while the flocculation ability of Fe (III) also captured some TP and SS. However, the high reaction rate of oxidation, to some extent, defected the flocculation and sedimentation, and the flocculation removal of TP and SS was much less than the oxidation removal of organic matter. These observations in Fig. 2 could be explained by that, with the dosing or reaction time increasing, there were more contact opportunity and duration between potassium ferrate and organic, resulting in more COD and TOC eliminated. Meanwhile, more floccus formed in the solution, promoting the removal phosphorus and suspended matter to a certain degree. On the other hand, it was reported that the stability of potassium ferrate was affected by the concentration in water; usually, the smaller concentration of potassium ferrate resulted in higher stability [25]. As time increasing, the decomposition rate of potassium ferrate gradually increased [29]. Therefore, with the concentration of potassium ferrate or time increasing, the removal of organic matter would not continuously rise.

3.2. Flocculation experiments of potassium ferrate

Base on the above experiments in section 3.1, in the oxidation stage, potassium ferrate has a certain removal effecton the organic matter, accompanied by the flocculation of a slight amount of TP and SS. The appropriate oxidation conditions were chosen as 40 mg/L of potassium ferrate, 10.31of pH, 25 min of oxidation time, which were used for the flocculation experiments. It was reported that the coalescence of ferrate is more efficient than that of ferric salts and ferrous salts [16], and the flocculation time of 15 min was determined in the following section 3.2.2 for investigation of optimal flocculation time.

3.2.1. Effect of pH

Fig. 3 shows that with the increase of pH, the concentrations of COD and TOC decreased firstly and then increased. When pH was 6.93, the COD and TOC reached the minimum with total removal rate of 26.02% and 25.93%, respectively. Compared with the removal rate of COD and TOC

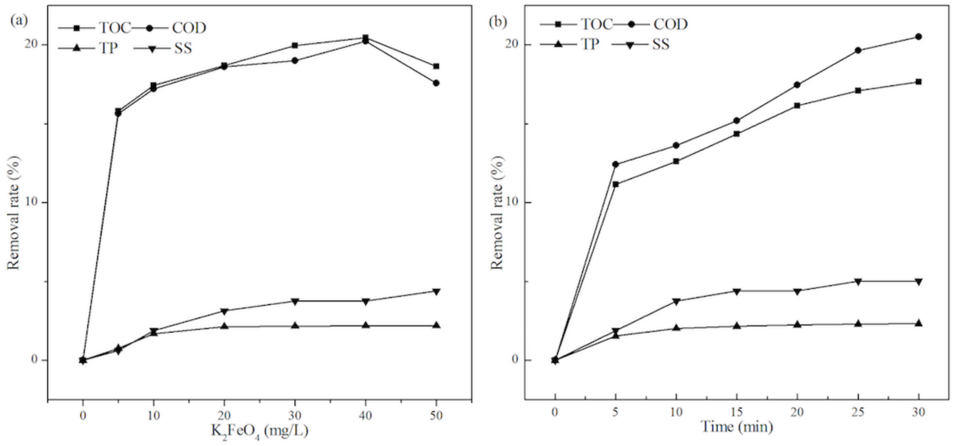

Fig. 2. Removal rate of TOC, COD, TP, SS with (a) different ferrate (VI) dose (pH 10.31 and oxidation time of 10 min) and (b) different oxidation times (pH 10.31 and ferrate (VI) dose of 40 mg/L), respectively.

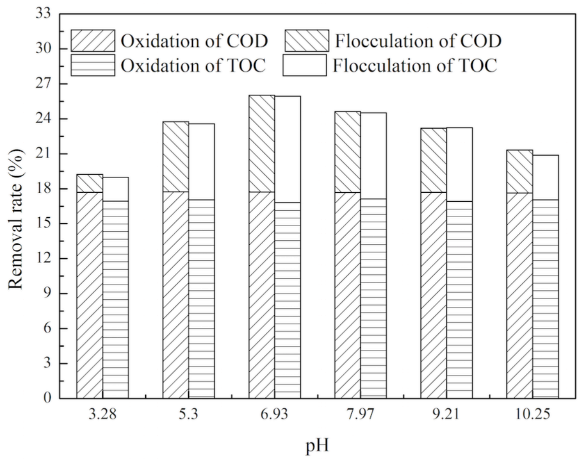

Fig. 3. Removal rate of COD and TOC at different flocculation pH (ferrate (VI) dose of 40 mg/L, flocculation time of 15 min and oxidation pH 10.31).

in the oxidation stage of 17.72% and 16.79% respectively, the removal rate in the flocculation stage were 8.30% and 9.13%, which were smaller than oxidation stage obviously, indicated that the process of potassium ferrate was mainly due to the removal of organic matter through the oxidation stage.

It was reported that, under acidic conditions, the ferrate decomposed into low polymerization degree and high charge hydrated Fe_2O_3, while likely into ferric hydroxide under neutral condition [30]. The hydroxide compound tended to bridge organic molecules into higher inorganic polymers, increasing flocculation area for organic matters. As shown in Fig. 3, at the neutral conditions, more organic matters were removed.

As shown in Fig. 4a, at acidic conditions, the removal of TP at the flocculation stage was better, especially when pH was 3.28, the total removal rate was 48.22%. Compared with the removal rate of oxidation stage, it could indicate that potassium ferrate removed TP mainly by flocculation. Potassium ferrate had the dual effect of oxidation and flocculation, which can be based on the oxidation of organo phosphate to use its flocculation to further remove inorganic phosphorus. After the oxidation stage, potassium ferrate was mainly in the form of potassium ferric. At acidic conditions, the unstable potassium ferric produced a large number of Fe^{3+} ions, promoting flocculation and TP removal by accelerating the formation of ferric phosphate co-precipitate compounds [31].

From Fig. 4b it can be seen that when pH was 3.28, the removal effect of SS at flocculation stage was the best and the total removal rate was 62.39%. In contrary, oxidation removal rate of SS only accounted for 3.83%, implying that the flocculation stage dominated the removal of SS. When pH was 6.93, SS ended up with only 9 mg/L, meeting the emission requirements of SS in discharge standard of pollutants for municipal wastewater treatment plant (GB18918-2002).

3.2.2. Effect of flocculation time

It can be seen from Fig. 5 that COD and TOC were gradually removed with time, and the descent trend of COD and TOC were slowed down. When the time reached 35 min, the total removal rates of COD and TOC were 23.99% and 23.41%, respectively. The removal rates of COD and TOC were 17.01% and 17.04% respectively in the oxidation stage while 6.98% and 6.37% in the flocculation stage. Meanwhile, TP and SS also decreased gradually with time, at the end of reaction, the total removal rates of TP and SS were 59.37% and 68.75%, respectively. The removal rates of TP and SS were 57.34% and 64.45%, which indicated that the ferrite removed TP and SS mainly by flocculation.

The FeO_4^{2-} in water was reduced to Fe (III) from Fe (V), Fe (IV) or other intermediates. The hydrated complexes formed larger network structure, compressing and electrically neutralizing the water impurity diffusion layer, resulting in flocculation [17], which could effectively promote the settlement of TP and SS. It can be seen from Fig. 5 that most of the COD and TOC removed by the oxidation stage while most of the TP and SS were removed through the flocculation stage.

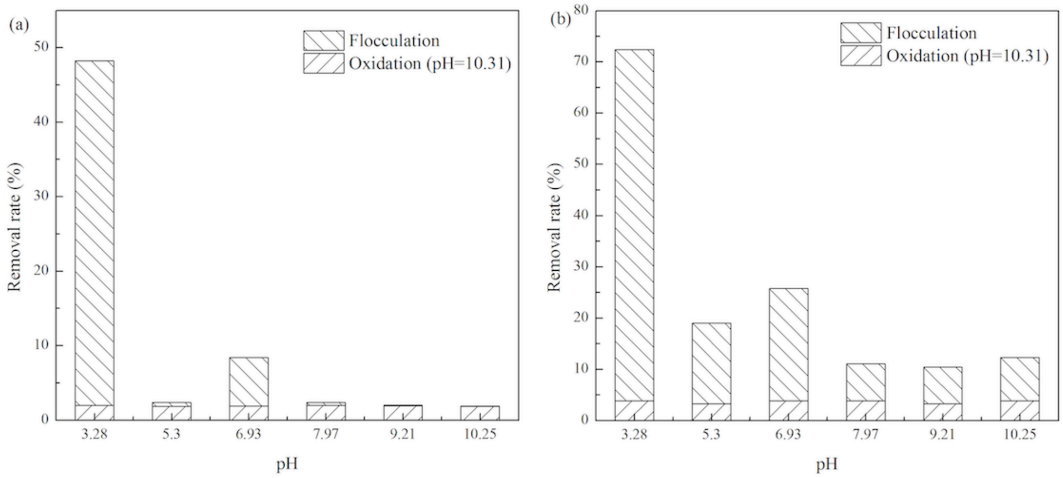

Fig. 4. Removal rate of (a) TP and (b) SS at different flocculation pH (ferrate(VI) dose of 40 mg/L and flocculation time of 15 min), respectively.

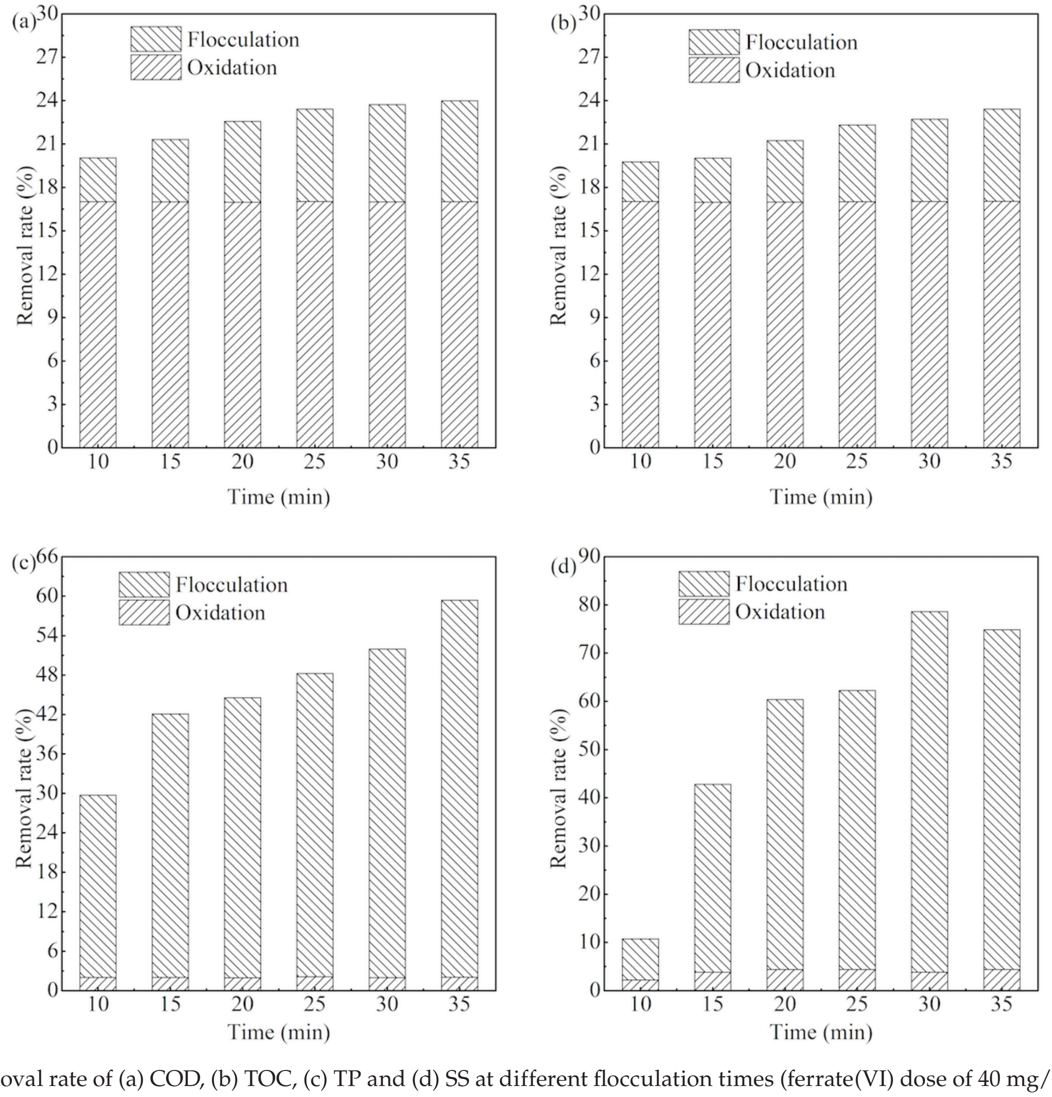

Fig. 5. Removal rate of (a) COD, (b) TOC, (c) TP and (d) SS at different flocculation times (ferrate(VI) dose of 40 mg/L), respectively.

3.3. Treatment of secondary effluent by potassium ferrate

3.3.1. Removal of organic matters

Previous studies [27] have shown that oxidation and flocculation can both remove organic matter by direct oxidizing or removing SS. Potassium ferrate could play both roles in water treatment. The effects of concentrations of potassium ferrate and reaction time are shown in Figs. 6 and 7.

It can be seen from Fig. 6 that when the concentration of potassium ferrate was 20 mg/L, the removal rates of COD, TOC and UV_{254} were 35.61%, 37.80%, and 22.43%, respectively, when the removal of organic matter was better than the advanced treatment of Ever bright WWTP (shown in Table 2). When the concentration increased to 60 mg/L, the removal rates were 44.25%, 45.82%, and 52.34%, respectively, which were the largest. In addition, Ferrate (VI) efficiently removed humus and conjugate structures as shown in Fig. 6. When the concentration was 60 mg/L, the removal rate was up to 23.80%. Decreasing trends of COD and TOC are shown in Fig. 7, indicating that longer time promoted the removal of organic matter by potassium ferrate. In the oxidation stage (the first 25 min), the removal rates of COD and TOC were up to 40.27% and 32.02%, respectively. In the flocculation stage (25–55 min), the removal rate was 9.28% and13.14%, respectively. For organic matter, oxidation stage was more important than flocculation. After 45 min, both removal rates were relatively stable. As can be seen from Fig. 7 that UV_{254} and SUVA decreased gradually with time in the oxidation stage (0–25 min). The highest removal rates were 31.67% and 3.16%, respectively, indicating that potassium ferrate can effectively remove humus substances in sewage.

3.3.2. Removal of TP and SS

As shown in Fig. 8, potassium ferrite had a significant effect on removing TP and SS from sewage.

As the increase of potassium ferrate concentration and reaction time, the removal rates of TP and SS were improved. The treatment effect was better than the actual advanced treatment in Ever bright WWTP (shown in Table 2). Based on the result of section 3.1.2, potassium ferrate removed most of the TP and SS through the flocculation stage. As shown in Fig. 8b, the removal trend of TP and SS in the flocculation stage was more significant than in the oxidation stage. In the meantime, higher concentration of potassium ferrate resulted the greater removal rate of TP and SS. Therefore, application of potassium ferrate can effectively reduce the possibility of water eutrophication caused by effluent in wastewater treatment.

Generally, the concentration of potassium ferrate between 50–60 mg/L, the most removal of pollutants (COD, TOC, TP and SS)were obtained without changing the original pH of these secondary effluent. Considering both the economic availability of purchasing potassium ferrate and the removal effect of pollutants, it is suggested that the optimum concentration of potassium ferrate should be 40–50 mg/L for a sewage treatment plant. Based on result of section 3.2.2 that most of the COD and TOC was removed during the oxidation stage while most of the TP and SS was removed during the flocculation stage,as shown in Figs. 7 and 8b, 25 min of oxidation time and 30 min of flocculation time were suggested for practical usage to maximize the removal of COD, TOC, TP, and SS.

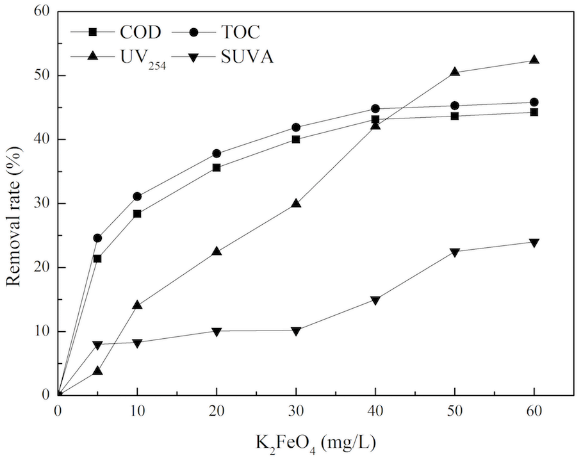

Fig. 6. Removal rate of COD, TOC, UV_{254} and SUVA with different ferrate (VI) dose (pH 6.76, oxidation times of 1 min and flocculation times of 15 min).

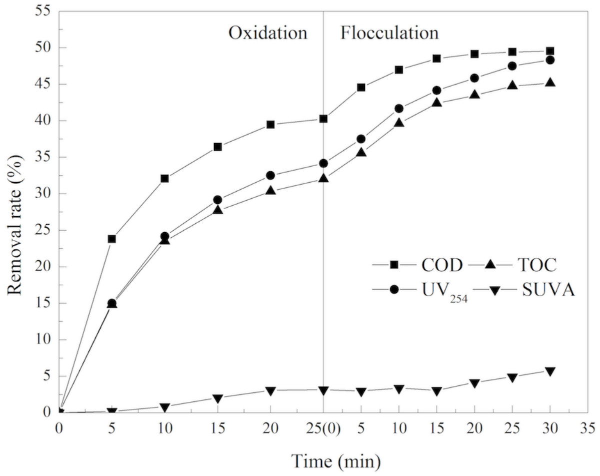

Fig. 7. Removal rate of COD, TOC, UV_{254} and SUVA at different treatment times (pH 10.32 and ferrate (VI) dose of 40 mg/L).

4. Conclusions

Potassium ferrate could be used to remove water pollutants, resulting from the combination effect of oxidation and flocculation, both of which could remove part of organic matters while the removal of TP and SS mainly relied on flocculation. Potassium ferrate concentration, pH, and reaction time could affect the treatments. The optimum treatment conditions in lab experiments were as follows: The potassium ferrate concentration of 40 mg/L, oxidation time of 20 min (pH of 10.31), and flocculation time of 30 min (pH of 3.28) except TP and SS removals. For practical water treatment plant, it is recommended to use potassium ferrate of 40–50 mg/L, 25 min of oxidation, 30 min of flocculation.

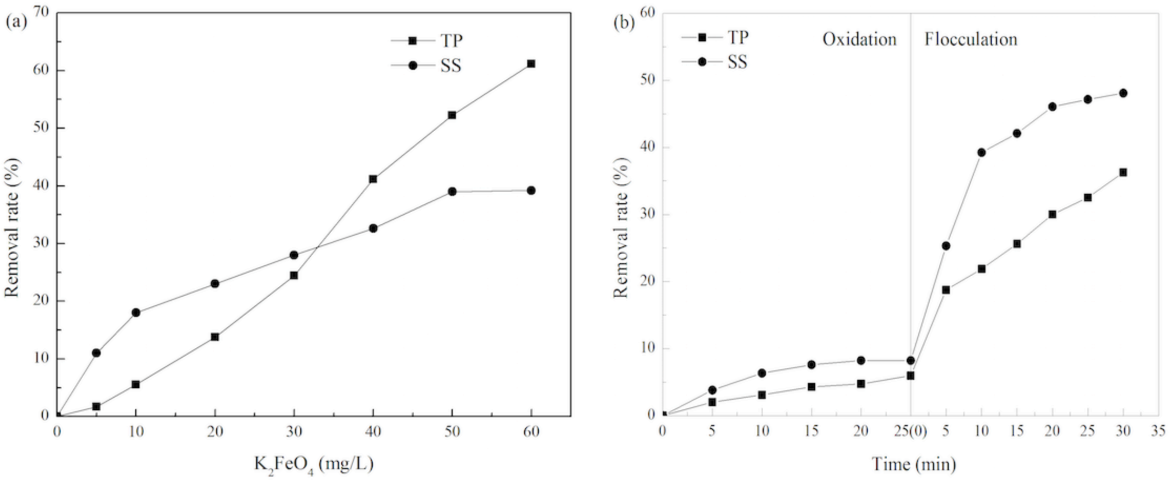

Fig. 8. Removal rate of TP and SS with (a) different ferrate (VI) dose (pH 6.76, oxidation times of 1 min and flocculation times of 15 min) and (b) different treatment time (pH 10.32 and ferrate (VI) dose of 40 mg/L), respectively.

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