Optimization of Enhanced Ultrafiltration Conditions for Cd with Mixed Biosurfactants Using the Box-Behnken Response Surface Methodology

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Abstract: A mixture of the environmentally friendly biosurfactants rhamnolipids and sophorolipids was used as a source of micelles in this study. The Box-Behnken design and response surface methodology was used to investigate the influence of factors on micellar-enhanced ultrafiltration (MEUF). Simulated Cd-containing wastewater was used for testing. Based on single-factor experiments, the initial Cd²⁺ concentration, biosurfactant mixing ratio (α) and pH were chosen as influential variables, and both the Cd²⁺ rejection coefficient and permeation flux were used as responses. A predictive model based on a quadratic polynomial regression equation was established to determine the optimized enhanced ultrafiltration conditions for Cd. The results show that the regression equation is extremely significant and fits the data accurately. The optimal enhanced ultrafiltration conditions are as follows: initial Cd²⁺ concentration of 10.0 mg/L, α of 0.30 and pH of 9.58. Under these conditions, the rejection coefficient and the permeation flux of Cd²⁺ are 99.14% and 37.36 L/m²·h, respectively. The experimental results confirm that the experimental values agree well with the values predicted by the model. Further, these results provide theoretical support for using MEUF to treat heavy metal-containing wastewater when biosurfactants are used for micelle formation.

Keywords: micellar-enhanced ultrafiltration; Box-Behnken response surface methodology; cadmium; wastewater; optimization

1. Introduction

With increasing industrialization, cadmium (Cd) has become widely used in various fields, including mining, metal working, electroplating and paint manufacturing [1]. As a persistent pollutant, Cd persists for a very long time after entering water bodies [2,3]. Cd also harms human health through accumulation in food chains [4,5]. The common treatment methods for heavy metal ions in aqueous solutions include adsorption [6–9], bio-based methods [10–12], membrane separation techniques [13–15], electrolysis [16,17] and chemical precipitation [12,18]. Among these, micellar-enhanced ultrafiltration (MEUF), one of the membrane separation techniques, has received recent attention because it has the advantages of low energy consumption, easy operation, high permeation flux and high removal efficiency [1]. This technique removes heavy metal ions by adding surfactants to wastewater to form micelles that heavy metal ions in water adsorb or bind to, and thus

are retained by ultrafiltration membranes. The performance of the MEUF process may be influenced by some dominant factors including surfactant concentration, pH of solution and operating pressure of the process [19]. Currently, the surfactants used in MEUF are generally chemical in nature [20–22]. However, chemical surfactants have the shortcomings of high critical micelle concentrations (CMCs), the need to use large amounts for functionality and a high possibility of introducing secondary pollution, which restrict the promotion and application of MEUF. Although biosurfactants feature low CMCs and no secondary pollution, there are very few studies of their application in MEUF [23,24]. Therefore, this work uses a mixture of the environmentally friendly biosurfactants rhamnolipids and sophorolipids as a source of micelles and investigates their effect on the ultrafiltration of Cd. In addition, the Box-Behnken response surface is used to optimize the ultrafiltration conditions with the aim of providing theoretical support for using MEUF to treat heavy metal-containing wastewater when rhamnolipids and sophorolipids are used for micelle formation.

2. Materials and Methods

2.1. Experimental Setup

The experimental setup is shown in Figure 1. This includes a membrane pump, a pipeline, a pressure gauge and an ultrafiltration membrane. The equipment's technical parameters and the main performance parameters of the ultrafiltration membrane are shown in Tables 1 and 2, respectively.

Figure 1. Diagram of the experimental setup.

Table 1. Technical parameters of the equipment.

Size of Main Engine (cm)	Membrane Area (cm ²)	Power (Kw)	Minimum Circulating Volume (mL)	Working Pressure (MPa)	Filtration Capacity (L/h)	Applicable Temperature (°C)
23 × 27 × 30	150	0.06	80	≤0.5	0.3–3	5–50

Table 2. Main performance parameters of the ultrafiltration membrane.

Membrane Type	Membrane Material	Molecular Weight Cutoff (Dalton)	Permeation Flux (L/m ² ·h)	Membrane Area (cm ²)	pH
SMU-420	PVDF	10,000	24–53	150	2–12

2.2. Experimental Methods

Specified amounts of the biosurfactants, rhamnolipids and sophorolipids were weighed and added to simulated Cd-containing wastewater. After stirring with a magnetic stirrer on a hot plate, the solution was left standing for 20 min and then ultrafiltered. The simulated wastewater entered the membrane assembly through the membrane pump. The concentrated solution flowed back to the feed

chute for ultrafiltration, and the filtrate was collected in a special container to measure the permeation flux. An appropriate amount of filtrate was collected to measure the Cd concentration.

2.3. Box-Behnken Response Curve Design

Box-Behnken designs have served as a popular choice for second-order models, and have been widely used in completely randomized experiments, split-plot experiments and within the robust parameter design setting [25]. By choosing three different levels of the initial Cd²⁺ concentration, the mixing ratio of biosurfactants (α : the ratio of the sophorolipid concentration to the total surfactant concentration) and pH and using the rejection coefficient and permeation flux of Cd²⁺ as the two indexes, Minitab 16 was used to design the Box-Behnken response curve experiments. The three different levels of influential variables are shown in Table 3.

Table 3. Independent variables, levels and symbols for the Box-Behnken design.

Variable	Symbol	Variable Levels		
		Low (−1)	Middle (0)	High (+1)
Cd ²⁺ Concentration (mg/L)	A	10	25	40
α	B	0.05	0.175	0.3
pH	C	4	7	10

2.4. Sample Measurements and Calculation

1. The Cd²⁺ concentration of the wastewater was measured using an inductively coupled plasma optical emission spectrometer (PE ICP-OES Optima 7000DV).
2. The rejection coefficient was calculated using the following equation:

$$R = 1 - \frac{C_p}{C_f}, \quad (1)$$

where C_p is the concentration of pollutants in the filtrate (mg/L), C_f is the concentration of pollutants in the feed solution (mg/L), and R is the rejection coefficient for the pollutants.

3. The permeation flux was calculated using the following equation:

$$J = \frac{Q}{A\Delta t}, \quad (2)$$

where Q is the volume of filtrate (L), Δt is the running time (h), A is the area of the ultrafiltration membrane (m²), and J is the permeation flux (L/m²·h).

3. Results and Analysis

3.1. Analysis of the Cd²⁺ Rejection Coefficient

The experiments were conducted according to the Box-Behnken response surface design. The experimental and predicted values of the Cd²⁺ rejection coefficient (R_{Cd}) are shown in Table 4. Table 4 shows that the maximum experimental value of the Cd²⁺ rejection coefficient was 99.8%, which is close to the predicted value, indicating that the design of the Box-Behnken response surface is reasonable and scientific.

Minitab 16 was used to fit the experimental values of the Cd²⁺ rejection coefficient to obtain a complete quadratic regression model for the relationship among the Cd²⁺ rejection coefficient (R_{Cd}) and the initial Cd²⁺ concentration, the biosurfactant mixing ratio (α) and the pH, that is,

$$R_{Cd} = -63.730 - 0.962A + 23.920B + 40.062C - 0.013A^2 - 124.533B^2 - 2.422C^2 + 0.520A \times B + 0.139A \times C + 0.467B \times C, \quad (3)$$

The regression equation shows that the Cd^{2+} concentration had a negative effect on the Cd^{2+} rejection coefficient, while α and pH had positive effects on the Cd^{2+} rejection coefficient. These results indicate that the larger α and the pH are, the better the effect on the Cd^{2+} rejection coefficient is, and the greater the Cd^{2+} concentration is, the worse the effect on the Cd^{2+} rejection coefficient is.

Table 4. Experimental and predicted values of the Cd^{2+} rejection coefficient.

Std	Run	Factor			Experimental Cd^{2+} Rejection Coefficient (%)	Predicted Cd^{2+} Rejection Coefficient (%)
		A	B	C		
1	15	−1	−1	0	99.7	98.2
2	2	1	−1	0	76.2	80.1
3	5	−1	1	0	99.3	95.4
4	4	1	1	0	79.7	81.2
5	3	−1	0	−1	50.5	54.1
6	7	1	0	−1	27.2	25.4
7	12	−1	0	1	98.0	99.9
8	8	1	0	1	99.8	96.3
9	9	0	−1	−1	43.3	41.3
10	11	0	1	−1	39.7	40.1
11	6	0	−1	1	99.6	99.3
12	1	0	1	1	96.7	98.8
13	10	0	0	0	94.1	93.6
14	14	0	0	0	94.4	93.6
15	13	0	0	0	92.2	93.6

Various statistical parameters are presented in Table 5 for analysis of the results related to the Cd^{2+} rejection coefficient. The F values in Table 5 show that the factors' impacts on R_{Cd} are in the following order: $\alpha < \text{Cd}^{2+}$ concentration $< \text{pH}$. The P value is the probability value used to specify the statistically significant effects in the model. The P values of each item in Table 5 show that the effect of the pH on R_{Cd} is extremely significant, whereas the effects of the Cd^{2+} concentration and α on R_{Cd} are insignificant, and the effect of the interaction between the Cd^{2+} concentration and the pH on R_{Cd} (AC) is significant. The similar rule has been observed in previous experiments on mono-rhamnolipid micelles using MEUF [24].

Table 5. Tests of response function R_{Cd} .

Source	df	SS	MS	F-Value	P	Significant
Model	9	9253.67	1028.19	65.52	<0.001	**
Linear	3	7332.54	768.63	48.98	<0.001	**
Square	3	1759.71	586.57	37.38	0.001	**
Interaction	3	161.43	53.81	3.43	0.109	
A	1	521.64	41.76	2.66	0.164	
B	1	1.44	2.08	0.13	0.731	
C	1	6809.45	2004.08	127.7	0.000	**
A ²	1	5.38	30.96	1.97	0.219	
B ²	1	0.27	13.98	0.89	0.389	
C ²	1	1754.06	1754.06	111.77	<0.001	**
AB	1	3.80	3.80	0.24	0.643	
AC	1	157.50	157.50	10.04	0.025	*
BC	1	0.12	0.12	0.01	0.933	
Residual	5	78.47	15.69			
Lack of fit	3	75.62	25.21	17.71	0.054	
Pure error	2	2.85	1.42			
Cor total	14	9332.14				
R ² = 0.992						

Note: $P < 0.05$ indicates that this item is significant and is denoted by "**", $P < 0.01$ indicates that the item is extremely significant and is denoted by "**". A is the Cd^{2+} concentration; B is the ratio of the sophorolipid concentration to the total surfactant concentration (α); C is the pH; AC represents the interaction of the Cd^{2+} concentration (A) and the pH (B), and AB and BC are similarly defined; A² is the quadratic term of the Cd^{2+} concentration (A), and B² and C² are similarly defined.

Using the quadratic regression model, surface and isoline plots (Figures 2–4) can be drawn to investigate the effects of the three factors (A, B and C) and their interactions (AB, AC and BC) on the Cd^{2+} rejection coefficient.

Figure 2 shows that the interaction of the Cd^{2+} concentration and α is insignificant when the pH is 7; α has a smaller effect on R_{Cd} than the Cd^{2+} concentration does; and R_{Cd} decreases as the Cd^{2+} concentration increases. Figure 3 shows that the interaction of the Cd^{2+} concentration and the pH (AC) is significant when α is held at 0.175, and the optimum point is a Cd^{2+} concentration of approximately 13.2 mg/L at a pH of 8.7, where the Cd^{2+} rejection coefficient exceeds 98%. Figure 4 shows that the interaction of α and the pH (BC) is insignificant when the Cd^{2+} concentration is held at 25 mg/L; the pH has a greater effect on the Cd^{2+} rejection coefficient than α , and the Cd^{2+} rejection coefficient increases with the pH.

Figure 2. Response surface (a) and isolines (b) for the Cd^{2+} rejection rate, the Cd^{2+} concentration and α .

Figure 3. Response surface (a) and isolines (b) for the Cd^{2+} rejection rate, the Cd^{2+} concentration and the pH.

Figure 4. Response surface (a) and isolines (b) for the Cd^{2+} rejection rate, α and the pH.

3.2. Analysis of Permeation Flux

The experiments were conducted according to the Box-Behnken design (the response surface design). The experimental and predicted values of the permeation flux (J) are shown in Table 6.

Table 6. Experimental and predicted values of the permeation flux.

Std	Run	Factor			Experimental Values of Permeation Flux (L/m ² ·h)	Predicted Values of Permeation Flux (L/m ² ·h)
		A	B	C		
1	15	−1	−1	0	19.4	19.5
2	2	1	−1	0	21.3	21.0
3	5	−1	1	0	26.9	27.2
4	4	1	1	0	20.4	20.3
5	3	−1	0	−1	4.4	3.3
6	7	1	0	−1	3.5	2.9
7	12	−1	0	1	28.4	29.0
8	8	1	0	1	22.9	24.0
9	9	0	−1	−1	3.3	4.3
10	11	0	1	−1	3.8	4.5
11	6	0	−1	1	25.1	24.4
12	1	0	1	1	32.1	31.2
13	10	0	0	0	15.5	15.1
14	14	0	0	0	14.9	15.1
15	13	0	0	0	14.9	15.1

Minitab 16 was used to fit a quadratic regression model for the relationship among the permeation flux (J), the Cd²⁺ concentration, α and the pH to the experimental values of the permeation flux,

$$J = -17.532 - 0.342A - 79.793B + 8.618C + 0.012A^2 + 261.600B^2 - 0.346C^2 - 1.120A \times B - 0.026A \times C + 4.333B \times C, \quad (4)$$

The regression equation shows that the Cd²⁺ concentration and α had a negative effect on the permeation flux (J), while the pH had a positive effect on J. The results indicate that the higher the pH is, the higher the membrane flux is; the larger the Cd²⁺ concentration and α are, the smaller the membrane flux is.

According to the F values in Table 7, the factors' impacts on the permeation flux are in the following order: Cd²⁺ concentration < α < pH. In addition, the P values in Table 8 show that the effects of α and the pH on J are extremely significant; the effect of the Cd²⁺ concentration on J is insignificant; that the effects of the quadratic terms (A², B² and C²) of the Cd²⁺ concentration α and the pH on J are extremely significant and that the effects of the interactions between the Cd²⁺ concentration and α (AB), and between α and the pH (BC), on J are very significant.

Using the quadratic regression model, the surface and isolines (Figures 5–7) can be drawn to investigate the effects of the three factors and their interactions on the permeation flux.

Figure 5 shows that the interaction between the Cd²⁺ concentration and α (AB) is significant when the pH is kept at 7; when the Cd²⁺ concentration is 10–19.5 mg/L and α is greater than 0.24, the permeation flux is greater than 22.5 L/m²·h. Figure 6 shows that the interaction between the Cd²⁺ concentration and the pH (AC) is insignificant when α is kept at 0.175; the pH has a greater effect on the permeation flux than the Cd²⁺ concentration does, and the permeation flux increases with the pH. Figure 7 shows that the interaction between α and the pH (BC) is significant when the Cd²⁺ concentration is kept at 25 mg/L; α has a smaller effect on the permeation flux than the pH does, and J is greater than 20 L/m²·h when the pH is above 8.6.

Table 7. Tests for the response function, J.

Source	df	SS	MS	F-Value	P	Significant
Model	9	1299.65	144.405	113.57	<0.001	**
Linear	3	1132.76	50.798	39.95	0.001	**
Square	3	133.40	44.465	34.97	0.001	**
Interaction	3	33.49	11.164	8.78	0.019	*
A	1	15.12	5.272	4.15	0.097	
B	1	24.85	23.115	18.18	0.008	**
C	1	1092.78	92.741	72.94	<0.001	**
A ²	1	28.09	29.207	22.97	0.005	**
B ²	1	69.54	61.690	48.52	0.001	**
C ²	1	35.77	35.770	28.13	0.003	**
AB	1	17.64	17.640	13.87	0.014	*
AC	1	5.29	5.290	4.16	0.097	
BC	1	10.56	10.562	8.31	0.034	*
Residual	5	6.36	1.271			
Lack of fit	3	6.12	2.039	16.99	0.056	
Pure error	2	0.24	0.120			
Cor total	14	1306.00				
R ² = 0.995						

Note: $P < 0.05$ indicates that the item is significant and is denoted by “*”, $P < 0.01$ indicates that the item is extremely significant and is denoted by “**”. A is the Cd²⁺ concentration; B is the ratio of the sophorolipid concentration to the total surfactant concentration (α); C is the pH; AC represents the interaction of the Cd²⁺ concentration (A) and the pH (C), and AB and BC are similarly defined; A² is the quadratic term for the Cd²⁺ concentration (A), and B² and C² are similarly defined.

Figure 5. Response surface (a) and isolines (b) for the permeation flux, the Cd²⁺ concentration and α . Figure 6. Response surface (a) and isolines (b) for the permeation flux, the Cd²⁺ concentration and the pH.

Figure 7. Response surface (a) and isolines (b) for the permeation flux, α and the pH.

3.3. Prediction and Validation of Optimum Filtration Conditions

The response optimizer in Minitab 16 was used to examine the responses of the Cd^{2+} rejection coefficient and the permeation flux simultaneously. The weight and degree of importance of each index was set to 1 and the optimization results are shown in Table 8.

Table 8. Response to optimization results.

Optimization Condition	Response Project	Response Result	Desirability	Optimized Composite Desirability
Cd^{2+} concentration = 10.0 mg/L $A = 0.30$ pH = 9.58	Cd^{2+} rejection rate	99.14%	1.00	1.00
	Permeation flux	37.36 L/m ² ·h	1.00	

Table 8 shows that when the Cd^{2+} concentration is 10.0 mg/L, α is 0.30 and the pH is 9.58, the Cd^{2+} rejection coefficient and the permeation flux are optimized simultaneously; the optimized composite desirability is 1.00. To further validate the reliability of the model's predictions, the optimized operating conditions were used to conduct experiments. For convenience in actual operation, the Cd^{2+} concentration was set to 10.0 mg/L, α was set to 0.30, and the pH was set to 9.5 in the experiments. The measured Cd^{2+} rejection coefficient and the permeation flux were 98.74 % and 37.02 L/m²·h, respectively. The relative error was no greater than 1%, and the experimental values were close to the predicted values, which indicates that the optimization model used in the paper is very reliable.

4. Conclusions

This work uses a mixture of the environmentally friendly biosurfactants rhamnolipids and sophorolipids as a source of micelles and investigates their effect on the ultrafiltration of Cd. Furthermore, this work optimizes the ultrafiltration conditions by using the Box-Behnken response surface, and provides theoretical support for using MEUF to treat heavy metal-containing wastewater when biosurfactants are used for micelle formation. The main conclusions are as follows:

- (1) Interactions among the three factors (the initial Cd^{2+} concentration, the ratio of biosurfactants (α) and the pH) affect the enhanced ultrafiltration of Cd^{2+} , and the significance of their effects on the rejection coefficient follows the order $\alpha < \text{Cd}^{2+}$ concentration $<$ pH. The degree of impact on the permeation flux follows the order Cd^{2+} concentration $<$ α $<$ pH.
- (2) The optimum Cd^{2+} removal conditions obtained from the experiments are as follows: Cd^{2+} concentration: 10.0 mg/L; α : 0.30; and pH: 9.58. Under these conditions, the Cd^{2+} rejection coefficient reaches 99.14 %, and the permeation flux reaches 37.36 L/m²·h. The experimental results confirm that the experimental values agree well with the theoretically predicted values.

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