



# Iron Recovery from Water of Acid Sulfate Soil Using Fixed-bed Cation Exchange: an Optimization Study



[https://doi.org/10.47125/jesam/2025\\_1/05](https://doi.org/10.47125/jesam/2025_1/05)

## ABSTRACT

The Design Expert 12 software was used to optimize the recovery of iron from naturally iron alum water by ion exchange using a fixed bed column of 225H resin (India). This method also showed the ability to recover aluminum, calcium, and magnesium from water. At first, the study found suitable values of parameters such as cation exchange resin bed height of 80 cm, pH 6.5, and flow rate of 30 L h<sup>-1</sup> by examining each individual parameter based on the evaluation of iron removal efficiency in water. Then, the optimization process was carried out with an initial iron content (iron content in natural iron-alum water before passing through the column) of 8.25±0.18 mg L<sup>-1</sup>. The optimal conditions were found through Design Expert 12 software with the values of parameters such as 80 cm (height of ion exchange resin layer), 6.5 (pH value), and 30 L h<sup>-1</sup> (flow rate through the column). This was consistent with the conditions found initially (investigating each individual parameter) with an output iron content in the water of 0.5 mg L<sup>-1</sup>. The actual tested output iron content was less than 0.5 mg L<sup>-1</sup>, with a treatment efficiency of about 94%. The output iron concentration was below 0.5 mg L<sup>-1</sup> with a treatment efficiency of over 93% (the actual iron ion content after treatment was 0.25-0.35 mg L<sup>-1</sup> compared to the result from the predicted model of 0.49 mg L<sup>-1</sup>). Besides the ability to remove iron, other ions such as aluminum, calcium, and magnesium after passing through the column also had a relatively high treatment efficiency of over 85%. This result proved the effectiveness of iron treatment in natural iron-alum water by cation exchange with different iron contents, as well as the relevance of the model in practice.

**Keywords:** natural iron-alum water; cation exchange method; recovery of iron ions, iron treatment

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## INTRODUCTION

The so-called iron-alum water is the natural water with high levels of iron, aluminum, and other metals (e.g., Ca, Mg, and Si) released from acid sulfate soil (Nguyen *et al.* 2021; Tran *et al.* 2022). Iron plays an important role in human through its involvement in hemoglobin, myoglobin, and various enzymes. Iron deficiency in the body can cause anemia and negatively impact overall health. However, when the iron content exceeds the body's ability to absorb it, it will cause serious problems for the human body such as liver cancer, diabetes, cirrhosis, heart disease, and infertility. Additionally, high iron content alters the color and taste of natural water, causes yellowing on clothes, and causes water pipe corrosion problems (Kumar *et al.* 2017b). For plants, iron is a vital element, playing a crucial role in plant growth and development. However, when the iron concentration is too high, exceeding the plant's allowable limit, it can reduce productivity. When iron is absorbed by

plants in excess, it becomes phytotoxicity which affects plant growth (Asati *et al.* 2016). Therefore, the iron content in natural iron-alum water needs to be reduced before being used for domestic purposes to ensure people's health and other production activities.

The common methods of treating iron in water include oxidation and precipitation (Bordoloi *et al.* 2013; Boyle *et al.* 1977; Kan *et al.* 2012), zeolite softening/ion exchange (Al-Anber and Al-Anber 2008; Bulai and Cioanca 2011; Dąbrowski *et al.* 2004), calcium carbonate materials (Aziz *et al.* 2004; Mettler *et al.* 2009), filter media separation (Cho 2005; Nemade *et al.* 2008; Vries *et al.* 2017). Among these, the ion exchange is an effective method of removing iron at concentrations less than 5 mg L<sup>-1</sup> (Colter and Mahler 2006), with high selectivity of metals, wide applicable pH range, and high regeneration capacity (O'Connell *et al.* 2008). Besides, the method of using

ion exchange resins is also considered to current development needs. Besides using polymeric frameworks, metal organic frameworks (MOFs) were also being developed (Kumar *et al.* 2017a) with nanostructured compounds (Shehzad *et al.* 2021). This process also demonstrates effectiveness for both wastewater and dilute streams (Sole *et al.* 2018), which is considered a huge advantage of the ion exchange method. It was shown that the current demand for using ion exchange materials is increasing in line with the goal of sustainable development (Ali *et al.* 2023). The ion exchange process is also considered an economic process and does not harm the ecological environment. Besides, through physical and chemical means, it is possible to modify ion exchange materials to increase the exchange surface area, active sites and functional groups (SenGupta 2017). Thus, it can be seen that the ion exchange method is considered a highly effective method for treating harmful substances in water (Barman *et al.* 2023). Moreover, it can separate and recover iron ions and other ionic metals (e.g., aluminum, calcium, and magnesium) in natural iron-alum water to avoid secondary pollution from the treatment process (Tran *et al.* 2022). In addition, it is possible to take advantage of iron ions and other ions separated through regeneration solutions to study and synthesize materials capable of treating other environmental pollution. This research helps avoid wasting natural resources and minimize the harmful effects of these metals in surface water.

The treatment of metals in water using the ion exchange method has been widely carried out in research and industry (Ali *et al.* 2023; Hubicki and Kołodyńska 2012). In this study, 225H resin beads were applied for the first time in the recovery of iron in naturally iron-alum water. In particular, the use of Design Expert 12 optimization software to find the values of parameters (pH, flow rate, and resin layer height) affecting the iron content in naturally iron-alum water after passing through the ion exchange column has not been done in previous studies. This is considered a novelty compared to previous studies in the treatment (recovery) of metal ions in water in column or batch form (Dąbrowski *et al.* 2004; Jasim and Ajjam 2024; Rajoria *et al.* 2022; Verbych *et al.* 2005). The method of applying this optimization software can help find the values of the parameters more accurately than the method of surveying each parameter individually (without using software), as it simultaneously evaluates the influence of many parameters on the iron content after the treatment process. Therefore, in this study, the method of treating natural iron-alum water with ion exchange resins was implemented to treat the water sources with high iron

content to reach iron content lower than 0.5 mg L<sup>-1</sup> after the ion exchange process (according to National Technical Regulation on Surface Water Quality of Vietnam). In this study, the survey of factors affecting ion exchange efficiency (height of ion exchange resin layer (cm), pH value, and flow rate of natural iron-alum water passing through the ion exchange column) using Design Expert 12 optimization software in Tri Ton district, An Giang province was carried out for the first time. In which, 225H ion exchange resin originating from India was first used to treat iron in iron-alum water at this location. This study used 225H resin beads from India (Total exchange capacity: 1.8 meq mL<sup>-1</sup>, minimum; Functional group: sulphonic acid; Ionic form as supplied: hydrogen, H<sup>+</sup>; Appearance: golden yellow to brown beads), which is a type of resin beads commonly used in Vietnam, with a price that can compete with other types of ion exchange resin beads currently used on the market. The use of resin beads with an H<sup>+</sup> ion exchange base aims to minimize the impact on water sources after the ion exchange process (only a small change in pH value). Design Expert 12 is an experimental design software that evaluates the suitability of models with reality, while also determining optimal 'conditions, thereby reducing space, time, and research costs. When the iron content in natural iron-alum water exceeds the standard for iron content in surface water, it is necessary to use this optimized Design Expert 12 software to simultaneously evaluate all three factors affecting the efficiency of iron ion exchange in water (height of ion exchange resin layer, pH value, and flow rate) instead of evaluating each factor individually with different value ranges of each factor using a continuous flow test model. Following the manufacturer's recommendations, ion exchange process in column form is selected, a process commonly practiced today for treating water hardness. Within the limits of this report, the researchers only conducted a survey of these 3 factors, which were considered the main factors affecting the efficiency of the ion exchange process of 225H resin mentioned by the manufacturer. The survey selection values were also based on the manufacturer's recommendations for the water hardness treatment process (Ion Exchange Ltd (India)). At the same time, the research has also determined the optimal parameters of the iron treatment process in natural iron-alum water.

## MATERIALS AND METHODS

### Chemicals

Chemicals used include: HCl (36% w/w), H<sub>2</sub>SO<sub>4</sub> (98% w/w), HNO<sub>3</sub> (65% w/w), NaOH, NH<sub>4</sub>Cl, ethylenediamine tetraacetic acid disodium salt (C<sub>10</sub>H<sub>14</sub>N<sub>2</sub>O<sub>8</sub>Na<sub>2</sub>·2H<sub>2</sub>O),

$\text{CH}_3\text{COONH}_4$ ,  $\text{NH}_2\text{OH}\cdot\text{HCl}$ ,  $\text{CaCl}_2$ ,  $\text{MgCl}_2$ , 1,10-phenanthroline chloride monohydrate ( $\text{C}_{12}\text{H}_9\text{ClIN}_2\cdot\text{H}_2\text{O}$ ), potassium peroxydisulfate ( $\text{K}_2\text{S}_2\text{O}_8$ ), iron standard solution ( $1000 \text{ mg L}^{-1}$ ), aluminum standard solution ( $1000 \text{ mg L}^{-1}$ ) were all from Merck. Deionized water was produced from the laboratory. Ion exchange resin 225H was manufactured by Ion Exchange (India) Ltd.

### **Ion Exchange Test**

The experimental model consists of a round, cylindrical plastic pipe with a diameter of 4.2 cm and a height of 1.5 m, along with a pump capable of producing a flow rate ranging from  $10 \text{ L h}^{-1}$  to  $50 \text{ L h}^{-1}$ . The natural iron-alum water was taken directly from the surface water source and passed through a coarse filter screen (Whatman filter paper 101, 20-25  $\mu\text{m}$ ) to remove suspended sediments before being passed directly through the column. The solution pH was adjusted using HCl and NaOH solution. After running through the column, the natural iron-alum water sample was analyzed for iron and other metal ion contents. Iron content was analyzed according to TCVN 6177:1996 (ISO 6332: 1988 (E), Water quality- Determination of iron- Spectrometric method using 1,10-phenantrolin) and aluminum content was analyzed by atomic absorption spectroscopy (AAS) (Model: AAnalyst 400; Hollow cathode lamps (HCLs) and Electrodeless discharge lamps (EDLs); Manufacturer: Pekin Elmer-USA). Calcium and magnesium were analyzed by the Ca and Mg analysis method according to TCVN 6224- 1996 (ISO 6059 : 1984 (E), Water quality- Determination of the sum of calcium and magnesium- EDTA titrimetric method).

This study was conducted in June 2023, consisting of two parts. In the first part, the study investigated individual factors, namely ion exchange layer height, pH, and flow rate, that affect the remaining iron ion content in natural iron-alum water. In the second part, based on the optimal values from the process of surveying factors (material layer height, pH value, and inlet flow rate of ion exchange column) affecting the ion exchange efficiency of iron in water to serve as the basis for designing the

values of each factor in the optimization software Design Expert 12. These parameters were the independent variables and the remaining iron content in natural iron-alum water was the response variable (**Table 1**).

The model was set up with 20 real experiments and was distributed at different levels ( $-\alpha$ ,  $-1$ ,  $0$ ,  $+1$ ,  $+\alpha$ ), which was automatically set by Design Expert 12 software. Of these, six experiments were repeated at the center (the optimal value of the parameters: material layer height, pH value, and water flow rate through the column found from individual investigations) and 14 experiments at the remaining positions to evaluate errors. The quadratic polynomial model for four parameters is expressed as Equation 1:

$$Y = \beta_0 + \sum_{i=1}^3 \beta_i X_i + \sum_{i=1}^3 \beta_{ii} X_i^2 + \sum_{i=1}^3 \sum_{j=i+1}^3 \beta_{ij} X_i X_j \quad (1)$$

Where Y is the remaining iron content in water after treatment.  $\beta_0$  is the zero-order regression coefficient.  $X_i$  is the independent variable  $i$  that affects the objective function Y.  $\beta_i$  is the first-order regression coefficient describing the influence of the factor  $X_i$  on Y.  $\beta_{ii}$  is the interaction regression coefficient describing the influence of the factor  $X_i$  on Y.  $\beta_{ij}$  is the interaction regression coefficient describing the simultaneous influence of the factor  $X_i$  and  $X_j$  on Y.

The assessment of the fit between the provided data and the predictions from the model was done through fit statistics. The analysis of variance (ANOVA) was used to determine the effects of independent variables on the dependent variable in the research model. In calculation, if the p-value  $< 0.05$  and the coefficient of lack of fit  $> 0.05$  then it shows that the second-order model will be suitable for prediction (Sood *et al.* 2012). Finally, the model predicting the iron content results after ion exchange was tested by evaluating real samples based on the parameters designed from the predictive model using the optimization software Design Expert 12. In which, the study used three actual samples randomly taken in Vinh Phuoc commune, Tri Ton district, An Giang province, where the iron and metal content in naturally

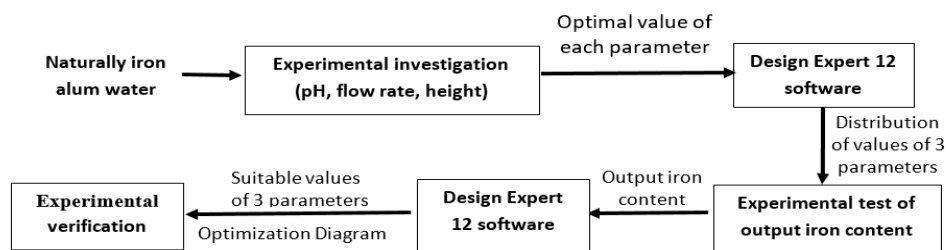


Figure 1. The flow diagram for the optimization of iron treatment by the ion exchange method.

Table 1. The range limits and variation levels of three factors (bed height, pH value, and flow rate).

| Independent Variable<br>(encoding) | Unit              | The Range of Values and Level of the Independent Variable |    |     |     |           |
|------------------------------------|-------------------|-----------------------------------------------------------|----|-----|-----|-----------|
|                                    |                   | $-\alpha$                                                 | -1 | 0   | +1  | $+\alpha$ |
| Height ( $x_1$ )                   | cm                | 46                                                        | 60 | 80  | 100 | 114       |
| pH ( $x_2$ )                       | -                 | 5.65                                                      | 6  | 6.5 | 7   | 7.35      |
| Flow ( $x_3$ )                     | L h <sup>-1</sup> | 13                                                        | 20 | 30  | 40  | 47        |

iron-alum water exceeds the permissible standard and is distributed over a wide area (Nguyen *et al.* 2024).

## RESULTS AND DISCUSSION

### Survey of Background Factors

In this experiment, the effect of flow rate (L h<sup>-1</sup>) on the removal of iron ions in natural iron-alum water by ion exchange resin 225H was investigated, with an iron ion content in the input water of 8.25 mg L<sup>-1</sup>, the height of the resin layer of 100 cm (diameter of 4.2 cm), and the pH of 6.5. It was when the flow rate was less than or equal to 30 L h<sup>-1</sup> that the remaining iron content in natural iron-alum water was less than 0.5 mg L<sup>-1</sup> to ensure that the iron content in water always meets Vietnam's surface water standards (QCVN 08-2023/BTNMT) (Figure 2). This can be because increasing the height of the ion exchange resin layer will prolong the contact time between iron ions in naturally iron-alum water and the resin, leading to increased efficiency in the iron removal process in water (Nur *et al.* 2015; Pirsaeheb *et al.* 2017), which is also consistent with the reference results (CaCO<sub>3</sub> removal efficiency by ion exchange resin) from the manufacturer (Ion Exchange Ltd, India).

When the flow rate is higher, there may not be enough contact time between iron ions and resin particles, leading

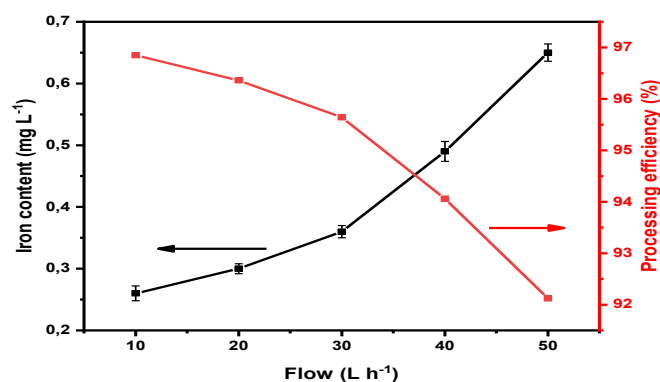


Figure 2. Effect of flow rate on ion exchange capacity of 225H resin for iron ions. (Experimental conditions: iron content of 8.25 mg L<sup>-1</sup>; height of the resin layer of 100 cm (diameter of 4.2 cm); the pH of 6.5; flow rate of 10-50 L h<sup>-1</sup> and number of repetitions (n=3))

to a reduced ion exchange efficiency. From there, it can be seen that the smaller the flow rate (i.e., the longer the contact time), the greater the ion exchange efficiency. When the flow rate was 30 L h<sup>-1</sup>, a survey of pH values ranging from 3.5-8 was conducted to investigate the effect of water pH on the iron removal efficiency of natural iron-alum water iron using the ion exchange model. In particular, when the pH value is greater than 6.5, the efficiency of the exchange process gradually decreases and the exchange efficiency decreases clearly at pH 8 (Figure 3). Thus, the reduced treatment efficiency is because the iron ions in natural iron-alum water may have changed from a dissolved state to a precipitated state, so the ion exchange process cannot be carried out and these precipitates stick to the surface of the ion exchange resin, causing interference (expanding the phase diagram of iron ions in the solution). Previous studies have shown that when the pH value is greater than 8.5, the ferrous iron form is transformed into the solid Fe(OH)<sub>2</sub> form (precipitate) (Morato-Lara 1998; Nkumanda 2017; Skousen 2014; Zhao 2004), which can cover the ion exchange centers on the surface of the ion exchange resin, making the exchange process difficult, directly affecting the efficiency of separating iron ions from water.

In the next experiment, the influence of ion exchange layer height was investigated at pH 6.5 and a flow rate of 30 L h<sup>-1</sup>, within the investigation range from 20 to 120 cm (Figure 4). When the ion exchange resin layer was larger than 80 cm, the efficiency of the ion exchange process reached over 90%, and the remaining iron content in natural iron-alum water was less than 0.5 mg L<sup>-1</sup>, this is also very close to the manufacturer's recommended conditions (0.75 m with co-flow regeneration and 1 m with counter-current regeneration). It can be seen that increasing the height of the ion exchange resin layer increases the ion exchange centers through the ion exchange column, which will help increase the efficiency of iron ion exchange in water, making the iron content after removal is lower and this has also been shown in previous studies (Nur *et al.* 2015; Pirsaeheb *et al.* 2017). From this, it can be seen that the appropriate height of the ion exchange resin layer selected for the iron treatment process in natural iron-alum water to achieve an iron content after ion exchange of less than 0.5 mg L<sup>-1</sup> (according to the iron content standard in

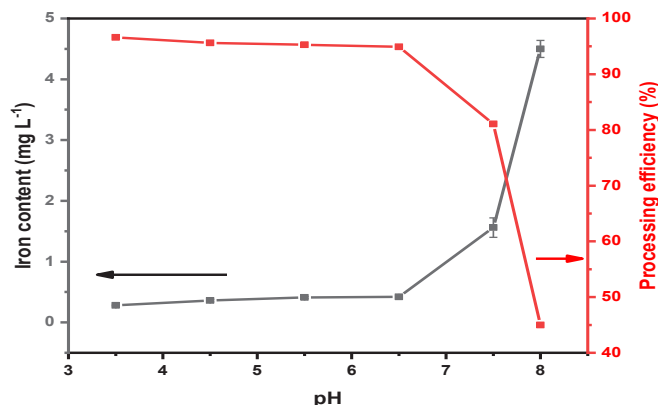


Figure 3. Effect of pH on iron treatment ability of ion exchange 225H resin (Experimental conditions: iron content of 8.25 mg L<sup>-1</sup>; height of the resin layer of 100 cm (diameter of 4.2 cm); pH of 3.5-8; flow rate of 30 L h<sup>-1</sup> and number of repetitions (n=3)).

surface water of Vietnam) was 80 cm (0.8 m).

After surveying three factors affecting the ion exchange efficiency of 225H resin beads for iron ions in natural iron-alum water, the appropriate conditions were selected as follows: pH value 6.5, flow rate 30 L h<sup>-1</sup>, material layer height 80 cm. This is because when applying the above-selected values to the iron ion exchange process, the iron content in iron-alum water after the ion exchange process was lower than 0.5 mg L<sup>-1</sup> (ensuring the iron content standard in surface water of Vietnam). A survey to evaluate the effectiveness of iron treatment in natural iron-alum water using the selected parameters showed that the remaining iron content after treatment was 0.42 mg L<sup>-1</sup>, with a treatment efficiency of 94.91%. Besides, the aluminum treatment efficiency reached 87.9%, while calcium and magnesium were not found after the treatment process (Table 2).

### Statistical Analysis and Assessment of Model Suitability

A target design with 20 experiments based on software calculations with six central repetitions to test

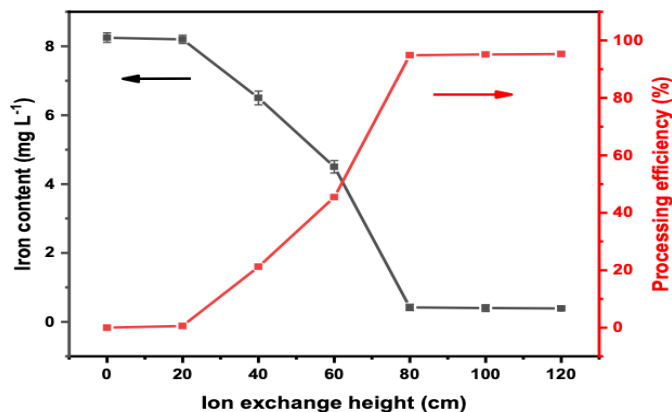


Figure 4. Effect of layer height on iron treatment ability of ion exchange resin (Experimental conditions: iron content of 8.25 mg L<sup>-1</sup>; height of the resin layer of 10-120 cm (diameter of 4.2 cm); pH of 6.5; flow rate of 30 L h<sup>-1</sup> and number of repetitions (n=3)).

the influence of factors: resin layer height, flow rate, and solution pH on iron content remaining after the ion exchange process. The results of the investigation of individual factors were set to the central value of 20 experiments (Table 3).

The F-value was 48.02 with a p-value < 0.0001, indicating that the model is statistically significant, as the p-value is below the commonly used threshold of 0.05. The parameters A, C, and A<sup>2</sup> indicated importance with the p-value < 0.05. Therefore, the model can be considered to eliminate the remaining unimportant parameters (B, AB, AC, BC, B<sup>2</sup>, and C<sup>2</sup>). This was because the p-values (Table 4) of the above parameters were greater than 0.05, which made the parameters insignificant in the multivariate quadratic model of Design Expert 12 software.

The correlation coefficient R<sup>2</sup> was a measure of model fit. According to the software analysis results, the value R<sup>2</sup> = 0.9774 has been obtained. This value is very close to 1, which shows that the experimental polynomial model calculated from the software is consistent with the actual experimental parameters previously performed. The predicted correlation coefficient R<sup>2</sup> = 0.8183, which is consistent with an adjusted R<sup>2</sup> correlation coefficient

Table 2. The treatment efficiency of iron and other cations in natural iron-alum water by ion exchange column 225H (n=3).

| Parameters                                                                                                 | Ion Metal |           |           |           |
|------------------------------------------------------------------------------------------------------------|-----------|-----------|-----------|-----------|
|                                                                                                            | Iron      | Aluminum  | Calcium   | Magnesium |
| Input water (mg L <sup>-1</sup> )                                                                          | 8.25±0.18 | 1.24±0.06 | 0.68±0.08 | 0.52±0.10 |
| After treatment (mg L <sup>-1</sup> )                                                                      | 0.42±0.04 | 0.15±0.05 | <0.01     | <0.01     |
| Treatment efficiency (%)                                                                                   | 94.91     | 87.9      | 100       | 100       |
| Maximum value of iron content affecting human health according to QCVN 08:2023/BTNMT (mg L <sup>-1</sup> ) | 0.5       |           |           |           |

Calcium and magnesium contents were analyzed by the analytical method according to TCVN 6224 – 1996 (Standard Method 2320B:2005)

Table 3. The iron removal efficiency of the ion exchange column based on the values of three parameters designed by Design Expert 12 software).

| No. | Height<br>(X <sub>1</sub> ) | pH<br>(X <sub>2</sub> ) | Flow Rate<br>(X <sub>3</sub> ) | Output Iron Content<br>(Y) |
|-----|-----------------------------|-------------------------|--------------------------------|----------------------------|
|     | cm                          | -                       | L h <sup>-1</sup>              | mg L <sup>-1</sup>         |
| 1   | 60                          | 6                       | 20                             | 1.2                        |
| 2   | 100                         | 6                       | 20                             | 0.26                       |
| 3   | 60                          | 7                       | 20                             | 1.02                       |
| 4   | 100                         | 7                       | 20                             | 0.28                       |
| 5   | 60                          | 6                       | 40                             | 1.3                        |
| 6   | 100                         | 6                       | 40                             | 0.3                        |
| 7   | 60                          | 7                       | 40                             | 1.7                        |
| 8   | 100                         | 7                       | 40                             | 0.32                       |
| 9   | 46                          | 6.5                     | 30                             | 2.2                        |
| 10  | 114                         | 6.5                     | 30                             | 0.15                       |
| 11  | 80                          | 5.65                    | 30                             | 0.38                       |
| 12  | 80                          | 7.35                    | 30                             | 0.44                       |
| 13  | 80                          | 6.5                     | 13                             | 0.28                       |
| 14  | 80                          | 6.5                     | 47                             | 0.52                       |
| 15  | 80                          | 6.5                     | 30                             | 0.41                       |
| 16  | 80                          | 6.5                     | 30                             | 0.39                       |
| 17  | 80                          | 6.5                     | 30                             | 0.41                       |
| 18  | 80                          | 6.5                     | 30                             | 0.4                        |
| 19  | 80                          | 6.5                     | 30                             | 0.41                       |
| 20  | 80                          | 6.5                     | 30                             | 0.41                       |

Note: The values X<sub>1</sub>, X<sub>2</sub>, and X<sub>3</sub> have been set by the software; Y was the result of the remaining iron content after each experiment.

response (iron content after the process in the water after the column) for certain levels of each factor. In addition, the values must be expressed in the original units for each parameter. In addition, this equation should not be used to determine the relative impact of each factor because the coefficients are scaled to match the units of each factor.

$$\begin{aligned} \text{Iron content} = & 12.10120 - 0.116467 \times \text{Height} - \\ & 1.62699 \times \text{pH} - 0.064409 \times \text{Flow rate} - \\ & 0.002250 \times \text{Height} \times \text{pH} - 0.000437 \times \text{Height} \times \\ & \text{Flow rate} + 0.014500 \times \text{pH} \times \text{Flow rate} + \\ & 0.000730 \times \text{Height}^2 + 0.109579 \times \text{pH}^2 + \\ & 0.000239 \times \text{Flow rate}^2 \end{aligned} \quad (2)$$

The coefficient table contains a row with the predicted values of the parameters for each response (iron content after passing the column) (Table 6). Each column contains the coefficient estimate for the model term coded for that particular parameter and the p-value for that coefficient. This coefficient describes the size and direction of the relationship between a predictor and a response variable. Coefficients are the numbers by which the values of the terms are multiplied to form a regression equation. In this case, p-values < 0.01 are highly significant, 0.01 < p-values < 0.05 are significant, 0.05 < p-values < 0.10 are marginally significant and p-values ≥ 0.10 are considered insignificant. P-values less than 0.10 but greater than

Table 4. The ANOVA analysis results for multivariate quadratic model.

| Source         | Sum of squares | df | Mean square | F-value | p-value  |             |
|----------------|----------------|----|-------------|---------|----------|-------------|
| Model          | 5.64           | 9  | 0.6262      | 48.02   | < 0.0001 | significant |
| A-Height       | 4.13           | 1  | 4.13        | 316.78  | < 0.0001 |             |
| B-pH           | 0.0095         | 1  | 0.0095      | 0.7292  | 0.4131   |             |
| C-Flow rate    | 0.1167         | 1  | 0.1167      | 8.95    | 0.0135   |             |
| AB             | 0.0041         | 1  | 0.0041      | 0.3106  | 0.5896   |             |
| AC             | 0.0613         | 1  | 0.0613      | 4.70    | 0.0554   |             |
| BC             | 0.0421         | 1  | 0.0421      | 3.22    | 0.1028   |             |
| A <sup>2</sup> | 1.27           | 1  | 1.27        | 97.38   | < 0.0001 |             |
| B <sup>2</sup> | 0.0112         | 1  | 0.0112      | 0.8565  | 0.3765   |             |
| C <sup>2</sup> | 0.0085         | 1  | 0.0085      | 0.6538  | 0.4376   |             |
| Residual       | 0.1304         | 10 | 0.0130      |         |          | significant |
| Lack of Fit    | 0.1301         | 5  | 0.0260      | 371.60  | < 0.0001 |             |
| Pure Error     | 0.0003         | 5  | 0.0001      |         |          |             |
| Cor Total      | 5.77           | 19 |             |         |          |             |

of 0.9570 with a difference of less than 0.2. This was considered a good fit between the model parameters. Additionally, the adequate precision measures the signal-to-noise ratio, and a ratio of 25.416 (a ratio greater than 4 was desirable) indicates an adequate signal (Table 5).

The final equation is calculated from the software based on the actual parameters (2). In this case, this equation can be used to make predictions about the

Table 5. Results from Fit Statistics of Design Expert 12 software for iron ion exchange in water.

|                          |         |
|--------------------------|---------|
| Std. Dev.                | 0.1142  |
| Mean                     | 0.6390  |
| C.V. %                   | 17.87   |
| R <sup>2</sup>           | 0.9774  |
| Adjusted R <sup>2</sup>  | 0.9570  |
| Predicted R <sup>2</sup> | 0.8183  |
| Adequate Precision       | 25.4158 |

Table 6. The predicted values of coefficients of parameters for the response variable (iron ion content after leaving the column) from Design Expert 12 software.

|              | Intercept | A         | B       | C         | AB      | AC      | BC     | A <sup>2</sup> | B <sup>2</sup> | C <sup>2</sup> |
|--------------|-----------|-----------|---------|-----------|---------|---------|--------|----------------|----------------|----------------|
| Iron content | 0,402377  | -0.547533 | 0.02627 | 0.0920174 | -0.0225 | -0.0875 | 0.0725 | 0.292101       | 0.0273948      | 0.0239346      |
| p-values     |           | < 0.0001  | 0.4131  | 0.0135    | 0.5896  | 0.0554  | 0.1028 | < 0.0001       | 0.3765         | 0.4376         |

0.05 are marginally significant. P-values greater than or equal to 0.10 are considered insignificant.

### Results of Response Surface Analysis

Based on the results from 20 experiments, the model's calculation process revealed different impacts of independent variables on the dependent variable (**Figure 5**) as illustrated through the model, three-dimensional surface graphs, and contour plots. Obviously, when the independent variables change values, the objective function also changes, maybe more or less, and this depends on the influence of each independent variable on the dependent variable. The analytical results showed that height and flow rate had a direct influence on the remaining iron content in alum-contaminated water with height having the greatest influence on iron content in the effluent water. This is evident from the ANOVA analysis and the surface response plot. For pH value when, combined with other independent variables, the contour representing these variables is usually a straight line. When considered in terms of statistical significance, these variables have a very small or insignificant effect on the remaining iron content after the ion exchange process.

The design parameters need to be selected in region (b) to ensure the output iron content in water is always below 0.5 mg L<sup>-1</sup> (**Figure 6**). Calculation results from the model show that the optimal value includes pH 6.5, low rate 28.7 L h<sup>-1</sup>, height 76.5 cm, corresponding to

the iron content in the initial alum-water of 8.25 mg L<sup>-1</sup>, with the target function of output iron content being 0.5 mg L<sup>-1</sup>. When applying the parameters found from the model into practice, the result after six repetitions is that the output iron content is always equal to or less than 0.5 mg L<sup>-1</sup>. This result has shown the compatibility of the calculation results obtained from Design Expert 12 software and the experimental process for natural iron-alum water samples based on the values of the parameters (pH, natural iron alum-water flow through the column, and ion exchange resin layer height) from Design Expert 12 software. In addition, insignificant independent variables are considered to be retained in some cases to ensure systematicity for the entire model.

The test at three water sampling locations with different initial iron content in water showed that the iron content out of column was all below 0.5 mg L<sup>-1</sup> (with the conditions at pH 6.5, flow of 30 L h<sup>-1</sup>, height of 82.3 cm and calculated output iron content of 0.34 mg L<sup>-1</sup>), the metal treatment efficiency also reached over 93% for iron and consistently above 85% for aluminum, calcium, and magnesium (**Table 7**). The pH value of actual water samples before and after the ion exchange process was theoretically significantly reduced because H<sup>+</sup> ions replace iron ions in the water. However, in actual water samples, the pH value did not change significantly during the exchange process because the content of iron ions in water was usually very low (usually only a few to more than ten mg L<sup>-1</sup>).

Table 7. Metal content in natural iron-alum water before and after treatment (n=3).

| Water sample                                                                                               | Parameters                            | Metal ions |           |           |           |
|------------------------------------------------------------------------------------------------------------|---------------------------------------|------------|-----------|-----------|-----------|
|                                                                                                            |                                       | Iron       | Aluminum  | Calcium   | Magnesium |
| Location 1                                                                                                 | Input water (mg L <sup>-1</sup> )     | 30.26±0.24 | 2.25±0.15 | 2.06±0.08 | 1.45±0.11 |
|                                                                                                            | After treatment (mg L <sup>-1</sup> ) | 0.35±0.12  | 0.28±0.14 | 0.2±0.06  | 0.15±0.10 |
|                                                                                                            | Treatment efficiency (%)              | 98.84      | 87.56     | 87.86     | 89.86     |
| Location 2                                                                                                 | Input water (mg L <sup>-1</sup> )     | 12.0±0.16  | 1.84±0.10 | 0.96±0.04 | 0.48±0.09 |
|                                                                                                            | After treatment (mg L <sup>-1</sup> ) | 0.34±0.10  | 0.2±0.05  | 0.12±0.04 | <0.01     |
|                                                                                                            | Treatment efficiency (%)              | 97.19      | 85.87     | 87.5      | 100       |
| Location 3                                                                                                 | Input water (mg L <sup>-1</sup> )     | 3.84±0.18  | 0.38±0.12 | 0.45±0.07 | 0.56±0.11 |
|                                                                                                            | After treatment (mg L <sup>-1</sup> ) | 0.25±0.13  | <0.01     | <0.01     | <0.01     |
|                                                                                                            | Treatment efficiency (%)              | 93.49      | 100       | 100       | 100       |
| Maximum value of iron content affecting human health according to QCVN 08:2023/BTNMT (mg L <sup>-1</sup> ) |                                       | 0.5        |           |           |           |

Note: Location 1, 2, and 3 were water samples at 3 different locations containing metals in natural water of acid sulfate soil ;

Note: Ca, Mg, and Al contents were detected by the analytical method according to TCVN 6224 – 1996 (Standard Method 2320B:2005) and AAS.

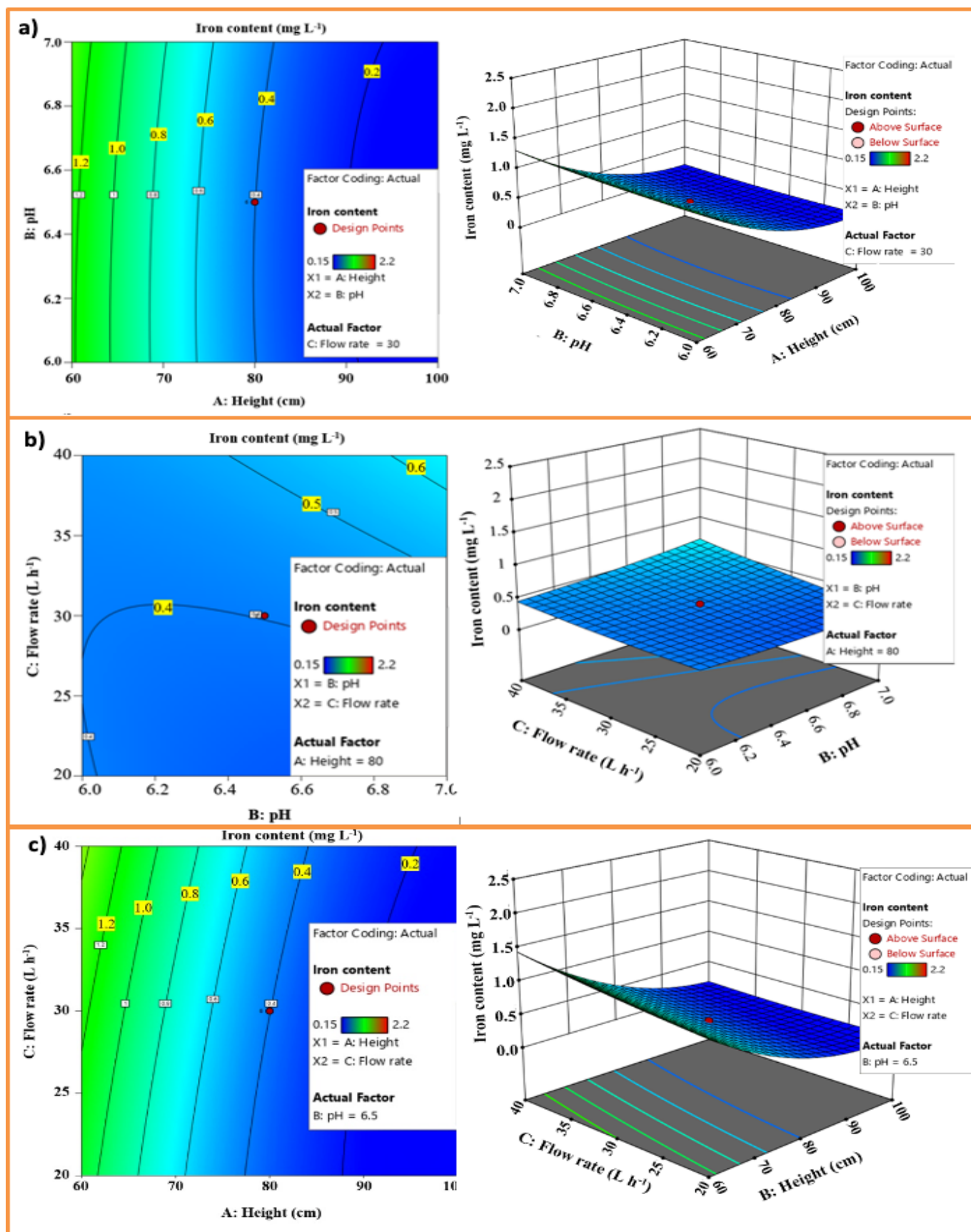


Figure 5. The interactive results are calculated using Design Expert 12 software between the three independent variables and the dependent variable in natural iron-alum water. (includes: a) Describe the correlation of pH and height with effluent iron content; b) Describe the correlation of pH and flow with effluent iron content and c) Describe the correlation of flow and height with effluent iron content)

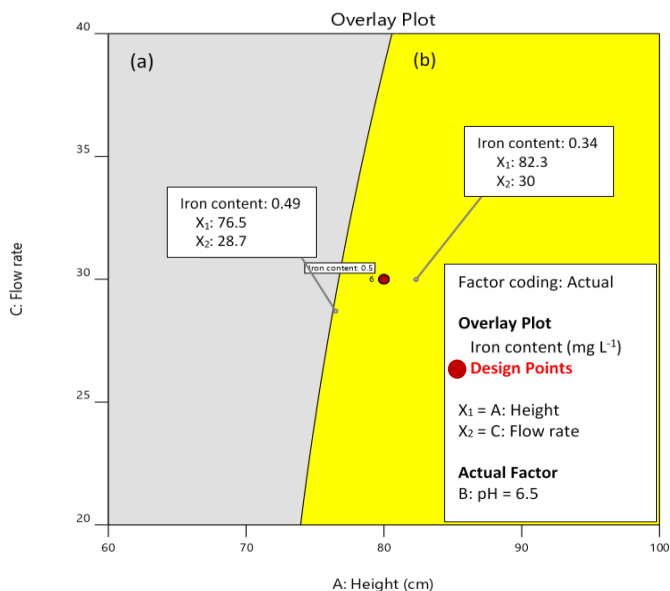


Figure 6. The resulting overlay diagram was calculated from Design Expert 12 software with two zones divided by an iron content of 0.5 mg L<sup>-1</sup>.

## CONCLUSIONS AND RECOMMENDATIONS

Research results have proven that the ion exchange method can be applied to the treatment of iron-alum water, with three factors directly affecting the efficiency of iron ion recovery: flow rate, pH, and exchange layer height. The study also built a multivariate quadratic equation using Design Expert 12 software. The calculation results from the software found the influence of independent variables on the remaining iron content in iron-alum water. In addition, through the calculation process using Design Expert 12 software, the study also showed the influence of each independent variable (flow rate through the column, pH value, and resin layer height) on the target function (residual iron content) after ion exchange (Figure 5). The optimal values of parameters under related constraints (pH 6.5, flow rate of 30 L h<sup>-1</sup>, and height of 82.3 cm) gave an iron removal efficiency of over 93% and the remaining iron content in natural iron-alum water was less than 0.5 mg L<sup>-1</sup>. Additionally, the treatment efficiency of other metals, such as aluminum, calcium, and magnesium was also over 85%. In general, using Design Expert 12 software to optimize the iron treatment process in iron-alum water was very useful, in evaluating the influence of independent variables on the remaining iron content after performing the ion exchange process. This method can also be applied practically with the ability to select the output iron content from the software.

Subsequent studies can be based on these optimal parameters to evaluate the iron removal efficiency of wastewater containing iron and the influence on the

content of other water parameters after treatment with ion exchange resin.

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## **ACKNOWLEDGMENT**

The authors acknowledge the Ho Chi Minh City University of Technology (HCMUT) and An Giang University, VNU-HCM for supporting this research.