



A mechanistic approach and response surface optimization of the removal of oil and grease from restaurant wastewater by electrocoagulation and electroflotation

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ABSTRACT

Electrocoagulation and electroflotation are effective technologies for restaurant wastewater treatment, especially for the removal of oil and grease. The response surface methodology was used to establish a model of restaurant wastewater treatment using electrocoagulation and electroflotation. The model provides the optimum operation conditions. In addition, the contributions of electrocoagulation and electroflotation to the removal of oil and grease were determined under different conditions. The optimum operation conditions are an inter-electrode distance of 3.6 cm, a reaction time of 34 min, and a current density of 43 A/m². The removal efficiency of oil and grease was above 95% under such conditions. When wastewater conductivity was less than 3,000 µs/cm, electrocoagulation played the dominant role, and the contribution rates ranged from 68.1 to 72.5%. When the wastewater conductivity was above 3,500 µs/cm, electroflotation played the dominant role, and the contribution rates ranged from 51.6 to 65.8%. Hence, electrocoagulation dominated, whose contribution rates ranged from 68.1 to 90.7%, 68.1 to 75.8%, and 66.0 to 89.1% with the changes of the inter-electrode distance, reaction time, and current density, respectively.

Keywords: Response surface methodology; Oil and grease removal; Restaurant wastewater; Electrocoagulation; Electroflotation

1. Introduction

The cooking industry discharges millions of tons of wastewater every year, and the discharge of wastewater continues to increase. The restaurant wastewater has high oil and grease content and high chemical oxygen demand (COD). Electrocoagulation and electroflotation present good performance for restaurant wastewater

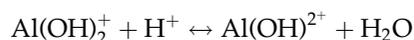
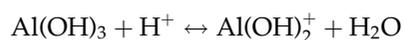
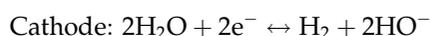
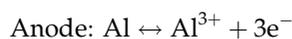
treatment. These technologies possess many advantages, such as short reaction time, no additional chemicals, small occupied areas, simple operations, and convenient management. Both are promising treatment technologies for oil and grease removal [1,2].

Chen et al. [3] preferred aluminum to iron as electrode material in electrocoagulation and electroflotation to deal with restaurant wastewater. The corrosion of iron electrode is more severe when the circuit is

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open. Restaurant wastewater is intermittent water, and therefore, aluminum is generally used as the anode plate. Mahvi et al. [4] improved the electroflotation oil removal efficiency from 70 to 99.5% under the best conditions by adding NaCl and a coagulant. The reaction obeys a first-order kinetic rate. Bande et al. [5] indicated that oil removal efficiency can reach 90% within 30 min by electroflotation. The electrocoagulation reactions are commonly used in COD removal. Tezcan et al. [6] studied vegetable oil refining wastewater and indicated that COD removal efficiency could reach 98.9% through electrocoagulation under optimal conditions. Bensadok et al. [7] showed that COD and turbidity removal efficiency of cutting oil emulsion could reach 92 and 99% by electrocoagulation, respectively, and about 1 g of oil could be removed using 10 mg of dissolved aluminum plate.

Electrocoagulation and electroflotation mainly rely on the electrochemical reaction to dissolve metal electrode (usually iron or aluminum). They generate metal flocculants to adsorb or precipitate pollutants and produce microscopic bubbles to remove the small colloidal particles. These microscopic bubbles have good lift ability. Their load capacity is twice as large as the pressurized air flotation [8,9]. In the electrochemical reaction, the hydrolysis product of the aluminum anode is related to the pH of the aqueous solution. The pH of restaurant wastewater is usually between 5 and 7. The reactions on the electrode surface and in the aqueous solution are as follows [10–12]:



When the anodic aluminum ion dissolves, the aluminum salt flocculants remove the pollutants from the wastewater. This phenomenon is known as the electrocoagulation effect. The tiny air bubbles generated on the electrodes form a flotation reaction zone which is known as the electroflotation effect. These phenomena are the two main effects of the electrochemical reaction.

The reaction is regarded as the simultaneous occurrence process of electrocoagulation and electroflotation [4]. Electroflotation mainly does with $\text{H}_2(\text{g})$ bubbles liberation from the cathode. And electroflotation

contribution may be increased when $\text{O}_2(\text{g})$ is produced from the anode, at the same time with metal ion liberation [13]. However, the contributions of electrocoagulation and electroflotation still need to be studied further. In this study, the response surface methodology (RSM) was used to establish a model of restaurant wastewater pretreatment. The model leads to the optimum conditions. In addition, the contributions of electrocoagulation and electroflotation to the removal of oil and grease were determined under different conditions.

2. Materials and method

2.1. Wastewater characteristics

The effluent was obtained from the oil separation tank of a university cafeteria. The oil and grease concentration ranged from 180 to 280 mg/L, and the pH was between 5 and 6. And the wastewater conductivity ranged from 1,625 to 3,480 $\mu\text{S}/\text{cm}$, while the corresponding salinity was between 1.2 and 3.2‰.

2.2. Reactor and testing methods

The reactor used was a $120 \times 100 \times 100$ mm (L \times W \times H) plexiglass vessel with an effective volume of 1 L, which is shown in Fig. 1. The anode was made of aluminum, and the cathode was made of stainless steel. The effective area of the electrode plate was 100×60 mm (L \times W), the other side was covered by plastic film to avoid the reaction, and the thickness of the plate was 2 mm. The space of the reactor plate at one end could be adjusted, and the other end had a stirrer at low speed.

The concentration of oil and grease was determined using infrared spectrophotometry [14] (using

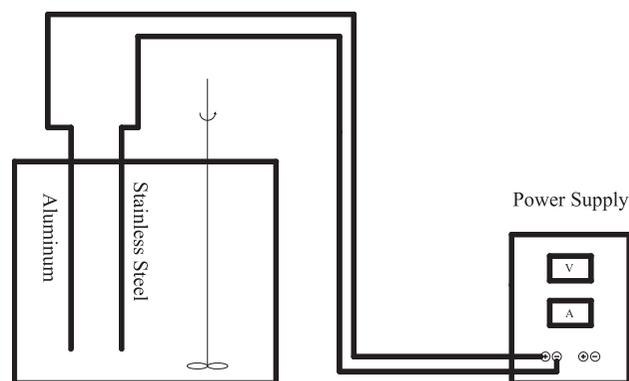


Fig. 1. Scheme of the electrocoagulation and electroflotation reactor.

an MAI-50G Infrared Oil Content Analyzer made by Little Swan Instruments Co., Ltd, China). The main analytical steps were as follows: (1) Mix water sample of 100 mL and CCl_4 of 25 mL into Separatory Funnel (250 mL), and shake for 5 min. (2) Remove the organic phase. (3) Determine oil concentration using the Infrared Oil Content Analyzer.

2.3. Experimental design

Two sections were considered: one was response surface optimization and another was removal contribution analysis of electrocoagulation and electroflotation.

RSM was regarded as one of the suitable methods for the electrochemical process to optimize the best operating conditions [15]. A Box–Behnken design with three factors and three levels was applied in this experiment. Four center points were added for each level of the categorical factor in order to estimate the experimental error and verify whether there was any curvature in the model to be fitted [16]. The independent variables included the inter-electrode distance, reaction time, and current density. The three-level value encodings of each independent variable were -1 , 0 , and 1 , which are shown in Table 1. The removal efficiency of oil and grease was regarded as the dependent variable. Table 1 shows the response values.

In RSM experiment, the water qualities of effluent were kept to their original values to lower the operational cost, such as pH and conductivity. The water samples conductivity (salinity) was focused on $2,980 \mu\text{s}/\text{cm}$ (2.2%), since those were the major part during sampling. The inter-electrode distance has impact on the capacity of reactor and energy consumption [17]. And the reaction time which is regarded as the hydraulics retention time in the pilot-scale reactor which is one of the most important parameters in water treatment. The current density not only determines the amount of coagulant that is generated, but also the rate of formation and size of the gas bubbles evolving on the electrode surface,

which help enhancing the mass transport of both Al^{3+} And OH^- to the bulk solution [18].

In the removal contribution analysis of electrocoagulation and electroflotation, we also discussed the impact of wastewater conductivities. As we know, the water qualities of restaurant wastewater effluent vary all the time. We sampled in different time, and determined samples conductivity and salinity to seek out the samples needed in the design.

2.4. Analysis method

The contributions of electrocoagulation and electroflotation technology to the removal of oil and grease were determined under different conditions. The electrocoagulation effect mainly depended on the role of the aluminum ions generated by the electrochemical reaction. The complex reaction between excess EDTA and Al ions could cover the electrocoagulation effect [19,20]. The removal efficiency of oil and grease (η_1) under this condition represented the contribution of electroflotation. The removal efficiency (η) without EDTA represented the total contributions of electrocoagulation and electroflotation. Eq. (1) expresses the contribution of electrocoagulation (η_2).

$$\eta_2 = \eta - \eta_1 \quad (1)$$

In this study, the contribution rate of electroflotation (A_1) is defined in Eq. (2). The contribution rate of electrocoagulation (A_2) is defined in Eq. (3).

$$A_1 = \eta_1 \times 100\%/\eta \quad (2)$$

$$A_2 = \eta_2 \times 100\%/\eta \quad (3)$$

Some control tests were conducted to examine whether EDTA addition influences the experimental results, by adding EDTA–Al complex saturated solution at the same concentration [21]. The ratios of the control and original tests were between 0.89 and 1.09, which indicated that EDTA addition did not influence

Table 1
RSM design

Variable, unit	Symbol		Level		
	Factor	Coded value	-1	0	1
Inter-electrode distance (cm)	X_1	A	2	4	6
Reaction time (min)	X_2	B	10	25	40
Current density (A/m^2)	X_3	C	10	30	50

the experimental results. Therefore, adding excess EDTA to the complex Al ion is a reliable experimental design for assessing the electrocoagulation effect.

3. Results and discussion

3.1. Experimental parameter optimization by RSM

RSM is a commonly used procedure in various fields for developing, improving, and optimizing processes. This procedure has been widely used to evaluate the relative significance of several factors, even in the presence of complex interactions [22]. The conditions were optimized through the design model using Minitab 16 software [23,24]. Table 2 shows the data.

Obtained in terms of coded factors:

$$Y_{(\text{Removalefficiency})} = 85.67 - 5.62A + 21.22B + 20.74C - 7.93AB + 5.26AC + 11.64BC - 15.90A^2 - 26.04B^2 - 20.69C^2 \quad (4)$$

Positive sign in front of the terms indicates synergistic effect, whereas negative sign indicates antagonistic effect. The removal efficiency results predicted by the Eq. (4), at each experimental point, are presented in Table 2.

It can be observed from Table 3 that the coefficients ($p < 0.01$ for all) are highly significant whereas the interaction terms (AC) are insignificant to the response. For a model to be reliable, the response

should be predicted with a reasonable accuracy by the model when compared to the experimental data. Fig. 2 compares experimental removal efficiency (%) with the predicted values obtained from the model. The figure indicated good agreements between the experimental and predicted values of removal efficiency.

The adequacy of the model was further supported by analysis of variance (ANOVA). The results of the ANOVA for removal efficiency are shown in Table 4. In this case, the p -value of 0.000 ($p < 0.05$) for regression model equation implies that the second-order polynomial model fitted to the experimental results well. The lack-of-fit was also calculated from the experimental error (pure error) and residuals. "Lack-of-fit F -value" of 51.34 implies the significance of model correlation between the variables and process response for removal.

The correlation coefficient R was 99.00%, the correction coefficient $R_{(\text{adj})}$ was 97.72%, and the coefficient of variation was 8.04%, which confirm the accuracy of the model. The model could be used to characterize the relationship between the dependent and independent variables because the variance was only 2.22% and the model was well-fitting. The interaction of A and B, and B and C were significant, while the interaction between A and C was general. The interaction term signs of the coefficients indicated a synergistic effect in AC and BC, however, an antagonistic effect was seen in the AB.

Table 2
RSM design and test results

No.	Inter-electrode distance (cm)	Reaction time (min)	Current density (A/m ²)	Removal efficiency of oil and grease (%)	
				Experimental	Predicted
1	6	25	10	12.80	17.46
2	4	10	10	12.80	8.62
3	4	25	30	86.99	85.67
4	4	10	50	27.61	26.83
5	2	40	30	78.00	78.49
6	4	40	50	88.35	92.53
7	4	40	10	27.00	27.78
8	6	25	50	68.19	69.46
9	6	10	30	25.31	24.82
10	2	10	30	14.76	20.21
11	6	40	30	56.85	51.40
12	2	25	50	74.85	70.18
13	4	25	30	84.72	85.67
14	4	25	30	86.32	85.67
15	4	25	30	84.92	85.67
16	2	25	10	40.50	39.23
17	4	25	30	85.40	85.67

Table 3
Estimated regression coefficients for removal efficiency (%) in coded units

Term	Coefficient	SE coefficient	<i>t</i>	<i>p</i>
Constant	85.67	2.046	41.870	0.000
A	-5.62	1.618	-3.474	0.010
B	21.22	1.618	13.115	0.000
C	20.74	1.618	12.820	0.000
A ²	-15.9	2.230	-7.130	0.000
B ²	-26.04	2.230	-11.680	0.000
C ²	-20.69	2.230	-9.278	0.000
AB	-7.93	2.288	-3.464	0.010
AC	5.26	2.288	2.299	0.055
BC	11.64	2.288	5.086	0.001

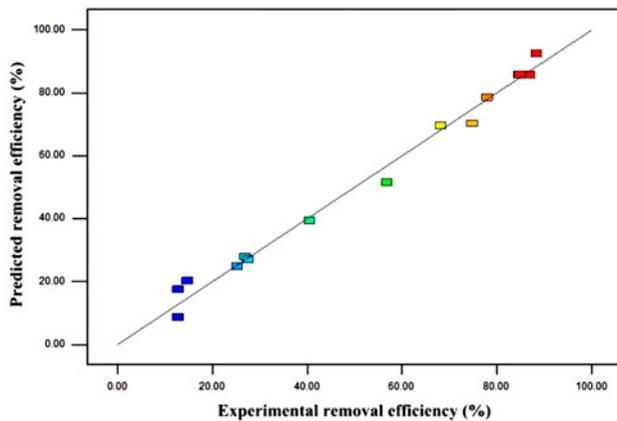


Fig. 2. Parity plot for the experimental and predicted value of removal efficiency.

To describe the effects of the inter-electrode distance, reaction time, and current density on the removal efficiency more directly, the three-dimensional response surfaces and contour maps were made, as shown in Figs. 3–5. The sparseness of contour maps indicates the rate of change of removal

efficiency with three dependent variables. A sparser contour line indicates a smaller rate of change of removal efficiency, whereas a denser line indicates a larger rate of change. The figures show that when the inter-electrode distance decreased, the reaction time and the current density increased, and the contour line was increasingly sparse. Under this condition, the removal efficiency of oil and grease was high and the rate of change was small. When the conditions reached a determined value (an inter-electrode distance of 3.6 cm, a reaction time of 34 min, and a current density of 43 A/m²), the increasing reaction time or current density or decreasing inter-electrode distance was not significant to removal efficiency. Table 5 shows the optimum conditions.

Also, the validation of the optimal values of parameters and response presented in Table 5 was performed in order to state whether the predicted values correspond to the experimental ones and thus to be concluded that the model obtained describes well the process. Under the model optimal parameters, the removal efficiency could reach 95.8 ± 0.6%, which was close to the predicted value. Thus, this model describes well the process, as the predicted values correspond to the experimental ones nearly.

3.2. Removal contribution analysis of electrocoagulation and electroflotation

This subsection mainly focuses on the oil and grease removal contributions by electrocoagulation and electroflotation with the changes of wastewater conductivity, inter-electrode distance, reaction time, and current density under the optimal conditions.

3.2.1. Wastewater conductivity

The optimal conditions were: inter-electrode distance of 4 cm, reaction time of 34 min, current density of 43 A/m². Wastewater conductivities: 1,625,

Table 4
ANOVA for removal efficiency (%)

Source	DF	Seq SS	Adj SS	Adj MS	<i>F</i>	<i>p</i>
Regression	9	14,555.6	14,555.6	1,617.28	77.26	0.000
Linear	3	7,293.7	7,293.7	2,431.2	116.15	0.000
Square	3	6,358.5	6,358.5	2,119.51	101.26	0.000
Interaction	3	903.4	903.4	301.1	14.39	0.002
Residual error	7	146.5	146.5	20.93		
Lack-of-fit	3	142.8	142.8	47.61	51.43	0.001
Pure error	4	3.70	3.70	0.93		
Total	16	14,702.1				

Note: $R^2 = 99.00\%$, $R^2(\text{adj}) = 97.72\%$.

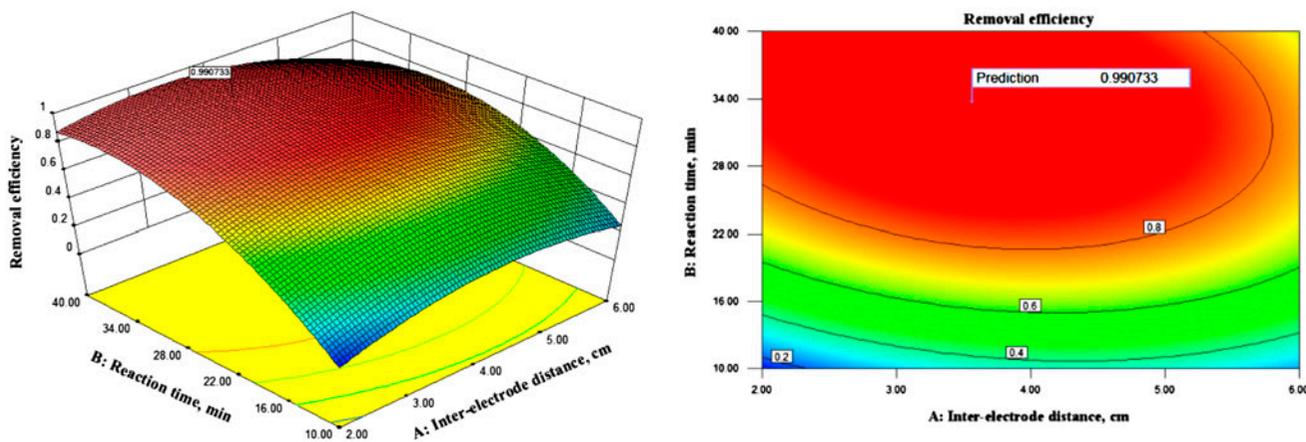


Fig. 3. Response surface and contour map with the effects of inter-electrode distance and reaction time.

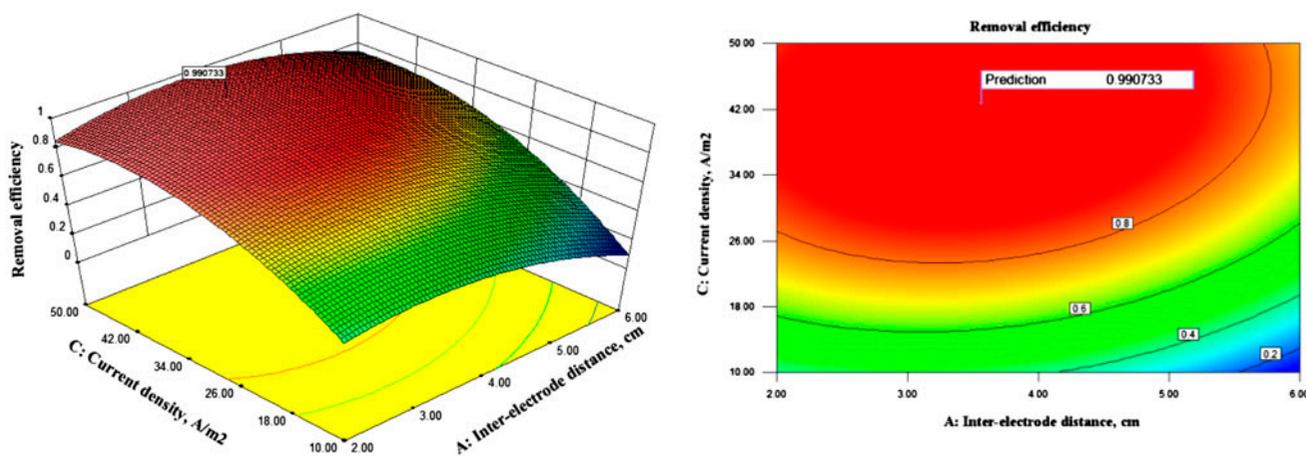


Fig. 4. Response surface and contour map with the effects of inter-electrode distance and current density.

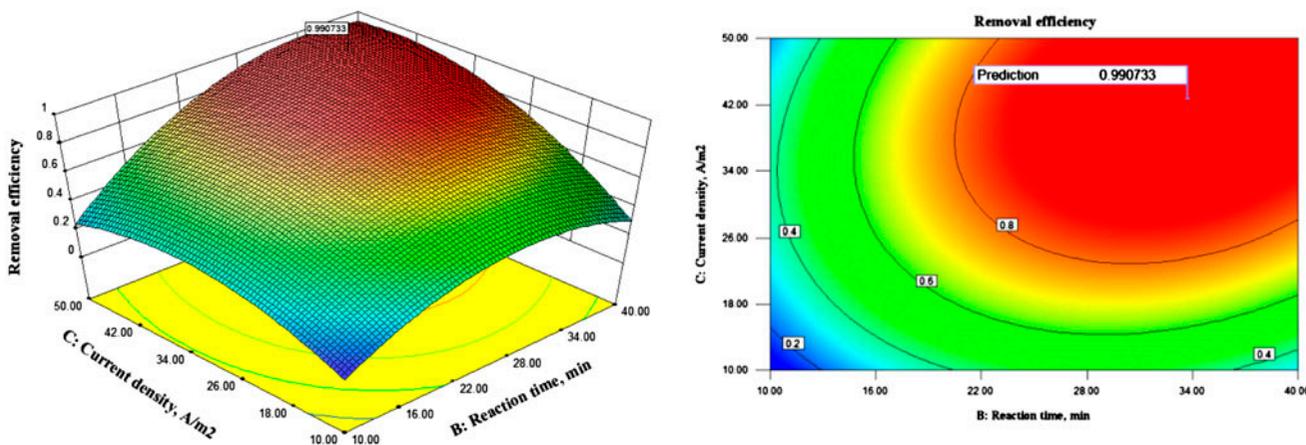


Fig. 5. Response surface and contour map with the effects of reaction time and current density.

Table 5
Model optimal parameters

Parameter, unit	Optimal value
Y (removal efficiency of oil and grease, %)	99
A (Inter-electrode distance, cm)	3.6
B (Reaction time, min)	34
C (Current density, A/m ²)	43

2,170, 2,980, 3,480, and 4,110 $\mu\text{S}/\text{cm}$; Corresponding wastewater salinities: 1.2, 1.7, 2.2, 2.7, and 3.2‰, respectively.

Fig. 6 shows that the removal efficiency increased with increasing wastewater conductivity. Therefore, larger wastewater conductivity is more conducive to the removal of oil and grease. The figure shows that the contribution of electroflotation increased with increasing wastewater conductivity. By contrast, the contribution of electrocoagulation decreased with increasing wastewater conductivity. When the wastewater conductivity was less than 3,000 $\mu\text{S}/\text{cm}$, the electrocoagulation played the dominant role with contribution rates of 68.1–72.5%. When the wastewater conductivity was above 3,500 $\mu\text{S}/\text{cm}$, the electroflotation played the dominant role with contribution rates between 51.6 and 65.8%.

3.2.2. Inter-electrode distance

Reaction time: 34 min; Current density: 43 A/m²; Wastewater conductivity: 2,980 $\mu\text{S}/\text{cm}$; Inter-electrode distances: 2, 4, 6 cm.

Fig. 7 shows that the removal efficiency increased with increasing the inter-electrode distance. Therefore, smaller inter-electrode distance is more conducive to

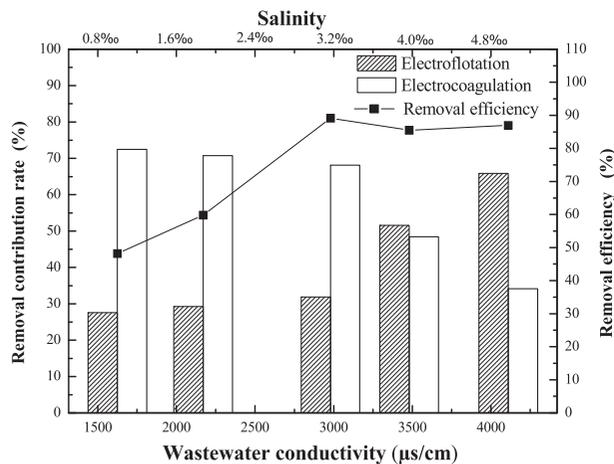


Fig. 6. Relationship among contribution rate, removal efficiency, and wastewater conductivity.

the removal of oil and grease. The figure also shows that the contribution rates of electroflotation ranged from 9.3 to 31.9%. Under this condition, the electrocoagulation played the dominant role with contribution rates of 68.1–90.7%.

3.2.3. Reaction time

Inter-electrode distance: 4 cm; Current density: 43 A/m²; Wastewater conductivity: 2,980 $\mu\text{S}/\text{cm}$; Reaction times: 10, 25, 34, 40 min.

When the reaction time increased, the removal efficiency increased and gradually tended to be stable, as shown in Fig. 8. Therefore, longer reaction time is more conducive to the removal of oil and grease. Fig. 8 also shows that the contribution rate of electroflotation increased with increasing reaction time and ranged from 24.2 to 31.9%. At this condition, the electrocoagulation played the dominant role with contribution rates of 68.1–75.8%.

3.2.4. Current density

Inter-electrode distance: 4 cm; Reaction time: 34 min; Wastewater conductivity: 2,980 $\mu\text{S}/\text{cm}$; Current densities: 10, 30, 43, 50 A/m².

When the current density increased, the removal efficiency increased and gradually tended to be stable, as shown in Fig. 9. Therefore, larger current density is more conducive to the removal of oil and grease. Fig. 9 also shows that the contribution rate of electroflotation increased with increasing current density and ranged from 10.9 to 34%. Under this condition, the electrocoagulation played the dominant role with contribution rates of 66–89.1%.

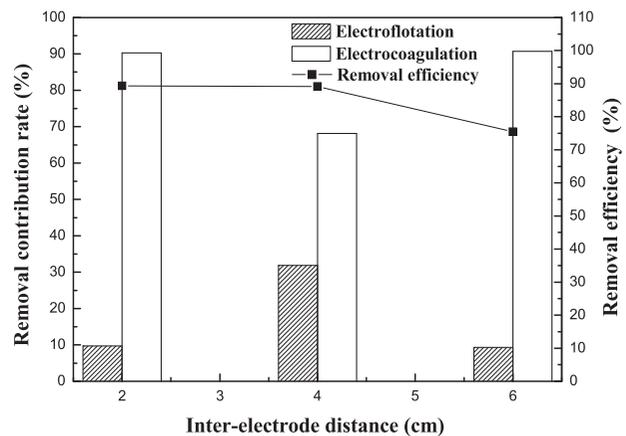


Fig. 7. Relationship among contribution rates, removal efficiency, and inter-electrode distance.

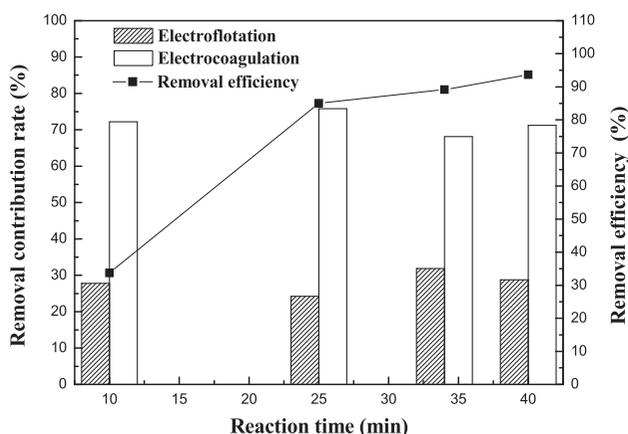


Fig. 8. Relationship among contribution rates, removal efficiency, and reaction time.

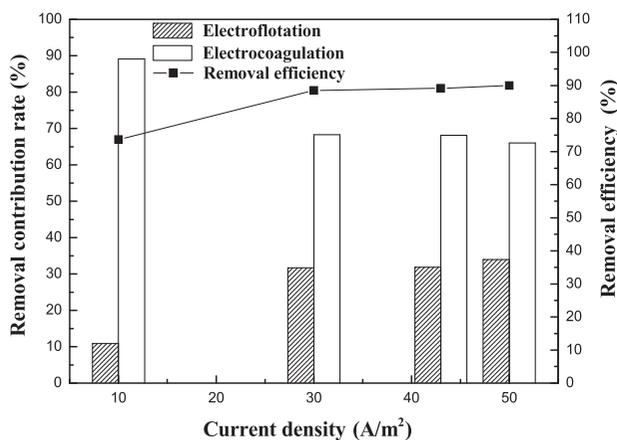


Fig. 9. Relationship among contribution rates, removal efficiency, and current density.

4. Conclusions

Electrocoagulation and electroflotation indicated a good performance on the removal of oil and grease. RSM was used to establish a model of the restaurant wastewater treatment. The model presented the optimum conditions. In addition, the contributions of electrocoagulation and electroflotation to the removal of oil and grease were determined under different conditions.

The optimum conditions were an inter-electrode distance of 3.6 cm, a reaction time of 34 min, and a current density of 43 A/m². The removal efficiency of oil and grease was above 95% under such conditions. When the wastewater conductivity was less than 3,000 $\mu\text{s}/\text{cm}$, electrocoagulation played the dominant role and the contribution rates ranged from 68.1 to 72.5%. When the wastewater conductivity was above

3,500 $\mu\text{s}/\text{cm}$, electroflotation played the dominant role, and the contribution rates ranged from 51.6 to 65.8%. Electrocoagulation dominated and the contribution rates ranged from 68.1 to 90.7%, 68.1 to 75.8%, and 66.0 to 89.1%, with the changes in inter-electrode distance, reaction time, and current density, respectively.

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