

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

Ngoc-Han T. Huynh^{1,*}, Bich- Ngoc T. Nguyen¹, Thanh Tran²



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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/m², pH 5.6, reaction time of 33.5 min, and initial phosphate (P-PO₄³⁻) of 66.249 mg/L. The correlation equation between treatment efficiency and factors was also determined as a quadratic model with R² = 0.9771. The interaction between the pairs of factors significantly affected P-PO₄³⁻ treatment efficiency, represented by 3D surface plots. The maximum P-PO₄³⁻ 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 m³ treated wastewater.

Key words: aluminum electrodes, electrocoagulation, phosphate

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1 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 others¹. Agriculture and livestock industry are the principal sources of wastewater containing phosphorus². 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 state¹. 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 cost⁶. 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 salts⁴. 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 to 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 others⁶. 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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53 achieved high efficiency. 99.99% of 50.13 mg/L initial P-PO₄³⁻ in the synthetic domestic wastewater 54 was removed by electrocoagulation with Al-Fe anode 55 and Ti cathode, at the optimal condition including 56 pH, current density 20 A/m², and reaction time of 57 80 min⁶. The treatment of phosphorus in synthetic 58 and microbrewery wastewater by electrocoagulation 59 using Al electrodes was investigated by James Aram- 60 barri et al.,³. The removal efficiency was 95% and 98% 61 after 10 min electrocoagulation with adding 15 mg/L 62 aluminum sulfate³. RSM (Response surface method- 63 ology) was applied to assess the effect of five oper- 64 ating factors the phosphorus removal including pH, 65 NaCl concentration, voltage, reaction time, and inter- 66 electrode distance⁷. The initial phosphate was fixed 67 at 90 mg/L. The removal efficiency was predicted as 68 high as 98%. It had not been evaluated the effect of 69 initial phosphorus concentration change on the treat- 70 ment efficiency. 71 In this study, the optimal operating conditions of pH, 72 current density, reaction time, and phosphorus were 73 determined by RSM. The experiments were set up 74 with three center points by the Expert Design 12 soft- 75 ware. The phosphorus treatment performance was 76 tested on the real livestock wastewater taken from Loc 77 Phat Farm, Hamlet 8B, Loc Hoa commune, Loc Ninh 78 district, Binh Phuoc province. 79

80 MATERIAL AND RESEARCH 81 METHODS

82 The wastewater

83 The wastewater influent was used in this study in- 84 cluding synthetic wastewater and the real livestock 85 wastewater. The synthetic wastewater was made by 86 mixing Na₃PO₄·12H₂O (Xilong China) in the tap wa- 87 ter. The real wastewater was taken after the biogas 88 tank at Loc Phat Farm. pH and electrical conductivity 89 were adjusted by NaOH 1N, H₂SO₄ 1N and NaCl.

90 Equipment

91 The equipment was used in this research:

- 92 • DC Power Supply, model QJ3010E with the 93 maximum output voltage of 30 V and current of 94 10 A;
- 95 • pH controller: Hanna, model HI98107 pHep®;
- 96 • Conductivity controller: Hanna model HI98304 97 DiST® 4;
- 98 • Magnetic stirrer: VELP;
- 99 • UV-5100, UV-VIS Spectrophotometer

Experimental procedure

Experimental bench scale

100 An experimental bench scale, 15 cm x 15 cm x 16.5 cm 102 in length, width, and height dimension, was operated 103 by batch mode. 02 plates-Aluminum electrodes (100 104 mm x 170 mm, 5 mm thick) were set up with a dis- 105 tance of 2.5 cm. Direct current was directly supplied 106 to 02 plate electrodes by a DC power supply. Voltage 107 and current can be adjusted and displayed on the DC 108 Power supply device. 109

Experimental procedure

110 Evaluating of pH, current density, phosphorus con- 111 centration, and reaction time were performed in a 112 batch reactor containing 3 liters of synthetic wastewa- 113 ter with experimental setup by Design Expert 12 soft- 114 ware. Conductivity and distances of electrodes were 115 fixed at 2 mS/cm and 2.5 cm, respectively. 116 Evaluate the treatment efficiency of real livestock 117 wastewater by electrocoagulation: 3 liters of real live- 118 stock wastewater were filled into the reaction tank. 119 The P removal performances were determined un- 120 der the determined optimal operating conditions by 121 RSM. After setup reaction time, the effluent sample 122 was taken and settled for 60 min to separate sludge 123 and water. Total P was analyzed to calculate the treat- 124 ment efficiency. 125 Every experiment was repeated 3 times to ensure the 126 collected results. 127

RESULTS AND DISCUSSION

Evaluation of optimal operating factors

128 In these experiments, 3 liters of synthetic wastewater 130 was put into a batch reactor. Four factors including 131 pH, current density, P initial concentration, and reac- 132 tion time were adjusted and controlled. The electrical 133 conductivity and distances of electrodes were fixed at 134 2 mS/cm and 2.5 cm, respectively. The optimal oper- 135 ating conditions were determined by RSM with De- 136 sign Expert 12 software based on the response of P 137 removal efficiency. The experiments were set up fol- 138 lowing Box Bhenken with 3 runs at the center point. 139 The collected results are obtained in Table 1. 140 From the experimental results in Table 1, using De- 141 sign Expert 12 to analyze model statistics, ANOVA, 142 prediction results, 3D surface, and optimization con- 143 dition. 144

The statistical model

145 The statistical model results showed that the total P 146 removal was suggested as a quadratic model, focus- 147 ing on the model maximizing the Adjusted R² and 148

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

Run	Factor 1 A: Current density, A/m ²	Factor 2 B: pH	Factor 3 C: Reaction time, min	Factor 4 D: P initial concentration, mg/L	Response 1 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

149 the Predicted R^2 . R^2 , adjusted R^2 values are 0.9771
 150 and 0.9542, respectively. They are closer to 1 so the
 151 model is suitable, and significant, and illustrates good
 152 agreement between the observation and prediction
 153 value^{8,9}. The predicted R^2 is 0.8685, the difference
 154 is less than 0.1. So, it is in reasonable agreement with
 155 the adjusted R^2 . The good relationship between the
 156 P treatment observation and prediction results was
 157 clearly illustrated in Figure 1 and Table 1.

158 After analyzing the experimental results by Design
 159 Expert 12, ANOVA analysis results showed that there
 160 are 4 independent variables including pH, current
 161 density, P concentration, and reaction time. The in-
 162 teraction effects of two factors when others are their
 163 central values are demonstrated as well. The results
 164 indicated that all pH, current density, P concentra-
 165 tion, and reaction time are the most effective factors
 166 and there is an interaction effect between factors. The
 167 3D response surface plots P removal efficiency are
 168 presented in Figure 2.

169 The regression equations in terms of coded factors (-
 170 1:+1) are found to express the dependence of the treat-
 171 ment efficiency on 4 main operating factors as follows:
 172 $Y = 81.43 + 12.17A - 12.56B + 30.9C - 11.06D +$
 173 $1.52AB - 8.01AC + 11.09AD + 4.07BC - 15.41BD +$
 174 $1.54CD + 3.07A^2 - 3.74B^2 - 16.38C^2 - 3.42D^2$

175 The optimal factors were determined from numerical
 176 solutions of optimization in Design Expert 12. The
 177 results indicated that there are 100 solutions found to
 178 reach the maximum efficiency of 99.6%. The first so-
 179 lution was chosen to perform continued experiments,
 180 including pH 5.6, a current density of 62 A/m², a re-
 181 action time of 33.5 min, and a P initial concentration
 182 of 66 mg/L.

183 Evaluate the treatment efficiency of real 184 livestock wastewater by electrocoagula- 185 tion.

186 The treatment efficiency of real livestock wastewa-
 187 ter was determined under optimal conditions includ-
 188 ing pH 5.6, current density of 62 A/m². The P
 189 initial concentration was 53.55±0.51 mg/L. The re-
 190 moval efficiency reached 98.5±0.74% at 15 min and
 191 99.74±0.12% at 30 min. In comparison to prediction
 192 efficiency by RSM and Design Expert 12, these results
 193 have so high agreement with prediction values. pH
 194 of the effluent was 7.3±0.1, illustrating that the real
 195 wastewater has high buffering properties.

196 Generated sludge was evaluated as well, 3±0.5 g/L
 197 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 live-
 stock 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
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 ology”

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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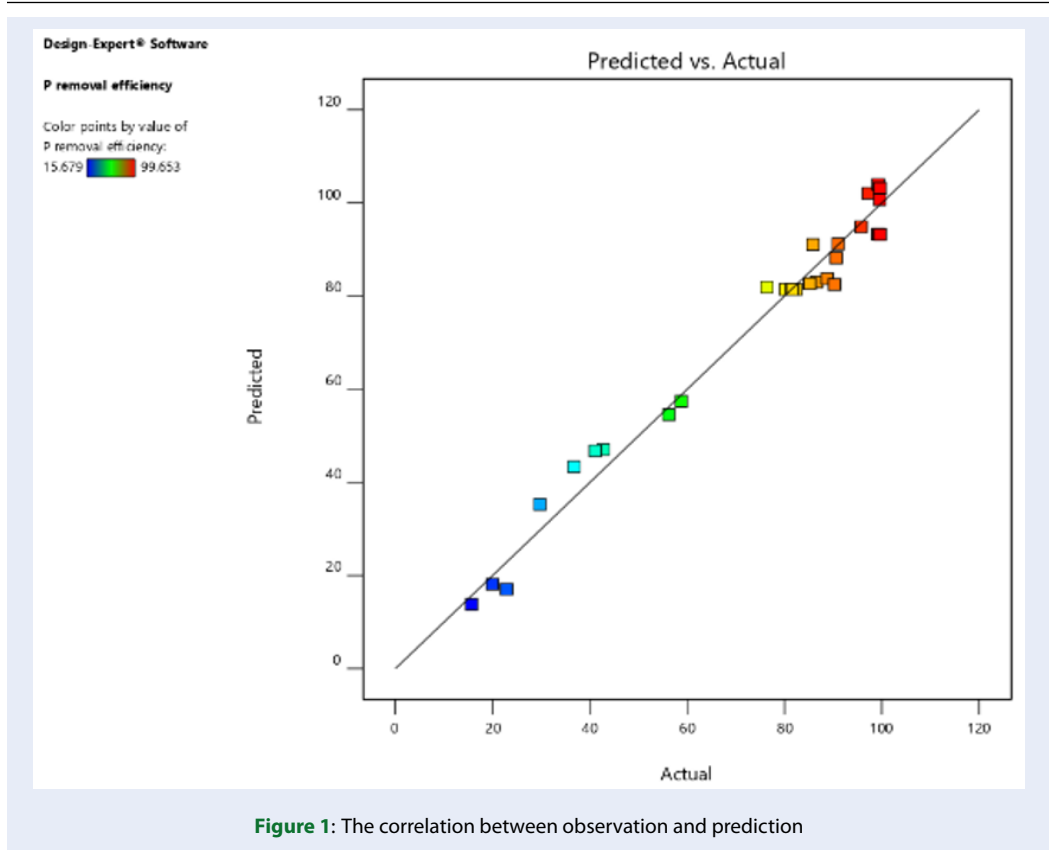


Figure 1: The correlation between observation and prediction

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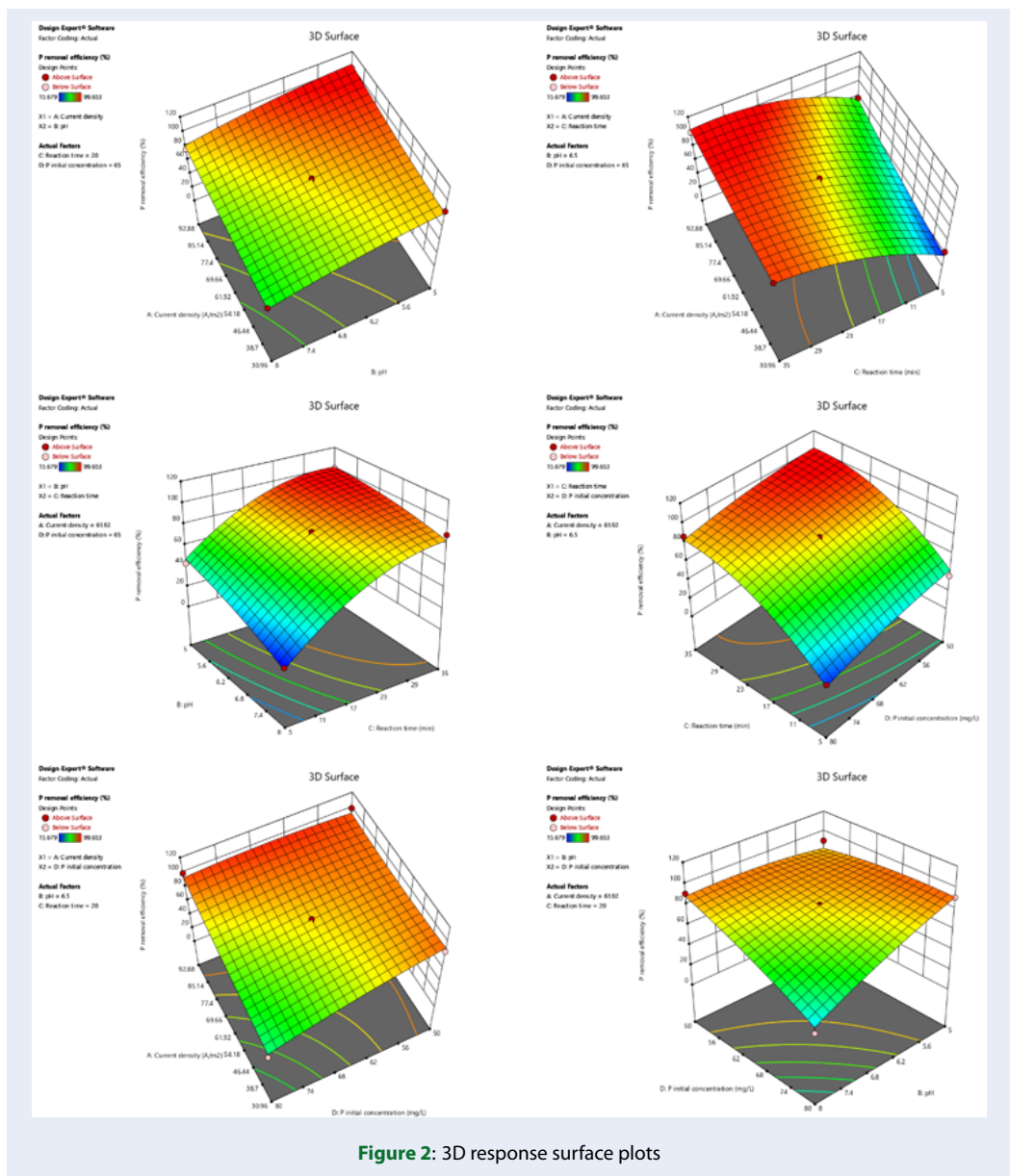


Figure 2: 3D response surface plots

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/m², pH 5,6, thời gian phản ứng 33,5 phút và hàm lượng phốt phát ban đầu (P-PO₄³⁻) 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 R² = 0,9771. Sự tương tác giữa các cặp yếu tố có ảnh hưởng đáng kể đến hiệu quả xử lý P-PO₄³⁻, được thể hiện bằng đồ thị bề mặt 3D. Hiệu suất xử lý P-PO₄³⁻ 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 m³ nước thải là 6.3±0.76 kg.

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

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