



## Application of potassium permanganate and diatomite in enhanced coagulation treatment of a black-odour water body

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### ABSTRACT

In this paper, a small, closed, black-odour water body with poor fluidity in Jurong City was taken as the research object, and the influencing factors and removal efficiency of enhanced coagulation with potassium permanganate (PP) and diatomite (DE) were evaluated. The quadratic regression models based on response surface methodology (RSM) were established to optimize the dosage of each reagent. The experimental results showed that treatment of the black-odour water body by enhanced coagulation combining PP and DE had a significantly greater effect than conventional coagulation with only the addition of polyaluminium chloride (PAC). The optimum pre-oxidation time of PP was 5 min, the optimum dosages of PAC, PP and DE were 34.047, 2.005 and 103.004 mg/L, respectively, and the removal rates of ammonia nitrogen ( $\text{NH}_4^+\text{-N}$ ), the chemical oxygen demand (COD) and the UV absorbance ( $\text{UV}_{254}$ ) reached 72.824%, 76.599% and 86.927%, respectively. The actual error between the experimental results and the values predicted by the models was less than 5%, indicating that the models can predict the actual effects of the combined use of PP and DE in the treatment of black-odour water well as well as guide engineering applications and reduce chemical costs.

**Keywords:** Black-odour water body; Response surface methodology; Potassium permanganate; Diatomite; Enhanced coagulation

### 1. Introduction

Black-odour water bodies have become a prominent problem in aquatic environments, due to their complex causes and numerous influencing factors, they present a difficult challenge for water pollution control and restoration. Currently, domestic and foreign studies on black-odour water bodies are mainly focused on the causes of the black colour and odour in water [1], the evaluation of these characteristics [2–4], the regulation and management of these water bodies [5], and so on. However, there are relatively few studies on emergency prevention and restoration measures for heavily polluted black-odour water bodies.

Purification technologies for black-odour water bodies mainly involve artificial aeration, coagulating sedimentation, etc. Artificial aeration technology requires a large amount of electricity and is not conducive to the deposition of suspended solids. However, coagulating sedimentation technology, which is widely used in water treatment because of its simplicity and high efficiency, uses coagulants to flocculate colloids and suspended particles to form heavy sediments, which are then removed to purify the water.

A commonly used coagulant is polyaluminium chloride (PAC), which has the advantages of good flocculation formation, strong adaptability, low corrosiveness and simple operation. However, it works poorly when used alone; the dosage

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must be large, so the cost is high and secondary pollution occurs easily.

As a strong oxidant, potassium permanganate (PP) is widely used to improve the conventional coagulation process because it does not produce toxic or harmful by-products during the oxidation process. Additionally, the degraded organic compounds and reducing products are easily separated from the solution.

However, most studies on PP-enhanced coagulation by both domestic and foreign scholars have involved powdered activated carbon, which has a low density, so the sediment is not easily flocculated.

In this paper, a mineral material, diatomite (DE), was selected as the coagulant aid because it has a porous structure, a large specific surface area, strong adsorption and low production cost. Compared with powdered activated carbon, its density and particle size are larger; its settling property is superior; and it is also a widely used filter medium [6–8]. Studies on the treatment of micro-polluted water with DE have been reported [9–13], but there has been no report on the combined use of DE and PP to effectively enhance coagulation treatment of black-odour water bodies even though DE has potential applications in enhanced coagulation.

Based on a small, closed, black-odour water body with poor fluidity in Jurong City, this paper discusses the optimal conditions and effects of combining PP and DE to enhance PAC coagulation treatment to prevent the seasonal appearance of black colour and odour in water. This study provides the basis and a reference for engineering applications and related research.

Because three kinds of chemicals are involved in enhancing PAC coagulation process with PP and DE and the interactions between the chemicals are nonlinear, the influencing factors are complex. Optimizing the experimental parameters and balancing the relationship between the chemicals are particularly important for reducing the cost, which requires an accurate and reasonable experimental design. Response surface methodology (RSM) is an effective experimental design for finding the optimal combination conditions, reducing production costs and solving the related problems of nonlinear data processing. Compared with orthogonal design, RSM has the advantages of reduced test times, shorter test periods, higher precision, and higher predictability of experimental results, and it also provides an intuitive 3D stereogram that can illustrate the interactions between factors. The most commonly used software for RSM is Design Expert, and the Box-Behnken Design (BBD) in the software is the most commonly used experimental design for RSM. At present, research on

optimizing the dosage of each reagent in enhanced coagulation by RSM has not been reported in the literature, so based on single-factor experiments, the influence of the interactions between reagents on the treatment of black-odour water was studied using the BBD module in Design Expert software. The prediction models were established, and the optimal technique parameters were determined.

## 2. Materials and methods

### 2.1. Sampling

The test water came from a small, closed water body characterized by poor fluidity, black water quality and unpleasant odour, i.e., a typical black-odour water body. Considering the pollution characteristics and the important factors affecting the blackness of the water body, the removal rates of the chemical oxygen demand (COD), the UV absorbance ( $UV_{254}$ ), the ammonia nitrogen ( $NH_4^+-N$ ) and the turbidity were selected to indicate the effectiveness of water body remediation. The water quality testing and monitoring methods are shown in Table 1.

### 2.2. Experimental methods

The technical process of enhanced coagulation by combining PP with DE is shown in Fig. 1.

The process was simulated by a static experiment that involved stirring samples in a beaker, specifically, six water samples in six 1,000 mL beakers. Each beaker was numbered and placed on a six-speed electric mixer, and according to the coagulation treatment process shown in Fig. 1, different amounts of coagulant and coagulant aid were added to each beaker. The mixing stage involved rapid stirring at a speed of 300 rpm for 1 min, while the flocculation stage involved stirring at a medium-high speed of 150 rpm for 2 min and then at a low speed of 50 rpm for 5 min. After 20 min of static settlement, the supernatant was removed from the six beakers to determine the indexes of COD,  $NH_4^+-N$ , turbidity and  $UV_{254}$  and their average values were obtained from three parallel determinations.

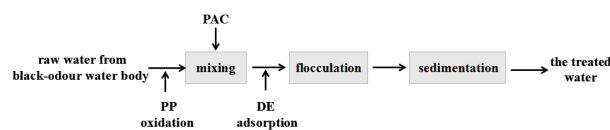


Fig. 1. Static experiment technical process.

Table 1  
Water quality testing and monitoring methods

Index	COD (mg/L)	$NH_4^+-N$ (mg/L)	Turbidity (NTU)	$UV_{254}$ ( $cm^{-1}$ )
Scope	58–86	0.43–4.95	10.81–154.10	0.091–0.209
Average value	72	3.06	60.27	0.135
Monitoring method	Potassium dichromate titration	Nessler reagent colorimetry	Light-scattering method	Microporous-membrane filtration
Instrumentation	JH-12 COD constant-temperature heater	7230G visible spectrophotometer	WGZ-200 turbidimeter	752 ultraviolet-visible spectrophotometer

3. Results and discussion

3.1. Determination of PP oxidation time and DE adsorption time

Previous research results [14,15] were referenced to select the PP pre-oxidation time.

The same amounts of PAC and PP were added to six 1,000 mL water samples at different time points. The effluent water treated by enhanced coagulation was monitored, and the removal effects on each index are shown in Fig. 2.

The six different oxidation times were as follows: (1) simultaneous addition of PAC and PP, (2) pre-oxidation of PP for 5 min before PAC addition, (3) pre-oxidation of PP for 10 min before PAC addition, pre-oxidation of PP for 15 min before PAC addition, (5) pre-oxidation of PP for 20 min before PAC addition, and (6) oxidation of PP for 10 min after PAC addition.

The results shown in Fig. 2 indicate that longer PP pre-oxidation times did not lead to better treatment effects, but the pre-oxidation effect of PP was better before PAC addition than after. Considering the removal effect on each index, the optimum PP pre-oxidation time was 5 min. However, the turbidity removal rate was higher under each process, and the effluent was clear.

To ensure sufficient time for adsorption, DE was added after rapid stirring (mixing stage) to further coagulate flocs and prevent them from being wrapped [16,17]. Therefore, the single-factor test for the DE addition time was not carried out.

3.2. Determination of the central PAC, PP and DE dosage values

The factors influencing enhanced coagulation are complex, the coagulant and coagulant aid dosages have a particularly strong effect on enhanced coagulation and the chemical costs in engineering applications. Single-factor experiments were performed to determine the central dosage values of PAC, PP and DE.

3.2.1. Determination of the central PAC dosage value

By adding different amounts of PAC to six 1,000 mL water samples, the conventional coagulation treatment was carried out according to the experimental methods described in 2.2, and the changes in the removal rates of each indicator are shown in Fig. 3.

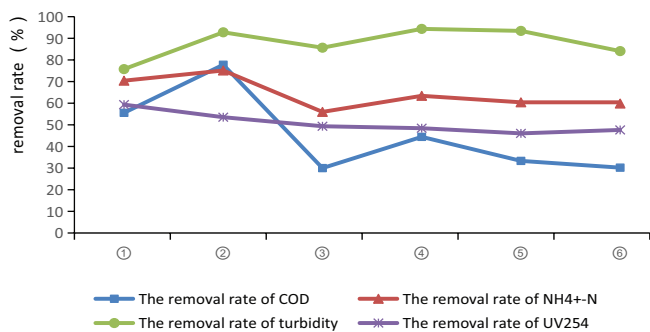


Fig. 2. Enhanced coagulation effect of PP at different oxidation times.

When the PAC dosage was increased, the removal rate of each index tended to first increase and then decrease. Considering the removal effect of each index, the coagulation treatment was more effective when the PAC dosage was 40 mg/L, after which the removal efficiency of COD, NH<sub>4</sub><sup>+</sup>-N and UV<sub>254</sub> decreased with increasing dosage. This reduction in efficiency occurs because colloidal particles in the water adsorb too many counterions, converting the original negative charges into positive ones and increasing the repulsive force, leading to the re-stability phenomenon [18,19]. Therefore, the PAC dosage of 30 to 50 mg/L was chosen as the optimal response surface range in subsequent multi-factor interaction experiments.

3.2.2. Determination of the central PP dosage value

By adding the optimum quantity (40 mg/L) of PAC and different amounts of PP to six 1,000 mL water samples, PP enhanced coagulation treatment was carried out according to the same experimental methods discussed above. The changes in the removal rates of each indicator are shown in Fig. 4.

When the PP dosage was increased from 0.5 to 3 mg/L, the removal rate obviously increased, but it increased slowly as the dosage continued to increase. Comprehensively, considering the removal effect of each index, the PP dosage of 1–3 mg/L was chosen as the optimal response surface range.

3.2.3. Determination of the central DE dosage value

The same amounts of PAC and PP but different amounts of DE were added to six 1,000 mL water samples to determine

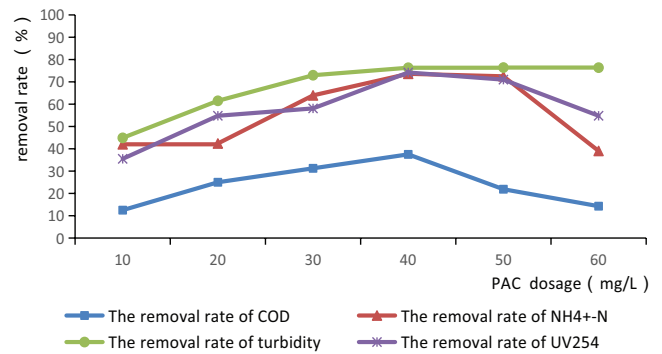


Fig. 3. Coagulation treatment effect of different PAC dosages.

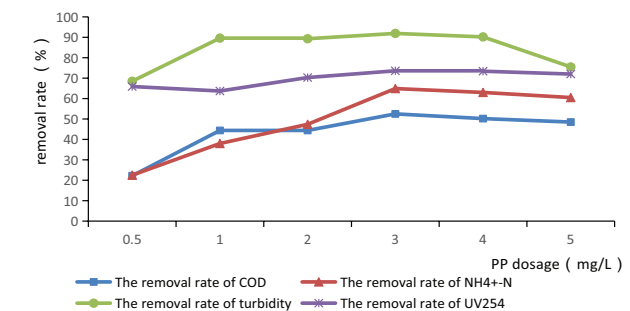


Fig. 4. Enhanced coagulation treatment effect of different PP dosages.

the change in removal efficiency with increasing DE dosage. The results are shown in Fig. 5.

Enhanced PAC coagulation treatment with PP and DE, the removal efficiency of turbidity and UV<sub>254</sub> were significantly higher than those using conventional coagulation treatments. The removal rates of COD and NH<sub>4</sub><sup>+</sup>-N increased with DE dosage, but at 100 mg/L, both removal rates reached their maxima. As the dosage continued to increase, the removal rate decreased slightly due to adsorption equilibrium, so the DE dosage of 80–120 mg/L was chosen as the optimal response surface range.

3.3. Enhanced coagulation experiments of response surface optimization

3.3.1. Response surface methodology

Single-factor experiments have some limitations and cannot reflect the interactions between factors, so an RSM with a short experimental period and high precision was adopted to optimize the conditions of the combined “PAC +

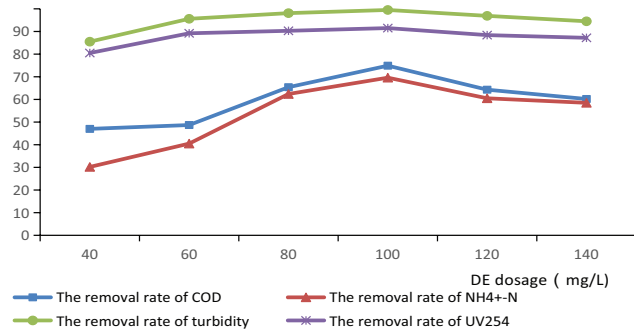


Fig. 5. Enhanced coagulation treatment effect of different DE dosages.

PP + DE” enhanced coagulation in this study [20–23]. Based on the single-factor experiments above and according to the central composite design principle, a total of 17 groups of experimental combinations with three factors and three levels were designed using the BBD module in Design Expert software to determine the best combined process for water quality improvement. The experimental design of the response surface optimization is shown in Table 2.

3.3.2. Experimental results of RSM and analysis of variance (ANOVA)

The RSM experimental results are shown in Table 3. The experimental data were analysed with the help of Design Expert software.

Because the turbidity removal rate was near the saturation value, only ANOVA was performed for the quadratic regression models of the NH<sub>4</sub><sup>+</sup>-N, COD and UV<sub>254</sub> removal rates.

$$Y_{\text{NH}_4^+-\text{N}} = 76.68 - 0.12 \times A + 0.40 \times B + 2.82 \times C + 6.12 \times AB + 0.72 \times AC + 2.72 \times BC - 3.80 \times A^2 - 9.75 \times B^2 - 5.00 \times C^2 \quad (\text{Model 1})$$

Table 2 Three factors and three levels of the RSM experiments

Factor (mg/L)	Code	Level		
		-1	0	1
PP dosage	A	1	2	3
DE dosage	B	80	100	120
PAC dosage	C	30	40	50

Table 3 RSM experimental results

Experiment number	Variable code values			Response values (removal rate/%)			
	A	B	C	NH <sub>4</sub> <sup>+</sup> -N	Turbidity	COD	UV <sub>254</sub>
1	0	1	-1	57.2	98.1	73.5	78.9
2	-1	0	-1	62.5	99.2	73.8	83.1
3	0	0	0	77.5	99.5	79.0	91.8
4	-1	1	0	60.5	99.2	72.1	86.3
5	1	0	-1	65.4	99.2	56.8	81.3
6	0	-1	-1	63.2	99.2	58.4	82.4
7	-1	0	1	68.9	98.7	68.5	90.1
8	0	1	1	66.1	99.2	67.3	90.8
9	0	0	0	77.0	99.5	78.5	89.3
10	1	0	1	74.7	99.4	63.8	91.2
11	0	0	0	77.0	99.6	76.9	92.1
12	0	-1	1	61.2	99.1	66.5	91.5
13	0	0	0	76.4	99.8	77.8	92.3
14	1	1	0	67.9	99.3	50.8	89.5
15	0	0	0	75.5	99.6	78.1	91.9
16	1	-1	0	53.5	99.2	65.4	90.2
17	-1	-1	0	70.6	99.1	62.3	90.5

$$Y_{\text{COD}} = 78.06 - 4.99 \times A + 1.39 \times B + 0.45 \times C - 6.10 \times AB + 3.07 \times AC - 3.58 \times BC - 8.06 \times A^2 - 7.36 \times B^2 - 4.28 \times C^2$$

(Model 2)

$$Y_{\text{UV}_{254}} = 91.48 + 0.28 \times A - 1.14 \times B + 4.74 \times C + 0.87 \times AB + 0.73 \times AC + 0.70 \times BC - 0.92 \times A^2 - 1.44 \times B^2 - 4.14 \times C^2$$

(Model 3)

The ANOVA results for the above models are shown in Table 4.

Table 4 shows that the  $F$  values of the three models of  $\text{NH}_4^+\text{-N}$ , COD and  $\text{UV}_{254}$  removal (model 1, model 2, and model 3) were 11.62, 14.78 and 22.67, respectively; the  $P$  values were all less than 0.05; and the correlation coefficients, namely, the  $R^2$  values, were all greater than 0.9. These results indicated that the models were significant and that the degrees of fit were good.

Table 4  
ANOVA results for the regression models

Source	Sum of squares			DF	F values			P values		
	$\text{NH}_4^+\text{-N}$	COD	$\text{UV}_{254}$		$\text{NH}_4^+\text{-N}$	COD	$\text{UV}_{254}$	$\text{NH}_4^+\text{-N}$	COD	$\text{UV}_{254}$
Model	866.53	1094.22	287.76	9	11.62	14.78	22.67	0.0019	0.0009	0.0002
A	0.13	199.00	0.61	1	0.015	24.19	0.43	0.9057	0.0017	0.5334
B	1.28	15.40	10.35	1	0.15	1.87	7.34	0.7060	0.2135	0.0302
C	63.85	1.62	179.55	1	7.71	0.20	127.33	0.0275	0.6706	<0.0001
AB	150.06	148.84	3.06	1	18.11	18.09	2.17	0.0038	0.0038	0.1840
AC	2.10	37.82	2.10	1	0.25	4.60	1.49	0.6299	0.0692	0.2616
BC	29.70	51.12	1.96	1	3.59	6.21	1.39	0.1002	0.0414	0.2769
$A^2$	60.88	273.19	3.53	1	7.35	33.21	2.50	0.0302	0.0007	0.1579
$B^2$	400.47	227.77	8.73	1	48.34	27.69	6.19	0.0002	0.0012	0.0417
$C^2$	105.37	77.13	72.17	1	12.72	9.38	51.18	0.0091	0.0183	0.0002
Residual	57.99	57.59	9.87	7						
Cor total	924.52	1151.81	297.63	16						

### 3.3.3. Response surface analysis

With the help of Design Expert software, 3D response surface diagrams were obtained to determine the influence of the interacting among three factors on the removal effect of each index as shown in Figs. (6)–(8).

Fig. 6 shows that the effect of the combined use of PP and DE was obviously better than that observed when only PP was added. The contour plots and the curvatures of the response surfaces shown in Fig. 6 indicate that the interaction between the PP and DE dosages had a significant effect on the removal of  $\text{NH}_4^+\text{-N}$  and COD. The  $\text{NH}_4^+\text{-N}$  removal rate first increased and then decreased. The COD removal rate obviously increased with increasing PP and DE dosages and then tended to stabilize. The  $\text{UV}_{254}$  removal rate remained high, with the content in the effluent tending to stabilize after precipitation, and the removal effect was not significantly affected by the interaction of the two coagulant aids.

Because PP has a strong oxidization potential, it can oxidize and decompose organic matter in water, and the

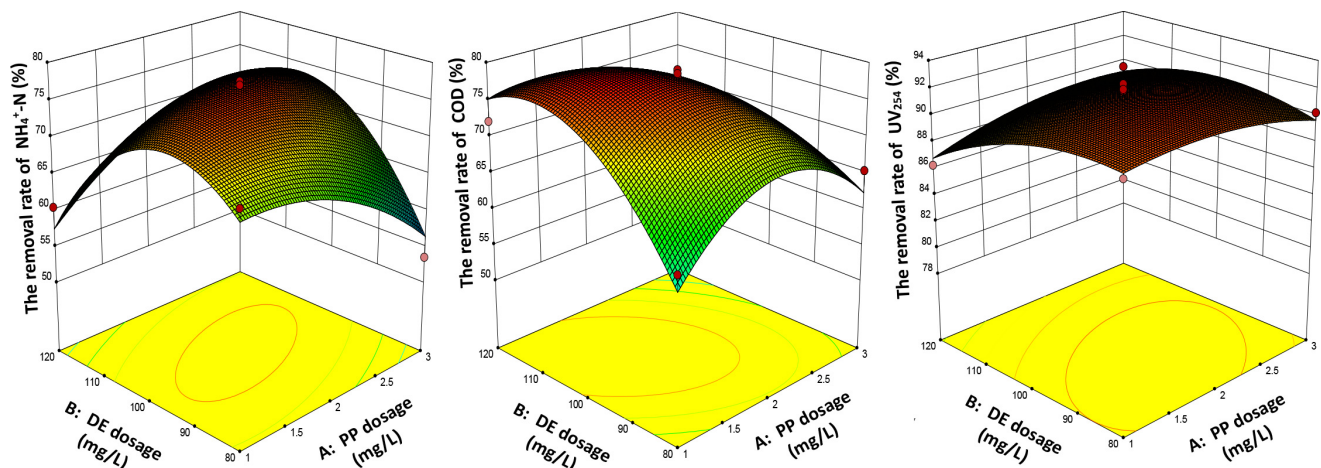


Fig. 6. Response surfaces for the interaction between PP and DE dosages.

intermediate products formed during oxidation, such as manganese dioxide, have the function in adsorption and coagulation [24,25], which can effectively improve the removal rate of organic matter in water. Moreover, DE has a large specific surface area and strong adsorption properties, and a large number of silica hydroxyl groups are distributed in the pore channels that can effectively adsorb organic pollutants in water. However, when the adsorption reached saturation in this study, the effluent tended to stabilize.

The results shown in Fig. 7 indicate that the interaction between the PP and PAC dosages had a significant effect on COD removal but not on  $\text{NH}_4^+\text{-N}$  and  $\text{UV}_{254}$  removal. With an increase in the PAC dosage, the removal rate of each pollutant first increased and then decreased, and when the dosage reached a certain value, the rates tended to stabilize, mainly due to the “re-stabilization” of colloidal particles in water that weakened the coagulation effect. Among them, the  $\text{UV}_{254}$  removal rate increased most rapidly with the increase in PAC dosage, mainly because  $\text{UV}_{254}$  involves humic organic compounds with negatively charged functional groups such as carboxylic groups and hydroxyl groups that can be neutralized by the positive charge produced during the hydrolysis of the polymeric coagulant PAC.

The results shown in Fig. 8 indicate that the interaction between the DE and PAC dosages had a significant effect on COD and  $\text{NH}_4^+\text{-N}$  removal but not on  $\text{UV}_{254}$  removal, which was consistent with the ANOVA results.

The significant degree of influence on the removal effect of the three indexes can be seen in Figs. 6–8 and in the *F* values in Table 4. The order of the effect on  $\text{NH}_4^+\text{-N}$  removal was  $\text{PAC} > \text{DE} > \text{PP}$ ; the order of the effect on COD was  $\text{PP} > \text{DE} > \text{PAC}$ ; and the order of the effect on  $\text{UV}_{254}$  was  $\text{PAC} > \text{DE} > \text{PP}$ .

### 3.3.4. Optimum analysis

According to the ANOVA results, the quadratic regression models of the effects of dosages of PAC, DE and PP on the removal rates of  $\text{NH}_4^+\text{-N}$ , COD and  $\text{UV}_{254}$  fit the results well. Keeping other experimental parameters unchanged and considering the removal rates of  $\text{NH}_4^+\text{-N}$ , COD and  $\text{UV}_{254}$ , the PAC, PP and DE dosages were optimized using Design Expert software to obtain the lowest chemical cost for engineering applications. To verify the accuracy of the model predictions, the experiment was carried out with the optimized dosage of each reagent and the average value of three parallel experiments was obtained, as shown in Table 5.

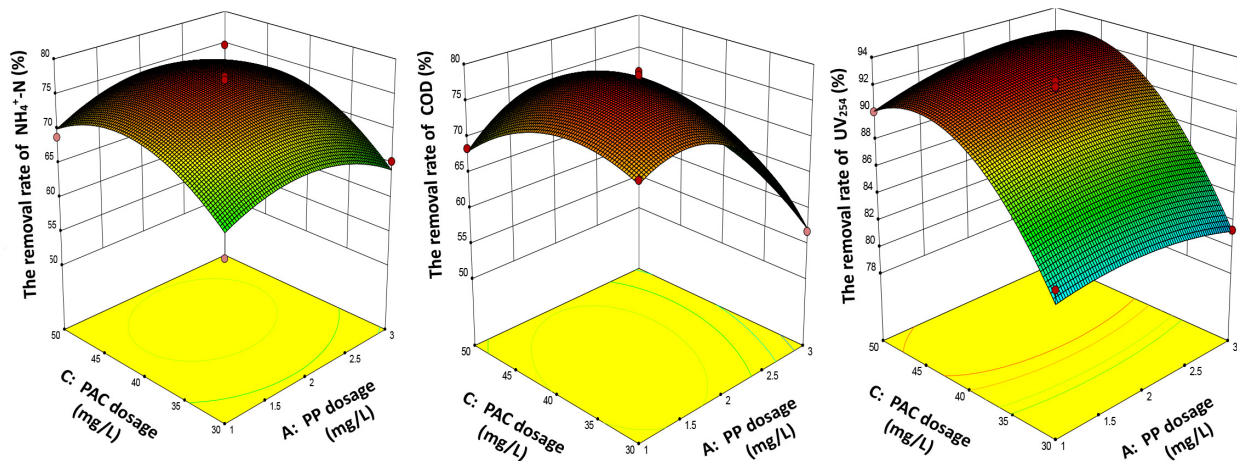


Fig. 7. Response surfaces for the interaction between PAC and PP dosages.

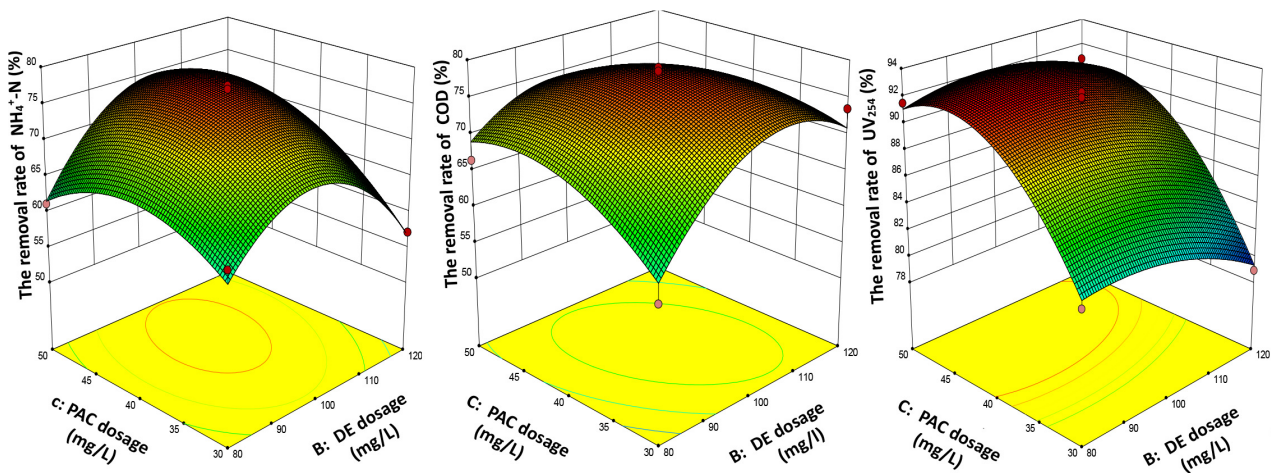


Fig. 8. Response surfaces for the interaction between PAC and DE dosages.

Table 5  
Model prediction and experimental results under optimum conditions

Model prediction and experimental verification	Dosage (mg/L)			Removal rate (%)		
	PAC	PP	DE	NH <sub>4</sub> <sup>+</sup> -N	COD	UV <sub>254</sub>
Model prediction	34.047	2.005	103.004	72.824	76.599	86.927
Experimental verification	34	2	103	69.5	73.8	84.0

Table 5 shows that the actual error between the experimental results and the predicted values was less than 5%, which indicated that the models could better predict the actual effects of the combined use of PP, DE and PAC in the treatment of black-odour water bodies. Therefore, it will be of great practical significance for improving the treatment effect and reducing chemical costs.

### 3.3.5. Cost estimation

The enhanced coagulation technique for the treatment of black-odour water by combining PP and DE does not require additional water treatment structures, so the cost of the chemicals will greatly impact on the cost of operation. In this study, the dosage of each reagent was optimized using RSM, and according to the results, the optimal dosages of PAC, PP and DE in each m<sup>3</sup> of water were 34.047, 2.005 and 103.004 g, respectively. According to the market prices of PAC, PP and DE, which are \$2,500, \$24,500 and \$1,800/ton, respectively, the cost of chemicals/m<sup>3</sup> of water is only approximately 0.32 yuan.

## 4. Conclusions

The removal efficiency of pollutants in a black-odour water body by enhanced PAC coagulation treatment with PP and DE was obviously better than that by conventional coagulation treatment.

The PP pre-oxidation time and the central values of three factors (PAC, PP, and DE dosages) were determined by single-factor experiments, and based on the results, quadratic response surface models were established using Design Expert software. Since the turbidity removal rate reached saturation, the removal rates of NH<sub>4</sub><sup>+</sup>-N, COD and UV<sub>254</sub> were chosen as the response values of the models. The *P* value of each regression model was less than 0.05, and the correlation coefficient, *R*<sup>2</sup>, was greater than 0.9. These results indicated that the models were significant; the degree of fit was better; and the models could analyze and predict the effects on pollutant removal well.

The order of the effect of three factors on the NH<sub>4</sub><sup>+</sup>-N removal efficiency was PAC > DE > PP; the order of the effect on COD was PP > DE > PAC; and the order of the effect on UV<sub>254</sub> was PAC > DE > PP.

The dosages optimized by Design Expert software were 34.047 mg/L PAC, 2.005 mg/L PP, and 103.004 mg/L DE, and the removal rates of NH<sub>4</sub><sup>+</sup>-N, COD and UV<sub>254</sub> reached 72.824%, 76.599%, and 86.927%, respectively. The results of the verification experiment yielded an error of less than 5% from the predicted value, which indicated that the models could better predict the actual effects of the combined use of PP, DE and PAC in the treatment of black-odour water bodies

and will prove to be of great practical significance for guiding engineering applications.

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