

Original Research

Multivariate Parameter Optimization of PAM Wastewater Treatment by Ti/Ir–Ta Electrocatalysis via the Box-Behnken Design

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Received: 6 May 2021

Accepted: 17 July 2021

Abstract

In this study, the electrocatalysis method is used to degrade polyacrylamide (PAM) from wastewater. This method can degrade PAM rapidly and effectively, and it has no secondary pollution but has clean and environmental protection. The Box–Behnken design of response surfaces is used to optimize the PAM degradation experiment of wastewater. In accordance with the regression analysis results, the second-order polynomial fitting model is significant. The best conditions for the experiment are a current density of 58.8 mA/cm², an electrode plate spacing of 3.07 cm, initial pH of the solution of 6.73 and reaction time of 2.225 h. The COD removal rate reaches 92.7%, and the predicted value is 92.24%. Therefore, electrocatalytic degradation of PAM has a good application prospect.

Keywords: electrocatalysis, PAM, Box-Behnken design, response surface method, COD removal efficiency

Introduction

Polyacrylamide (PAM) has the functions of flocculation, adhesion, resistance reduction and thickening. PAM is used not only in water treatment but also in printing and dyeing, papermaking and oil exploitation industries [1]. In the process of oilfield energy development, the injection of PAM solution can improve the oil–water velocity of flow ratio, increase the viscosity of driving fluid, reduce the permeability of formation water and improve oil recovery [2-3]. However, the discharge of water containing PAM to the environment will lead to many environmental problems

and cause many health issues for human beings and potential harm to the ecosystem. The viscosity of wastewater containing PAM is relatively high, which increases the ability of the wastewater to carry suspended solids, thus making the suspended solids in water exceed the standard [4]. PAM is beneficial to the growth of sulphate-reducing bacteria in water and has a strong corrosive effect on water treatment facilities. Therefore, how to remove the residual PAM in wastewater effectively has become a key concern in the field of environment. At present, the main methods for treating polymer-containing wastewater are physical treatment [5], biodegradation [6-8], chemical degradation [9-10] and combination method [11]. The use of an electrochemical method for the degradation of wastewater is characterized by many advantages, including flexible operation, simple equipment, clean

and environmental protection, no secondary pollution, fast degradation reaction speed, mild reaction conditions and evident degradation effect of PAM. Nevertheless, the effect of electrochemistry is influenced by electrode materials, operating conditions and other factors. Our team used an aluminium electrode to treat oilfield polymer wastewater [12]. Although good results have been achieved, the further promotion of this technology is limited due to the excessive consumption of electrode plate and secondary pollution in the electrochemical process. By contrast, the electrocatalysis technology uses an insoluble electrode as a material to carry out water treatment research, and the reaction is more thorough and does not produce secondary pollution [13-14]. The purpose of high efficiency and environmental protection degradation of oilfield wastewater can be achieved through the optimisation and improvement research of plate material, current density, plate spacing, reaction time and other factors.

Electrocatalysis is a technology that has achieved good results in the research of organic wastewater, such as phenol [15] and dinitrobenzene [16]. Yang [17] also tried to use an insoluble electrode to degrade PAM wastewater for the first time. Compared with the common chemical flocculation and biodegradation technology, it has the characteristics of short time and high efficiency. However, no systematic study has been conducted on the related parameters. The conventional one-factor-at-a-time approach has been widely used to optimise the effects of various factors to attain the best system performance. This approach determines the influence of various factors on degradation rate, that is, whilst changing one of the factors, the other factors remain unchanged to study the influence of each factor in turn. Accordingly, this approach needs to conduct multibatch experiments, which is a time- and labour-consuming process. It fails to consider any possible interaction amongst the factors. The response surface methodology (RSM) evaluates the influencing factors and their interactions by establishing mathematical models and determines the best experimental parameters. The number of experiments is minimal. The objectives of this study are to optimise the experiment of electrocatalytic oxidation degradation of PAM by using RSM and obtain the maximum response value and corresponding experimental parameters from a mathematical model to realise enhanced experimental results.

Experimental

Material

The solvent used in the experiment is distilled water. In order to simulate PAM wastewater, sodium chloride is added to distilled water as electrolyte, and sodium hydroxide and hydrochloric acid solution are used to adjust the pH value. The reagents used are of analytical

grade. The pH value is measured by pH meter, and the COD of the sample is determined by QCOD-2E rapid analyzer (Changhong Co., Ltd., Shenzhen, Guangdong). A 500 ppm PAM solution was prepared by dissolving 0.50 g PAM in 1000 mL water. The average molecular weight of PAM was 5 million and the degree of hydrolysis was 27%. Use 0-30 V DC regulated power supply (Hong Kong Longwei Instrument Co., Ltd.) for electrolysis.

Experimental Methods

The electrolysis experiment was conducted in a cylindrical glass cell. In this process, the anode was mainly titanium plate, and different elements were coated on the plate surface. The electrodes were Ru-Ir and Ir-Ta (size: 10 cm×12 cm; thickness: 0.2 cm), and the cathode was Ti plate (size: 10 cm×12 cm; thickness: 0.2 cm). The electrolyte (1000 mL) was poured into the experimental electrolyser, and the electrodes were immersed in the electrolyte to achieve the required effective electrolysis area. The electrodes were connected to a DC-regulated power supply with a current range of 0-5 A and a voltage range of 0-30 V to provide current density. All experiments were performed at room temperature (10°C), and the COD value was determined at the end of the experiment.

Experimental Design

The response surface method (RSM) was used for the experimental design, data analysis, and model construction. The aim was to investigate the relationship between COD removal efficiency and four operating parameters (i.e. current density, inter-electrode gap, pH and operation time). After further narrowing the range of experimental parameters, we selected the current density of 40-60 mA/cm², the plate spacing of 2-4 cm, the pH of 6-8 and the reaction time of 1.5-2.5 h. The Box-Behnken design (BBD) was specifically selected as it was more efficient than other RSM designs when there are three or four variables [18]. The four selected process variables, current density, inter-electrode gap, pH and operation time, were defined as X_1 , X_2 , X_3 and X_4 , respectively, COD removal efficiency were defined as Y . Each parameter was coded at three levels, -1 (minimum), 0 (central), and +1 (maximum), covering the entire study range. Table 1 shows the Box-Behnken design matrix. Analysis of variance (ANOVA) was used to analyze the experimental data. The adequacy of the developed models was measured by coefficient of determination (R^2) and adjusted coefficient of determination (R^2_{adj}) analyses. After fitting the models, the generated data were used for 3D response surface optimization. The statistical software Design Expert 8.0 (Stat-Ease Inc., Minneapolis, USA) was applied for the experiment design, analysis and optimisation.

A Box-Behnken design requires an experiment number according to $N = k^2 + k + C_p$, where k is the

Table 1. Experimental range and levels of the independent variables in Box-Behnken design.

Variables, unit	Factors	Ranges and levels		
		-1	0	1
Current density (mA/cm ²)	X_1	40	50	60
Inter-electrode gap (cm)	X_2	2	3	4
pH	X_3	6	7	8
Reaction time (h)	X_4	1.5	2	2.5

factor number and C_p is the replicate number of the central point. Experimental data from the Box-Behnken design can be analyzed and fitted to a second-order model using Design Expert 8.0 software and the following Equation (1):

$$Y = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^{j-1} \sum_{j=1}^k \beta_{ij} X_i X_j + \sum_{i=1}^k \beta_{ii} X_i^2 + e_i \quad (1)$$

...where Y is COD removal efficiency, β_0 is a constant coefficient, X_i and X_j are variables, β_i is the coefficient of response surface (linear term) that determines the influence of parameters on response surface, β_{ij} refers to the effect of the interaction among variables i and j , β_{ii} is a parameter that determines the shape of the curve, k is the number of studied factors, and e_i is the error. The coding value of process parameters can be determined by the following formula (2):

$$X_i = \frac{x_i - x_0}{\Delta x_i} \quad (2)$$

In the above formula, X_i is the dimensionless coded value of the i th independent variable, x_i is the uncoded value of the i th independent variable, x_0 is the uncoded independent variable of the center point, and Δx_i is the step change value from low level (-1) to high level (+1).

Results and Discussion

Effect of Anode Materials

As shown in Fig. 1, with the prolongation of electrolysis time, the degradation effect of PAM by using the two electrodes changed, the concentration of PAM decreased, and the degradation rate increased. After 3 h, the degradation rates of PAM were 80.54% and 88.19%. When the initial concentration was 500 mg/L, the current density was 30 mA/cm², the electrode spacing was 3 cm, and the electrolyte concentration was 5000 mg/L. The degradation rate of the Ru-Ir electrode was 80.54%, and the degradation

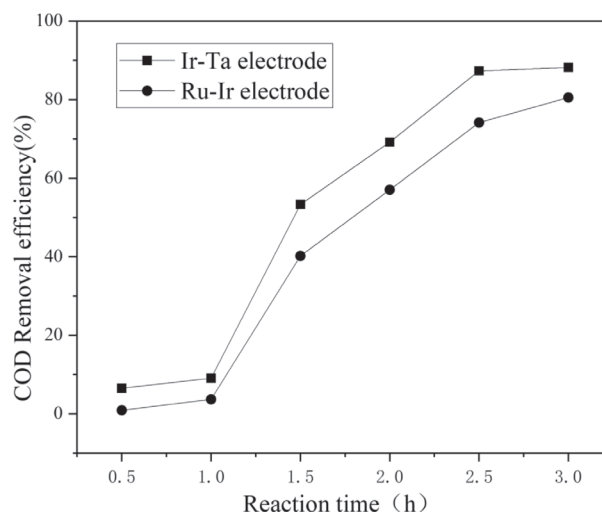


Fig. 1. Effect of two different electrodes on COD removal efficiency.

rate of the Ir-Ta electrode PAM was 88.19% after 3 h of electrolysis. The effect of the Ir-Ta electrode was better at any time of electrolysis. In the electrolysis process, the electrolyte solution could invade into the electrodes through the surface cracks, such that the electrode plates were slowly oxidised; in particular, the Ru-Ir electrode plate was corroded, thus affecting the electrolysis effect of the electrode [19]. Hwang [20] determined that Ta is an inert oxide. Doping Ta oxide in an electrode can improve the durability and stability of Ir, delay the loss of the electrode under acidic conditions and enhance the ability of the anode to resist oxygen passivation. The results showed that the degradation rate of PAM was higher when the Ir-Ta electrode was used as anode but lower when the Ru-Ir anode was used as anode. Therefore, in the next experiment, the Ir-Ta electrode were chose as anode to study the best reaction conditions.

Effect of Current Density

The degradation efficiency of PAM is related to the electron transfer capability and generation capability of $\cdot\text{OH}$ and other active free radicals. The electron transfer capability and generation capability of free radicals depend on the applied current density [21]. The higher the current density is, the more charge transfer in the solution will be, that is, the stronger the electron transfer capability will be, and the faster the electrode reaction rate will be. When the current density increases, the electrode potential of the anode increases, the electrode reaction rate accelerates, and substantial active substances, such as $\cdot\text{OH}$ radicals, are produced per unit time. The reaction with PAM accelerates, the concentration of PAM decreases, and the degradation rate of PAM increases. However, excessive current will also increase the energy consumption of the experimental system. This condition will present

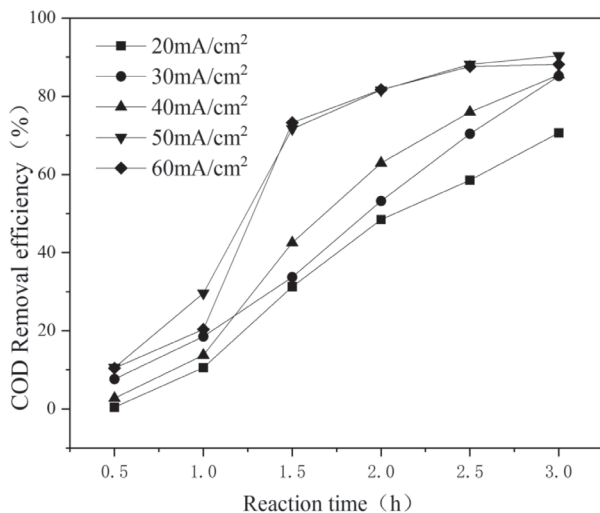


Fig. 2. Effect of current density on COD removal efficiency.

electrolytic water and other side reactions, thus reducing the current efficiency of electrocatalytic oxidation. In Arwa Abdelhay's experiment, the increase in the applied current density from 1.28 mA/cm² to 2.55 mA/cm² enhanced the COD removal efficiency. When the current density was further increased from 2.55 mA/cm² to 3.83 mA/cm², no significant increase in COD removal efficiency occurred. Calculation indicated that when the current density was 1.28 mA/cm², COD in the solution could be removed at the minimum cost [22]. Therefore, the current densities of 20, 30, 40, 50 and 60 mA/cm² were designed. In accordance with Fig. 2, when the current density reached 50 mA/cm², the degradation rate of PAM increased slowly, and the trend was unobvious. When the current density was 50 mA/cm², the degradation rate of PAM was 90.36%.

Effect of Inter-Electrode Gap

The distance between the two electrodes would affect the electric field strength of the solution, which will affect the degradation rate of PAM. When the plate spacing decreased, the electric field intensity and current density, the force between the electrodes and the conductivity increased. As a result, the mass transfer effect of the solution was increased, the migration rate of charged ions was accelerated, and the generation capability of active free radicals (such as $\cdot\text{OH}$ and $\text{O}_2^{\cdot-}$) in the solution was enhanced; the reaction with PAM accelerated, thus improving the removal efficiency of COD in water [23]. Nonetheless, an excessively small inter-electrode gap is un conducive to the electrochemical catalytic oxidation reaction because the solution between the two plates is difficult to flow and exchange with the solution outside the plate. This condition will lead to uneven solution mixing, resulting in a concentration difference and a waste of electric energy. Therefore, in this experiment, the inter-electrode gap was adjusted from 1 cm to 5 cm.

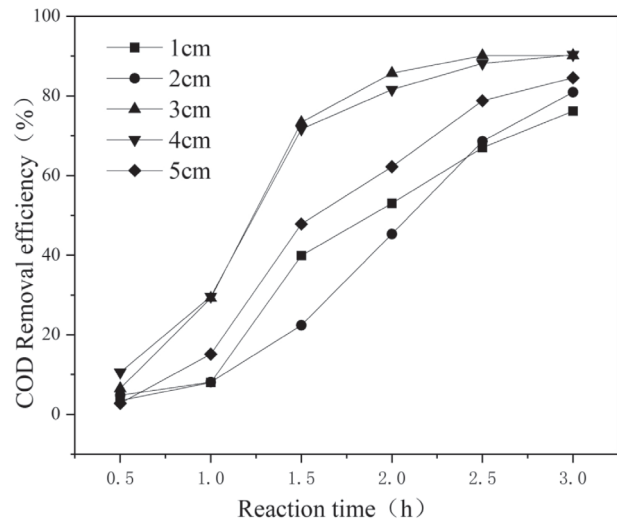


Fig. 3. Effect of inter-electrode gap on COD removal efficiency.

The other operating parameters were as follows: current density = 30 mA/cm², electrolyte concentration and initial pH = 7. As shown in Fig. 3, with the increase in plate spacing, the COD removal efficiency firstly increased and then decreased. The degradation rates of PAM at 1, 2, 3, 4 and 5 cm were 76.20%, 80.95%, 90.21%, 90.36% and 84.57%, respectively. When the inter-electrode gap was 3 cm, the degradation rate of PAM reached the maximum, that is, 3 cm was the best inter-electrode gap. In the subsequent experiments, the inter-electrode gap of 3 cm was selected as an optimum reaction condition.

Effect of pH

In Fig. 4, the degradation rate of PAM firstly increased and then decreased with the increase in pH. Moreover, the degradation rate of PAM under acidic conditions was significantly higher than that under

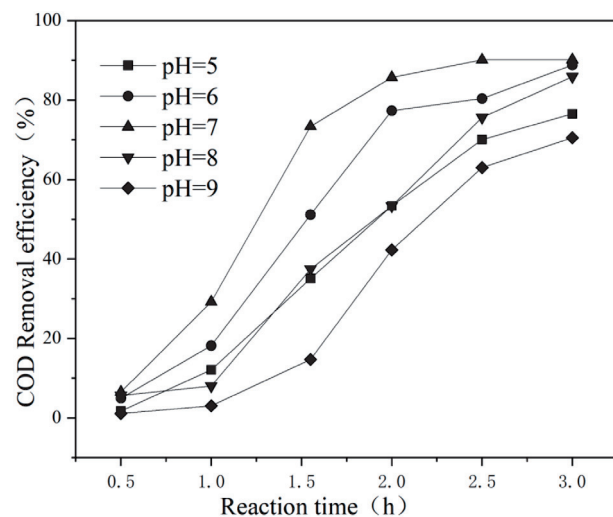


Fig. 4. Effect of pH on COD removal efficiency.

Table 2. Design matrix in coded units and the experimental responses.

Rum number	Current density (X_1)	Inter-electrode gap (X_2)	pH (X_3)	Reaction time (X_4)	COD Removal efficiency (Y , %)
1	0	0	-1	1	80.40
2	-1	0	0	-1	69.20
3	-1	-1	0	0	58.20
4	0	0	0	0	85.60
5	1	0	1	0	52.34
6	0	0	1	1	54.17
7	1	0	0	1	88.24
8	0	0	0	0	83.00
9	0	0	-1	-1	65.20
10	1	0	-1	0	74.30
11	0	0	1	-1	37.20
12	-1	0	-1	0	50.20
13	0	-1	-1	0	22.01
14	-1	0	0	1	70.98
15	0	1	0	-1	70.66
16	-1	1	0	0	59.93
17	0	-1	1	0	39.70
18	0	-1	0	-1	25.50
19	0	1	-1	0	75.20
20	0	1	1	0	49.34
21	1	0	0	-1	73.10
22	-1	0	1	0	44.30
23	0	1	0	1	82.40
24	1	-1	0	0	45.80
25	0	0	0	0	87.70
26	0	-1	0	1	83.40
27	1	1	0	0	83.10

alkaline conditions. Under acidic conditions, the acyl group of PAM in the solution was easy to be converted into a carboxyl group. Such conversion reduced the internal hydrogen bonding amongst molecules, thus increasing the solubility of PAM in water and facilitating the degradation of the solution. In alkaline conditions, the carboxyl group in the solution was hydrolysed to carboxylate ions, and the amide group in PAM combined with carboxylate ions to form a hydrogen bond easily. This phenomenon increased the stability of molecules and was un conducive to the degradation of PAM in the solution. Li et al. [24] found in an experiment of electrolytic petrochemical wastewater that a large amount of adsorbed $\cdot\text{OH}$ was produced at low pH, and the degradation performance would be reduced with the increase in pH. In the

experiment, 1 L of PAM solution with a concentration of 500 mg/L was used for electrolysis, the electrode spacing was 3 cm, the electrolyte concentration was 5000 mg/L, and the current density was 30 mA/cm². The degradation rates of PAM were 76.56%, 88.84%, 90.21%, 85.91% and 70.52% when pH was adjusted to 5, 6, 7, 8 and 9, respectively, with HCl and NaCl. Under alkaline conditions, the degradation rate of PAM was relatively low. Hence, when pH=7, the degradation effect of PAM was highest, and the degradation rate was 90.21%.

ANOVA and Model Fitting

Table 2 shows the experimental results for COD removal efficiency based on the Box-Behnken

Table 3. ANOVA for response function Y and variables selected to fit a model.

Source	Sum of squares	df	Mean square	F -value	p-value Prob> F	Remark
Model	8929.33	14	637.81	9.77	0.0002	S
X_1	342.08	1	342.08	5.24	0.0410	S
X_2	1776.82	1	1776.82	27.22	0.0002	S
X_3	678.91	1	678.91	10.40	0.0073	S
X_4	1174.73	1	1174.73	18.00	0.0011	S
X_1X_2	316.31	1	316.31	4.85	0.0480	S
X_1X_3	64.48	1	64.48	0.99	0.3399	NS
X_1X_4	44.62	1	44.62	0.68	0.4245	NS
X_2X_3	474.15	1	474.15	7.26	0.0195	S
X_2X_4	532.69	1	532.69	8.16	0.0144	S
X_3X_4	0.78	1	0.78	0.012	0.9146	NS
X_1^2	270.75	1	270.75	4.15	0.0644	NS
X_2^2	1439.93	1	1439.93	22.06	0.0005	S
X_3^2	2770.35	1	2770.35	42.45	<0.0001	S
X_4^2	57.38	1	57.38	0.88	0.3669	NS
Residual	783.21	12	65.27			
Lack of fit	772.12	10	77.21	13.93	0.0688	NS
Pure error	11.09	2	5.54			
Cor total	9712.54	26				

design. The obtain the ‘goodness of fit’, ANOVA was used to analyze the data from this study. The COD removal model was significant by the F -test at the 95% confidence level (prob> F <0.05) [25]. The results of the ANOVA of the regression parameters of the predicted response surface quadratic models for COD removal efficiency, with insignificant model terms excluded, are shown in Table 3. The relationships between COD removal efficiency and the four operating parameters (i.e. current density, inter-electrode gap, pH and operation time) were fitted to the second-order polynomial. The Equation (3) is as follows:

$$\begin{aligned}
 Y = & 85.43 + 5.34X_1 + 12.17X_2 - 7.52X_3 + 9.89X_4 + 8.89X_1X_2 \\
 & - 4.01X_1X_3 + 3.34X_1X_4 - 10.89X_2X_3 - 11.54X_2X_4 \\
 & + 0.44X_3X_4 - 7.13X_1^2 - 16.43X_2^2 - 22.79X_3^2 - 3.28X_4^2
 \end{aligned}
 \quad (3)$$

...where Y is the COD removal efficiency (%); X_1 , X_2 , X_3 and X_4 denote the coded values for current density (mA/cm²), inter-electrode gap (cm), pH and reaction time (h), respectively.

In Table 3, the model F -value of 9.77 implies the model is significant. There is only a 0.02% chance that a “model F -value” this large could occur due to noise. Value of “Prob> F ” less than 0.05 indicate model terms

are significant. In this case X_1 , X_2 , X_3 , X_4 , X_1X_2 , X_2X_3 , X_2X_4 , X_2^2 , X_3^2 are significant model terms. The “Lack of Fit F -value” of 13.93 implies there is a 6.88% chance that a “Lack of Fit F -value” this large could occur due to noise.

Fig. 5 shows the normal probability of COD removal efficiency of t -test residual, and the figure

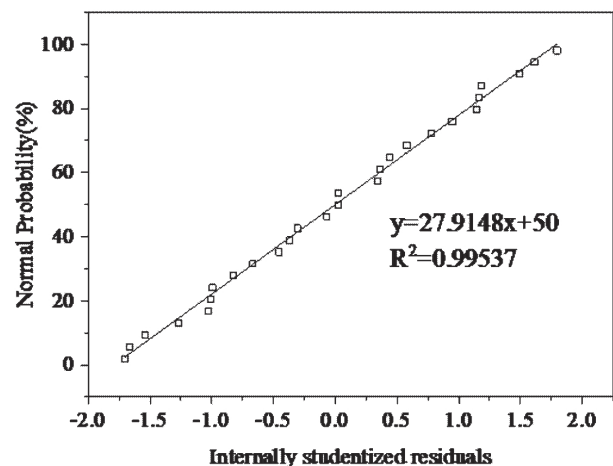


Fig. 5. Normal probability plot of the internally studentised residuals in the response surface model.

is an important diagnostic tool to check whether the model is sufficient. Most of the obtained data points followed a straight line, suggesting that the data were normally distributed in the responses of the model. The determination coefficient R^2 was 0.9954, which indicated the ratio of prediction variables. The value of R^2 was high, implying that the predicted value of response was not different from the actual value. The regression model of the adjusted decision coefficient ($R_{adj}^2 = 0.9253$) was close to 1, which indicated that the experimental value could be significantly predicted.

Interactive Effects Analysis and Optimization

Contour plots of the mathematical regression model were described using Design Expert software to study the interactive relationship between independent variables and response. Fig. 6 (a-f) show the contour plots. Three variables were varied within the experimental range, whilst the other variables were maintained constant.

Fig. 6a) describes the interaction between the current density and inter-electrode gap. When the current

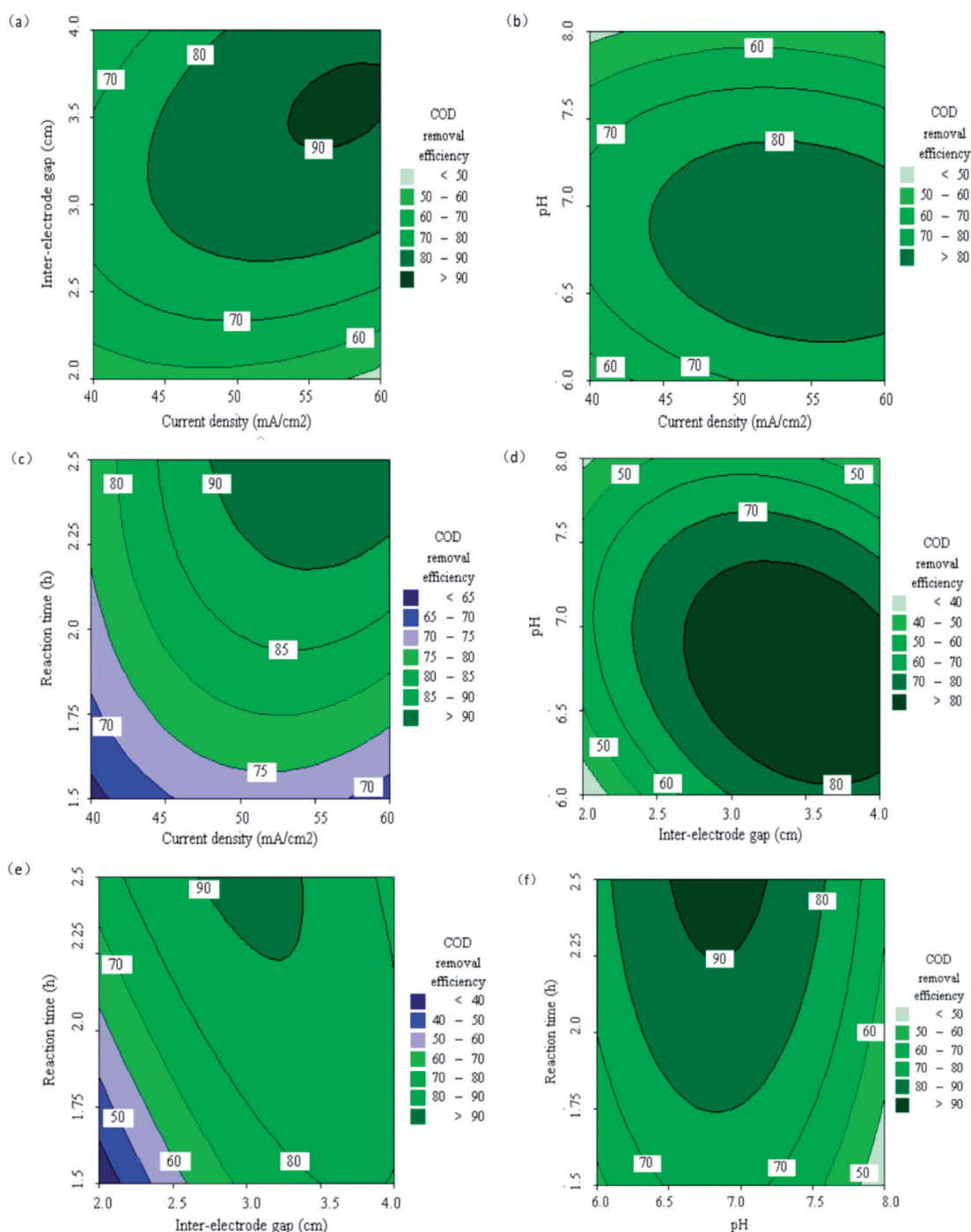


Fig. 6. Response surface plots of interactive effect among factors on (a-f) COD reduction efficiency (%).

Table 4. Verification test of the fitting models.

Test number	Operation parameters				Response value Y (%)		
	X_1	X_2	X_3	X_4	Observed value	Predicted value	Error
1	-0.36	-0.049	0.43	-0.049	75.08	76.16	-1.44
2	0.85	0.71	0.085	0.48	90.28	90.01	0.30
3	0.25	-0.91	0.45	-0.73	38.56	38.22	0.88
4	0.34	-0.23	-0.14	-0.76	70.29	69.73	0.80
5	-0.97	-0.46	0.41	-0.41	59.65	59.23	0.70

density was 40 mA/cm², the COD removal efficiency increased from 53.7% to 73.2% and then gradually decreased to 60.1% with the increase in plate spacing from 2 cm to 4 cm. When the current density was 60 mA/cm², the COD removal efficiency increased from 46.6% to 90.4% and then decreased to 88.4% with the increase in electrode plate spacing from 2 cm to 4 cm.

The interaction between current density and pH is illustrated in Fig. 6b). When the current density was 40 mA/cm², the pH increased from 6 to 8. The plot shows that the COD removal efficiency increased by 19.2% (from 54.2% to 73.4%) and then decreased by 26.3% (from 73.4% to 47.1%). At the current density of 60 mA/cm², the COD removal efficiency increased by 12.5% (from 72.7% to 85.2%) and then decreased by 35.5% (from 85.2% to 49.7%) when the pH increased from 6 to 8.

Fig. 6c) describes the interaction between current density and reaction time. At the reaction time of 1.5 h, the removal efficiency increased by 9.2% (from 63.3% to 72.5%) and then decreased by 5.1% (from 72.5% to 67.4%) when the current density increased from 40 mA/cm² to 60 mA/cm². At the reaction time of 2.5 h, the removal efficiency increased by 18.4% (from 76.3% to 94.7%) and then decreased by 1.0% (from 94.7% to 93.7%) when the current density increased from 40 mA/cm² to 60 mA/cm².

Fig. 6d) describes the interaction between inter-electrode gap and pH. At the inter-electrode gap of 2 cm, the removal efficiency increased by 25.7% (from 31.4% to 57.1%) and then decreased by 19.2% (from 57.1% to 37.9%) when the pH increased from 6 to 8. At the inter-electrode gap of 4 cm, the removal efficiency increased by 8.2% (from 77.0% to 85.2%) and then decreased by 44.7% (from 85.2% to 40.5%) when the pH increased from 6 to 8.

The interaction between reaction time and inter-electrode gap is illustrated in Fig. 6e). When the reaction time was 1.5 h, the inter-electrode gap increased from 2 cm to 4 cm. The plot shows that the COD removal efficiency increased by 48.2% (from 32.7% to 80.9%) and then decreased by 1.2% (from 80.9% to 79.7%). At the reaction time of 2.5 h, the COD removal efficiency increased by 17.0% (from 75.0% to 92.0%) and then decreased by 15.5% (from 92.0% to 76.5%) when the inter-electrode gap increased from 2 to 4 cm.

Fig. 6f) describes the interaction between reaction time and pH. At the reaction time of 1.5 h, the COD removal efficiency increased by 15.3% (from 57.8% to 73.1%) and then decreased by 30.9% (from 73.1% to 42.2%) when the pH increased from 6 to 8. At the reaction time of 2.5 h, the COD removal efficiency increased by 16.0% (from 76.6% to 92.6%) and then decreased by 29.9% (from 92.6% to 62.7%) when the pH increased from 6 to 8.

Optimisation and Validation Experiment

The desirable point prediction function in the experimental design was applied to identify the optimum conditions to maximise COD removal efficiency within the studied experimental ranges. The optimal results for COD reduction were predicted to occur under the following conditions: a current density of 58.8 mA/cm², an inter-electrode gap of 3 cm, pH of 6.73 and reaction time of 2.23 h. The actual value of COD removal efficiency was 92.70% , Compared with the team degradation of PAM by electro flocculation of aluminum plate, the COD removal efficiency is 90.9% after optimization of response parameters [12]. The degradation effect of Ir-Ta electrode is better, which was in good agreement with the predicted value of 92.24% from the regression model, with small error. To confirm the predicted results, experiments were conducted using the optimum conditions that were given by the optimisation software. The COD removal efficiency is described in Table 4. The experimental results were close to the predicted values but with a lower percentage of error (-1.44, 0.30, 0.88, 0.80, 0.70).

Conclusions

The effects of current density, plate spacing, initial pH and reaction time on the removal rate of PAM were studied. RSM was used to optimize the experimental parameters. Regression analysis showed that the experimental data fitted well with the second-order polynomial model. The coefficient of determination R^2 was 0.9954, F -value was 9.77, and the curve model was significant. When the current density was 58.80 mA/cm², the electrode plate spacing was 3.07 cm,

the initial pH of the solution was 6.73, and the reaction time was 2.23 h. The COD removal rate reached 92.70%. In accordance with the analysis of variance, *p*-values of current density (X_1), plate spacing (X_2), pH (X_3) and reaction time (X_4) were less than 0.05, implying that they were significant factors. The interaction between current density and inter-electrode, inter-electrode and pH, inter-electrode and reaction time significantly affected the COD removal efficiency. The successful application of the BBD design experiment showed that the electrocatalytic technology can effectively degrade PAM in wastewater.

Acknowledgments

The authors are grateful for financial support from the Scientific Research Foundation of Educational Commission of Hubei Province, China (Q20201315).

Conflict of Interest

The authors declare no conflict of interest.

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