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Research article

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Assessing the effect of operating factors on phosphate removal by electrocoagulation process using Response Surface Methodology

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ABSTRACT
This study investigated the electrocoagulation batch model with two aluminum plate electrodes. The effect of operating factors on phosphate removal efficiency by electrocoagulation process using aluminum electrodes was evaluated by Response Surface Methodology (RSM), including current density, pH, initial phosphate concentration, and reaction time. A total of 15 experiments were set up following the Box-Behnken method in Design – Expert 12 software, with three center points. The optimal operating conditions were determined by analyzed collected data in Design – Expert 12 software: current density of 16.7 A/m^2, pH 5.6, reaction time of 33.5 min, and initial phosphate (P-PO_4^{3-}) of 66.249 mg/L. The correlation equation between treatment efficiency and factors was also determined as a quadratic model with R^2 = 0.9771. The interaction between the pairs of factors significantly affected P-PO_4^{3-} treatment efficiency, represented by 3D surface plots. The maximum P-PO_4^{3-} treatment performance was reached 99.6%. The treatment efficiency of real livestock wastewater was determined and reached a high agreement with the predicted value, 99.7% after 30 min. Generated sludge was evaluated, approximately 6.3±0.76 kg per 1 m^3 treated wastewater.
Key words: aluminum electrodes, electrocoagulation, phosphate

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INTRODUCTION

Phosphorus enters into the water resources from both point and non-point sources such as agricultural drainage, livestock, domestic and industrial effluents, etc.,[1]. The point sources are more effectively controlled than others[1]. Agriculture and livestock industry are the principal sources of wastewater containing phosphorus[2]. In nature and wastewater, phosphorus is present in orthophosphate, polyphosphate, and organic phosphate[1,2]. Organic phosphate is usually found in raw wastewaters and orthophosphate is dominant in the effluent of wastewater treatment plants because the hydrolysis process occurs in the biological state[1]. Phosphorus is the main contaminant causing eutrophication in freshwater bodies[1,3,4]. Total phosphorus concentration has a strong relationship with the development of chlorophyll in lake water [5]. Therefore, the removal of phosphorus becomes the most important method to control the eutrophication [5]. So, it is necessary to reduce phosphorus before discharge into receiving sources to prevent the eutrophication.

Phosphorus is well-known removed by biological method or chemical method[4,5]. Conventional biological as biofiltration or activated sludge process can be applied to handle phosphorus in the wastewater but have low efficiency, only up to 40%[1,4]. The biological treatment has high efficiency, up to 97% in the case of low phosphorus concentration with low cost[6]. A/O technology or MBR process also has a high treatment of phosphorus but a long reaction time and is expensive. The chemical method is highly effective in phosphorus treatment via a precipitation process with aluminum, ferric, or calcium salts[4]. The treatment performance of phosphorus by the chemical process may approach to 99%[1,4]. However, there are some advantages including high pH controlled requirements, expensive operation, difficulty determining optimal chemical dosage, generating of secondary pollution, and corrosion [1], [4].

Electrocoagulation is an effective method used in remove phosphorus, up to 100% due to metal ions released from the anode[2–4,6,7]. The metal, usually used to make the anode, is iron or aluminum[2–4,6,7]. And aluminum plate anode has been proven higher efficient than others[6]. The initial pH, phosphorus concentration, current density, electrical conductivity, distance of electrode, and reaction time are the operating parameters that directly affects the treatment performance[2–4,6,7]. Phosphorus removal by electrocoagulation was performed in the previous studies and the results showed that electrocoagulation

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achieved high efficiency. 99.99% of 50.13 mg/L initial P-PO_4^{3-} in the synthetic domestic wastewater was removed by electrocoagulation with Al-Fe anode and Ti cathode, at the optimal condition including pH, current density 20 A/m^2, and reaction time of 80 min[6]. The treatment of phosphorus in synthetic and microbrewery wastewater by electrocoagulation using Al electrodes was investigated by James Arambarri et al.,[3]. The removal efficiency was 95% and 98% after 10 min electrocoagulation with adding 15 mg/L aluminum sulfate[3]. RSM (Response surface methodology) was applied to assess the effect of five operating factors the phosphorus removal including pH, NaCl concentration, voltage, reaction time, and inter-electrode distance[7]. The initial phosphate was fixed at 90 mg/L. The removal efficiency was predicted as high as 98%. It had not been evaluated the effect of initial phosphorus concentration change on the treatment efficiency.

In this study, the optimal operating conditions of pH, current density, reaction time, and phosphorus were determined by RSM. The experiments were set up with three center points by the Expert Design 12 software. The phosphorus treatment performance was tested on the real livestock wastewater taken from Loc Phat Farm, Hamlet 8B, Loc Hoa commune, Loc Ninh district, Binh Phuoc province.

MATERIAL AND RESEARCH METHODS

The wastewater

The wastewater influent was used in this study including synthetic wastewater and the real livestock wastewater. The synthetic wastewater was made by mixing $Na_3PO_4.12H_2O$ (Xilong China) in the tap water. The real wastewater was taken after the biogas tank at Loc Phat Farm. pH and electrical conductivity were adjusted by NaOH 1N, H_2SO_4 1N and NaCl.

Equipment

The equipment was used in this research:

- DC Power Supply, model QJ3010E with the maximum output voltage of 30 V and current of 10 A;
- pH controller: Hanna, model HI98107 pHep®;
- Conductivity controller: Hanna model HI98304 DiST® 4;
- Magnetic stirrer: VELP;
- UV-5100, UV-VIS Spectrophotometer

Experimental procedure

Experimental bench scale

An experimental bench scale, 15 cm x 15 cm x 16.5 cm in length, width, and height dimension, was operated by batch mode. 02 plates-Aluminum electrodes (100 mm x 170 mm, 5 mm thicken) were set up with a distance of 2.5 cm. Direct current was directly supplied to 02 plate electrodes by a DC power supply. Voltage and current can be adjusted and displayed on the DC Power supply device.

Experimental procedure

Evaluating of pH, current density, phosphorus concentration, and reaction time were performed in a batch reactor containing 3 liters of synthetic wastewater with experimental setup by Design Expert 12 software. Conductivity and distances of electrodes were fixed at 2 mS/cm and 2.5 cm, respectively.

Evaluate the treatment efficiency of real livestock wastewater by electrocoagulation: 3 liters of real livestock wastewater were filled into the reaction tank. The P removal performances were determined under the determined optimal operating conditions by RSM. After setup reaction time, the effluent sample was taken and settled for 60 min to separate sludge and water. Total P was analyzed to calculate the treatment efficiency.

Every experiment was repeated 3 times to ensure the collected results.

RESULTS AND DISCUSSION

Evaluation of optimal operating factors

In these experiments, 3 liters of synthetic wastewater was put into a batch reactor. Four factors including pH, current density, P initial concentration, and reaction time were adjusted and controlled. The electrical conductivity and distances of electrodes were fixed at 2 mS/cm and 2.5 cm, respectively. The optimal operating conditions were determined by RSM with Design Expert 12 software based on the response of P removal efficiency. The experiments were set up following Box Bhenken with 3 runs at the center point. The collected results are obtained in Table 1.

From the experimental results in Table 1, using Design Expert 12 to analyze model statistics, ANOVA, prediction results, 3D surface, and optimization condition.

The statistical model

The statistical model results showed that the total P removal was suggested as a quadratic model, focusing on the model maximizing the **Adjusted R^2** and

Table 1: Experiment result data and model prediction results on total P treatment

Run	Factor 1	Factor 2	Factor 3	Factor 4	Response 1	
	A: Current density, A/m2	B: pH	C: Reaction time, min	D: P initial concentration, mg/L	P removal efficiency, %	
					Actual value	Prediction
1	61.92	6.5	5	80	19.96	18.13
2	30.96	6.5	20	80	41.07	46.77
3	61.92	6.5	35	50	97.22	102.05
4	61.92	5	35	65	99.56	100.70
5	61.92	8	5	65	15.68	13.79
6	30.96	8	20	65	56.22	54.52
7	30.96	6.5	5	65	22.85	17.05
8	61.92	6.5	20	65	81.42	81.43
9	61.92	6.5	20	65	80.19	81.43
10	61.92	6.5	20	65	81.63	81.43
11	92.88	6.5	35	65	99.63	103.18
12	61.92	8	20	50	90.62	88.19
13	61.92	6.5	5	50	36.72	43.33
14	30.96	6.5	35	65	95.75	94.87
15	92.88	8	20	65	76.39	81.90
16	61.92	6.5	20	65	82.37	81.43
17	61.92	8	35	65	88.76	83.73
18	61.92	6.5	20	65	81.53	81.43
19	61.92	5	5	65	42.76	47.05
20	30.96	6.5	20	50	85.86	91.06
21	61.92	5	20	50	90.26	82.48
22	92.88	5	20	65	99.27	103.97
23	30.96	5	20	65	85.20	82.68
24	61.92	6.5	35	80	86.63	83.01
25	61.92	8	20	80	29.71	35.24
26	92.88	6.5	20	80	99.22	93.27
27	92.88	6.5	20	50	99.65	93.22
28	92.88	6.5	5	65	58.78	57.40
29	61.92	5	20	80	91.01	91.18

the **Predicted R^2**. R^2, adjusted R^2 values are 0.9771 and 0.9542, respectively. They are closer to 1 so the model is suitable, and significant, and illustrates good agreement between the observation and prediction value [8,9]. The predicted R^2 is 0.8685, the difference is less than 0.1. So, it is in reasonable agreement with the adjusted R^2. The good relationship between the P treatment observation and prediction results was clearly illustrated in Figure 1 and Table 1.

After analyzing the experimental results by Design Expert 12, ANOVA analysis results showed that there are 4 independent variables including pH, current density, P concentration, and reaction time. The interaction effects of two factors when others are their central values are demonstrated as well. The results indicated that all pH, current density, P concentration, and reaction time are the most effective factors and there is an interaction effect between factors. The 3D response surface plots P removal efficiency are presented in Figure 2.

The regression equations in terms of coded factors (-1:+1) are found to express the dependence of the treatment efficiency on 4 main operating factors as follows:

$$Y = 81.43 + 12.17A - 12.56B + 30.9C - 11.06D + 1.52AB - 8.01AC + 11.09AD + 4.07BC - 15.41BD + 1.54CD + 3.07A^2 - 3.74B^2 - 16.38C^2 - 3.42D^2$$

The optimal factors were determined from numerical solutions of optimization in Design Expert 12. The results indicated that there are 100 solutions found to reach the maximum efficiency of 99.6%. The first solution was chosen to perform continued experiments, including pH 5.6, a current density of 62 A/m^2, a reaction time of 33.5 min, and a P initial concentration of 66 mg/L.

Evaluate the treatment efficiency of real livestock wastewater by electrocoagulation.

The treatment efficiency of real livestock wastewater was determined under optimal conditions including pH 5.6, current density of 62 A/m^2. The P initial concentration was 53.55±0.51 mg/L. The removal efficiency reached 98.5±0.74% at 15 min and 99.74±0.12% at 30 min. In comparison to prediction efficiency by RSM and Design Expert 12, these results have so high agreement with prediction values. pH of the effluent was 7.3±0.1, illustrating that the real wastewater has high buffering properties.

Generated sludge was evaluated as well, 3±0.5 g/L with 15 min, and 6.3±0.76 g/L with 30 min.

CONCLUSIONS

The electrocoagulation has very impressive efficiency in phosphate removal of both synthetic and real livestock wastewater. RSM with Design Expert 12 gave a good prediction of the optimal operating conditions and efficiency. The P treatment performance was able to reach 99.78%.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest in the publication of the article "Assessing the effect of operating factors on phosphate removal by electrocoagulation process using Response Surface Methodology"

AUTHOR'S CONTRIBUTION

Ngoc-Han T. Huynh: Conceived the ideas; designed the research, analyzed the data, and wrote the paper.

Bich- Ngoc T. Nguyen: performed experiments.

Thanh Tran: provided equipment and collected the data.

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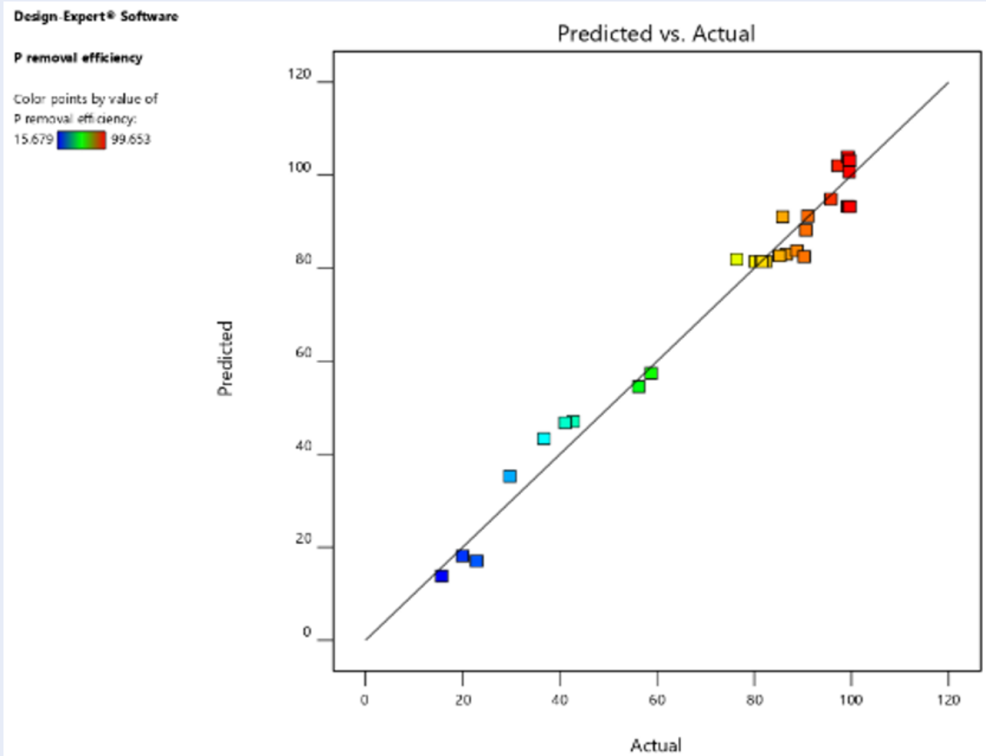

Figure 1: The correlation between observation and prediction

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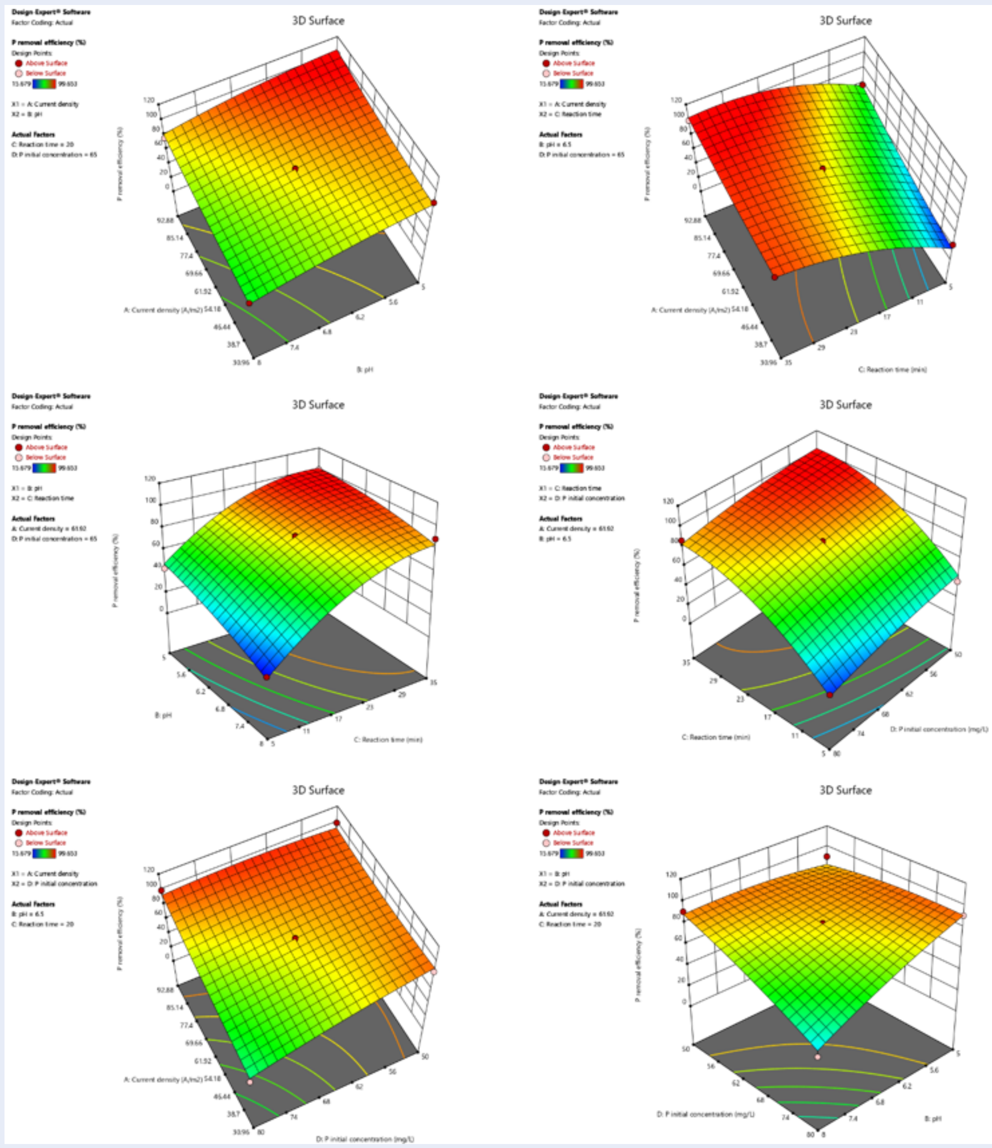

Figure 2: 3D response surface plots

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Bài nghiên cứu

Đánh giá ảnh hưởng của các yếu tố vận hành đến việc loại bỏ photphat bằng quá trình keo tụ điện hóa sử dụng phương pháp bề mặt đáp ứng

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TÓM TẮT

Nghiên cứu này đã được thực hiện trên mô hình keo tụ điện hóa dạng mẻ với 2 tấm điện cực nhôm phẳng. Ảnh hưởng của các yếu tố vận hành đến hiệu quả loại bỏ photphat bằng quá trình keo tụ điện hóa sử dụng điện cực nhôm được đánh giá bằng Phương pháp bề mặt đáp ứng (RSM), bao gồm mật độ dòng điện, pH, nồng độ photphat ban đầu và thời gian phản ứng. Tổng số 15 thí nghiệm được bố trí theo phương pháp Box-Behnken trong phần mềm Design – Expert 12, với 3 điểm trung tâm. Điều kiện vận hành tối ưu được xác định bằng số liệu thu thập từ thực nghiệm và phân tích trong phần mềm Design – Expert 12 gồm mật độ dòng điện 16,7 A/m ^{2}, pH 5,6, thời gian phản ứng 33,5 phút và hàm lượng phốt phát ban đầu (P-PO $_{4}$ $^{3-}$) là 66,249 mg/ L. Phương trình tương quan giữa hiệu quả xử lý và các yếu tố cũng được xác định, tuân theo phương trình bậc 2 với R^2 = 0.9771. Sự tương tác giữa các cặp yếu tố có ảnh hưởng đáng kể đến hiệu quả xử lý P-PO $_{4}$ $^{3-}$, được thể hiện bằng đồ thị bề mặt 3D. Hiệu suất xử lý P-PO $_{4}$ $^{3-}$ tối đa đạt 99,6%. Hiệu quả xử lý nước thải chăn nuôi thật cũng được đánh giá và đạt sự tương thích cao với hiệu suất dự báo từ mô hình, 99,7% sau 30 phút phản ứng. Lượng bùn sinh ra khi xử lý 1 m^3 nước thải là 6.3±0.76 kg.

Từ khoá: điện cực nhôm, keo tụ điện hóa, phốt phát

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