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Evaluation and Optimization of Electro-Fenton Process in Removal of Amoxicillin Antibiotic Using Chelating Agents at High pH Values from Aqueous Solutions

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ABSTRACT

Antibiotics are among the most consumed drugs worldwide. These compounds enter the environment from different sources, adversely affecting human and animal health given their high persistence and stability in the environment. Conventional water and wastewater treatment processes have not been designed for removal of such materials. Thus, a suitable method should be devised for removal of these compounds. Among antibiotics, Amoxicillin (AMX) has the minimum metabolism. The aim of this research is the removal of AMX using electro-Fenton (EF) method from aqueous solutions. One of the most important disadvantages of the electro-Fenton method is its high efficiency at acidic pH, causing secondary contamination. As such, for resolving this issue, chelating agents can be used. In this study, the effects of important parameters have been examined on AMX removal efficiency including initial concentration AMX (20-120) mg/L, initial pH (3-7), current intensity (10-130) mA, and EDTA concentration (0-1) mM as chelating agent via design of experiment method. This paper introduces a simplified version of quadratic polynomial Response Surface Methodology (RSM). After analysis of variance, it was found that all examined variables were significant and the model had sufficient validity. The results showed that EDTA in addition to pH has also been influential in enhancing removal efficiency. Regarding the optimal conditions for the examined parameters, the initial concentration of AMX 24 mg/L, the current intensity 85 mA, initial pH 5.6, and EDTA dose 0.6 mM, showed 95.71% AMX removal, eventually. Also, a graphite cathode made of pencil was used for constant production of hydrogen peroxide during the process. The extent of electric energy consumption at the optimal point was obtained 0.86 KWh/m^3.

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INTRODUCTION

Water pollution caused by human activities is a major global issue that poses a constant and widespread threat. The continuous release of organic and inorganic materials into aquatic environments has resulted in a decline in water quality, putting the survival and sustainability of living beings at risk. Industrial waste, chemicals, and pharmaceutical residues are some of the substances regularly discharged with wastewater streams (Brillas, 2020). Antibiotics are powerful therapeutic agents that are widely used and consumed throughout the world. These chemotherapeutic compounds specifically target bacterial cells by either destroying them or inhibiting their growth (Kümmerer, 2009; Aleksic et al., 2021). Antibiotics are extensively used in human medicine, veterinary medicine, and agriculture, resulting in their persistent release into the environment from anthropogenic sources (Michael et al., 2013). These compounds

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*Corresponding Author Email: *m.salari@sirjantech.ac.ir* possess traits such as stability, lipophilicity, and poor degradability. They are discharged through different pathways, including municipal and hospital wastewater treatment plants, runoff, landfill leachate, and fishpond leachate. Eventually, pollute the groundwater, they infiltrate water and soil systems either directly or indirectly (Vaou et al., 2021; Zhang et al., 2016).

Activated sludge and other traditional water and wastewater treatment methods are not designed to remove pharmaceuticals like antibiotics. This highlights the need for an effective and safe technique to address these pollutants (Shokoohi et al., 2020). Antibiotics are known to persist in the environment, which raises concerns about their potential harm to human health. As a result, innovative technologies must be utilized to remediate this type of pollutant (Basturk et al., 2021). Amoxicillin (AMX) is a penicillin class antibiotic used in human and veterinary medicine to treat gastrointestinal bacterial infections (Aranda & Rivas, 2022). Its chemical formula is $C_{16}H_{19}N_3O_5S$, and it has a molecular weight of 365.41 g/mol (Aranda & Rivas, 2022). The Electrochemical Advanced Oxidation Process (EAOP) is the most effective method for removing antibiotics from aquatic systems, according to various studies (Rahmatinia et al., 2016). The Electro-Fenton (E.F.) process is an advanced electrochemical oxidation process that is highly efficient and flexible, environmentally friendly, and cost-effective (Deng et al., 2020). It uses an electrode and an electrical source to provide divalent iron for the reaction, making it less costly than other methods that require more divalent iron consumption. Furthermore, it produces less sludge due to reversible conversion of divalent iron to its trivalent form, making it more desirable than other processes that produce more sludge (Jiang, 2022, Lama et al., 2022). One of the biggest challenges of traditional Fenton and Electro Fenton processes is that low pH is required due to the precipitation of Fe II and Fe III at high pH values (Shoorangiz et aL., 2019, Salari et al., 2021). This leads to iron sludge production, which limits the usefulness of homogeneous electro Fenton. However, this issue can be addressed by adding chemicals, with one possible solution being to modify the process using chelating agents to make it viable at high pH values. Numerous studies have been conducted in this area. Basturk et al. (2020) investigated removing antibiotics from medical laboratory wastewater using the electro-Fenton process. They reported a removal efficiency of 99.12%, 98.65%, and 99.38% for cephalexin, ciprofloxacin, and clarithromycin, respectively (Basturk et al., 2021). Deng et al. (2020) proposed an innovative electro-Fenton system that overcomes pH-related limitations using a nickel bottom cathode (Ni-F) and a tripolyphosphate (3-PP) electrolyte with near-neutral pH. Their study demonstrated how tripolyphosphate can boost hydrogen peroxide. Moreover, the research underscores the potential of the nickel bottom cathode as a suitable option for electro-Fenton under near-neutral circumstances when tripolyphosphate electrolyte is present. This represents an innovative addition to the field. (Deng et al., 2020). Kadji et al. (2020) conducted a study on the electro-Fenton process with a graphite cathode to remove AMX from an aqueous medium. Their findings showed that 95% decomposition and 74% mineralization of the pollutant could be achieved under optimal conditions, including a current intensity of 600 mA, a temperature of 25° C, an AMX concentration of 0.082 mmol/L, and a divalent iron concentration of 1 mmol/L (Kadji et al., 2021). Salari (2022) recently investigated the effectiveness of magnetite nanoparticles in removing ciprofloxacin (CIP) from aqueous solutions. The study tested various independent variables such as the initial concentration of ciprofloxacin, adsorbent dose, pH values, and reaction time to determine their effect on the percentage of CIP removal (Salari, 2022). Varindani et al. (2021) conducted research on the modified Electro Fenton process for treating mixed industrial wastewater. The Electro Fenton process was modified by introducing ethylenediaminetetraacetic acid (EDTA) as a chelating agent to enhance the Fenton reaction at a nearly neutral pH. To achieve this, a titanium electrode coated with a nanocomposite was used as the anode while graphite served as the cathode. The results showed that the electro-Fenton process with the chelate agent achieved 67% COD removal at near-neutral pH, which is comparable to the electro-Fenton process (66%) performed at acidic pH. In their study on AMX removal using electro-Fenton, Zhang et al. (2021) found that key operational parameters such as divalent iron concentration, pH, applied current, and initial concentration of AMX significantly affected degradation efficiency. Applying the response surface method (RSM) under optimal conditions of 10 mg/L AMX at pH 2.29, applied current of 366.08 mA, and ferrous sulfate concentration of 26.18 mg/L resulted in a 96.97% removal percentage of AMX (Zhang et al., 2021). In 2022, Salari et al. conducted a study to optimize the homogeneous Fenton-Like process for high initial pH. By optimizing simultaneously with EDTA, they found that a pH of around 7, an initial CIP concentration of 14.9 mg/L, a dose of divalent iron of 9.2 mmol, a ratio of divalent iron to hydrogen peroxide of 3.2, and an EDTA concentration of 6.60 mM over a 25-min period resulted in an 85.2% removal rate of ciprofloxacin and a predicted sludge-to-iron ratio of 2.24 g/mol (Salari et al., 2022). Previous studies have shown that chelating agents have various practical applications such as breaking down metal ions, inhibiting metal-catalyzed reactions, eliminating metal ions and increasing metal accessibility. Moreover, it seems that at a pH level of 6 to 7, introducing chelating agents generates stable clusters with iron ions. This makes the ions available for reacting with hydrogen peroxide, leading to the production of hydroxyl radicals. This reaction somewhat hinders the precipitation of iron ions (Messele et al., 2014).

The purpose of this study is to utilize experimental design techniques in a laboratory study to eradicate the antibiotic AMX from high pH water settings. The focus is on utilizing a chelating agent to improve the efficiency of antibiotic elimination. Furthermore, the research seeks to analyze the influence of important factors such as pollutant concentration, current intensity, initial pH, and chelating agent quantity on the most effective removal conditions, as well as to assess energy usage throughout the procedure.

MATERIALS AND METHODS

Reagents

The chemical materials and the sources of their supply are shown in the table 1.

Electro-Fenton (EF) Process

The electro-Fenton process represents an advanced oxidation method predicated on the Fenton reaction. The traditional Fenton method involves the direct addition of hydrogen peroxide (H_2O_2) and ferrous ions (Fe^{2+}) to the reaction. However, in electro-Fenton, H_2O_2 is generated through oxygen reduction at the cathode according to Equation (1), while a small quantity of iron is incorporated into the solution in the form of iron salt. Catalysts, such as Fe^{2+}, Fe^{3+}, or iron oxides, interact with H_2O_2 to initiate the Fenton reaction and produce hydroxyl ($O.H^0$) radicals. Fe^{3+} ions, which arise from the Fenton reaction, are subsequently converted into Fe^{2+} ions, catalyzing the production of $O.H^0$ from the Fenton reaction.

$$O_2+2H^++2e^-\rightarrow H_2O_2 \tag{1}$$

The EF process relies on the continuous production of oxygen, as detailed in relation (1),

Table 1. Type of used chemical material and the sources of their supply

Material	Source of the supply
AMX ($C_{16}H_{19}N_3O_5S$-$3H_2O$, 99% in purity)	Iran Pharmaceutical Company, (Iran)
Sodium hydroxide (NaOH, 99% in purity)	Kian Kaveh Azma Chemical and Pharmaceutical Industries Company, (Iran)
Sulfuric acid (H_2SO_4, 95e98%)	Merck Company, (Germany)
Iron sulfate ($FeSO_4\cdot 7H_2O$)	Merck Company, (Germany)
Ethylene diamine tetraacetic acid ($C_{10}H_{16}N_2O_8$, EDTA, 99%)	Bou Ali Sina Chemical Company (Iran)

which is facilitated by an air pump throughout the experiment period or via the electrolysis of water and dissolved oxygen at the anode, as demonstrated by reaction (2). The requisite Fe^{2+} ions, necessary for the Fenton reaction to occur at the cathode surface, are derived from the reduction of Fe^{3+} ions, as expressed with Eqs (3-4). (Matyszczak et al., 2020).

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (2)$$

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (3)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH\bullet + OH^- \quad (4)$$

The resulting highly reactive oxidant radical (OH•) ultimately facilitates the transformation of unsaturated compounds or aromatic pollutants into minerals, resulting in their conversion to CO_2, H_2O, and inorganic ions. The EF process has proven to be effective in removal organic pollutants from water, exhibiting a high rate of oxidation or mineralization (Babuponnusami & Muthukumar, 2012; Matyszczak et al., 2020; Panizza & Cerisola, 2009; He & Zhou, 2017).

$$R + OH\bullet \rightarrow CO_2 + H_2O + \text{Inorganic-matter} \quad (5)$$

The current investigation involves the production of iron ions through the addition of a salt, while hydrogen peroxide is generated using a graphite cathode fashioned from a pencil core.

Chelating agents

A majority of previous studies exploring the removal of AMX without the introduction of a specific quantity of chelating agent have reported an optimal pH level of roughly 3.5 (Verma et al., 2019; Ay and Kargi, 2010). Chelating agents can create chemical bonds with iron ions and establish complexes with Fe(II) and Fe(III) iron ions under high pH conditions, maintaining their solubility and preventing precipitation, thereby promoting oxidation (Zhang & Zhou, 2019).

The present study employs the chelating agent EDTA to form complexes with metal ions, particularly those that are strongly adsorbed by transition metals. EDTA finds widespread use in various industries, including textiles, pulp and paper, and food production (Zhang & Zhou, 2019). The implementation of chelators in oxidation procedures represents a novel approach. Chelating agents are organic compounds characterized by multiple rings, displaying a strong affinity for binding with metal elements.

Modeling by method (CCD)

The CCD method is the most widely used response surface method and it is a rotatable method. In this study, modeling is done using the CCD method by Design-Expert® software to design the experiment table and predict the responses, find the mutual effects of the variables and determine the optimal parameters. For further study of this method, can refer to the references (Eslami et al., 2016; Bezerra et al., 2008).

Selection of parameters affecting the process and preliminary experiments

Before beginning the experiment, it is important to select the variables that affect the EF process and determine their investigated intervals. Table 2, provides this information based on a review of past studies and initial experiments of the variables (Salari et al., 2022; Shoorangiz et al., 2019).

Additionally, preliminary experiments were conducted to choose between sodium chloride

Table 2 The actual and coded values designed for the independent variables in CCD experiments

Coded variable	**Description**	**Experimental Field**				
		-∝	**-1**	**0**	**1**	**+∝**
X_1	A: Concentration of AMX (mg/L)	20	45	70	95	120
X_2	B: Initial pH	3	4	5	6	7
X_3	C: Current intensity (mA)	10	40	70	100	130
X_4	D: Concentration of EDTA (mM)	0	0.25	0.50	0.75	1

(NaCl) and sodium sulfate (Na_2SO_4) as the electrolyte. The results indicated that sodium chloride has a higher pollutant removal percentage. The experiment was conducted for 15 minutes with a pH of 3, an electrolyte concentration of 0.05 mol, a current of 40 mA, and an initial AMX concentration of 10 mg/L.

Other study by Yousefian et al. (2017) examining the energy consumption required to remove ciprofloxacin with various electrolytes, it was found that sodium chloride had the lowest energy consumption among all electrolyte types (Yoosefian et al., 2017).

To determine the optimal concentration of $FeSO_4{\cdot}7H_2O$, an experiment was conducted for 15 min at pH 3, with sodium chloride electrolyte concentration of 0.05 mol, current intensity of 40 mA, and an initial AMX concentration of 10 mg/L. The results revealed that a concentration of 30 mg/L provided the highest AMX removal percentage.

Analytical procedures

To determine the maximum absorption of AMX (λ_{max}), a concentration of 10 mg/L of AMX was prepared and measured using a spectrophotometer. This process was repeated three times, and λ_{max} was found to be 203 nm in all three trials. Table (SI) outlines the design of the AMX antibiotic removal experiment using CCD.

This study employed an electrochemical reactor constructed from plexiglas with dimensions of 55 x 55 x 70 mm, as shown in Fig SI. The reactor was covered with aluminum sheeting to prevent the intrusion of light and to ensure the accuracy of the experimental results. The distance between the electrodes within the reactor was 3 cm, with a sample volume of 100 cm^3. All experiments were performed at room temperature and air pressure. Initially, 100 cm^3 of contaminated wastewater with a predetermined concentration was introduced into the reactor. Subsequently, 30 mg/L of $FeSO_4{\cdot}7H_2O$ was added to the solution, followed by 0.05 mol (NaCl) as an electrolyte. In all experiments, the duration of the reaction was 30 min, and a magnetic stirrer operating at 200 rpm was utilized to prevent the sedimentation of the suspension during the experiment. Upon completion of the reaction time, the sample was centrifuged at 30,000 rpm for 4 min to settle the suspended material and prepare the sample for the measurement of the remaining antibiotic.

To measure the residual concentration of the samples, a calibration curve was constructed by plotting the absorption values of five different concentrations of antibiotics. The line equation obtained from the calibration curve used to calculate the residual concentration of AMX in the samples. The effect of filtration on the removal of pollutants was examined by filtering the sample through a 0.45 μm filter, which showed that filtration had a negligible effect on the removal of A (less than 1%). To determine the absorption values, the sample was transferred to a cuvette and analyzed using a spectrophotometer at a wavelength of 203 nm, which corresponded to the maximum absorption of AMX. Finally, the residual concentration and the AMX removal percentage was determined using Eq (6).

$$CIP_{removal\,(\%)} = (\frac{C_0 - C_f}{C_0}) \times 100 \tag{6}$$

Where C_0 and C_f are initial and final concentrations (mg/L) of AMX in the solution based on wavelength λ_{max}= 203 nm, respectively.

The cathode employed in this study was constructed using 25 pencil tips with a diameter of 2mm, as illustrated in Fig 2. The tips were joined to the device using solder and had an effective surface area of 16.55 Cm^2 in the experiment solution. The titanium anode used in the experiment was depicted in Fig 1, and its effective surface area of 16.55 Cm^2 was immersed in the solution.

Electrical energy consumption (EEC)

The energy consumption (kWh/m³) at the optimal point was calculated using Eq (7).

$$\mathrm{EEC} = \frac{U\,I\,t}{V} \tag{7}$$

Where (U) the voltage in volts, (I) the current in A, (t) the time in hr, and (V) the volume of the reactor L, as outlined in the study conducted by Yoosefian et al., (2017).

Electrical energy consumption (ELC)

Eq (8) can be utilized to determine the electrode consumption under optimal conditions, expressed in gr.v

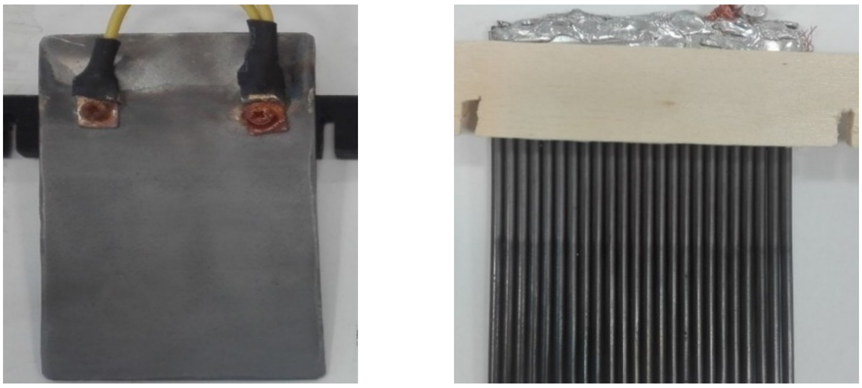
$$\mathrm{ELC} = \frac{I\,t\,M}{nF} \tag{8}$$

Where (I), (t), (M), (n), and (F) are current intensity (A) , reaction time (S), molar mass (g/M), number of electrons, and the Faraday constant which has a value of 96,485 Coulombs per mole, respectively (Yoosefian et al., 2017).

Pareto chart

To determine the significance of the primary, interaction, and quadratic variable effects in the overestimation model for AMX removal, the Pareto diagram is employed, and the proportional contribution of each parameter to the diagram is computed using Eq (9).

$$P_i = \frac{b_i^2}{\sum b_i^2} \times 100 \quad (\mathrm{i} \neq 0) \tag{9}$$



b) Titanium Anode **a) Graphic Cathode**

Fig. 1. Cathode and anode used in this study a) Graphic cathode and b) Titanium anode

In the above Equation, P_i and b_i are the participation percentage of each parameter and influence coefficient of each parameter (Ahmedzadeh & Dolatabadi, 2016).

RESULTS AND DISCUSSION

The experimental data were subjected to analysis using software, which facilitated the estimation of predicted values for the percentage of AMX removed through the implementation of the selected model. Eq (10) exhibits the prediction model for AMX removal efficacy, which is based on the software-encoded values.

$$\text{AMX } Removal\ (\%) = +84.15 - 4.68\,A + 3.22\,B + 4.81\,C + 5.34\,D - 5.24\,\mathrm{B}^2 - 3.54\,\mathrm{C}^2 \quad (10)$$

In this Eq, whereby A, B, C, and D, correspond to initial of AMX concentration (mg/L), initial pH, current intensity (mA), and EDTA concentration (mM), respectively. The inclusion of these variables in Eq (8) was determined by their P-value, as represented in the variance table, which indicated statistical significance. Notably, variables A, B, C, D, B^2, and C^2 demonstrated statistical significance, with a positive sign in proximity to their respective terms within Eq (8) denoting an increasing effect on the predicted removal percentage, while a negative sign indicates a decreasing effect. As reported by Eslami et al. (2016), insignificant variables with P-values exceeding 0.05 were excluded from the model.

The variance analysis was conducted to evaluate the predictive performance of the model for AMX removal percentage. The Design-Expert® software recommended the model that

Table 3. Results of experimental design values of CCD

Run	AMX removal (%)	Predicted values (%)
1	77.7	82.7
2	75.4	74.4
3	65.8	66.6
4	73.2	73.4
5	84.6	84.1
6	95.6	92.5
7	76.2	74.7
8	84.5	74.0
9	85.1	84.1
10	78.7	77.6
11	58.1	56.7
12	62.1	66.9
13	92.6	94.8
14	87.8	86.9
15	68.5	69.9
16	84.7	84.1
17	67.5	68
18	81.5	79.6
19	56.7	57.3
20	78.2	83.8
21	84.5	84.1
22	84.5	84.1
23	64.2	63.7
24	76.6	73.3
25	79.8	76.1
26	74.9	73.1
27	90.3	93.4
28	75.8	77.3
29	58.8	60.4
30	80.8	84.1

exhibited the highest correlation among variables, with a quadratic model being identified as the optimal model for this study. Table 3 displays the experimental outcomes alongside the values projected by the software.

The degree of fit of the model to the observed response changes as a function of the variables is indicated by the R^2 (Correlation coefficient) value, whereby the closer this value is to one, the greater the efficacy of the model furthermore, the values of R^2 and Adj. R^2 reflects the strength of the correlation between the predicted and observed values and the degree to which the model can fit the data. The Adj.R^2 parameter quantifies the magnitude of data changes around the mean. Adequate Precision (A.P.) reflects the values attained in the central points relative to the mean, with a value greater than 4 suggesting the model's adequacy. The Coefficient of Variation (C.V) signifies the degree of dispersion per unit of the mean and is calculated by dividing the standard deviation by the mean and then multiplying by 100 (Chavashqli et al., 2018).

Table 4 displays a P-value for the model that is less than 0.05, indicating its significance. The misfit index has a P-value of 0.1413, indicating its insignificance and confirming the validity of the model. The adequate precision, which measures the signal-to-noise ratio, is equal to 32.38, indicating the model's suitability as its value is greater than 4 (Eslami et al., 2016). The values for R^2 and Adj. R^2 in the Table S2 are estimated as 0.9572 and 0.9460, respectively, and logically are consistent, with a difference of less than 0.2. The C.V which expresses the model's repeatability, is equal to 3.16, and values less than 10% suggest that the experiments are highly accurate and reliable.

The significance of the primary, combined, and quadratic variables effect in removal of AMX were assessed using the Pareto chart, and the proportion of each parameter's contribution was computed through Eq. (7). Based on the Pareto analysis diagram, the most impact on pollutant removal was attributed to the quadratic pH (22.15) and EDTA (23.03) concentration variables. Conversely, the effect of the pH variable was found to be the least, indicating the chelating agent's effect in diminishing pH's effect on removal of AMX.

Statistical diagrams of model description

The graphs presented in Figs S2-S5 are powerful evidence that the chosen model is accurate and valid. Fig S2 clearly shows the correlation between actual and predicted values, helping to identify any outliers or inaccuracies. When points are closer to the line, it indicates a stronger agreement between observed and predicted values (Eslami et al., 2016). Fig S3 displays a normal distribution of residuals without any observable patterns, suggesting that the model is well-suited to the data. In Fig S4, residual distribution as a function of predicted values should be randomly

Table 4. Confirmatory experiments at optimum condition

Source	Sum of squares	Degree of freedom	Mean square	F-value	p-value	
Model	3047.19	6	507.86	85.69	< 0.0001	**Significant**
A-C_0	526.50	1	526.50	88.84	< 0.0001	
B-pH	249.42	1	249.42	93.64	< 0.0001	
C-I	554.98	1	554.89	93.64	< 0.0001	
D-EDTA	684.48	1	684.48	115.49	< 0.0001	
B^2	780.60	1	780.68	131.72	< 0.0001	
C^2	355.46	1	355.64	59.98	< 0.0001	
Residual	136.31	23	5.93	-	-	
Lack of Fit	123.43	18	6.89	2.66	0.1432	**Not significant**
Pure Error	12.88	5	2.58	-	-	

scattered within two lines for accuracy (Hasani et al., 2020). Finally, Fig S5 demonstrates that all standardized residuals fall within acceptable ranges. These results highlight the importance of using this model for reliable predictions. (Ahmedzadeh & Dolatabadi, 2016).

The Perturbation plot of the model is presented in Fig 4. The saturation diagram displays the effect of all parameters at the central point of the design space. A steep slope or curvature of a parameter signifies its sensitivity to the response, while a smooth line indicates insensitivity to a change in that particular factor (Salari et al., 2018). The diagram reveals that decreasing the initial concentration of AMX (A) has a positive impact on AMX removal. Similarly, increasing the amount of EDTA (D) enhances the percentage of AMX removal. As for the initial pH (B), the diagram demonstrates that increasing it from a low level to a certain amount leads to an increase in the percentage of antibiotic removal. However, a further increase in the initial pH causes a reduction in the percentage of antibiotic removal, indicating the effect of the chelating agent in increasing the initial pH up to a certain range.

The effect of the interaction of effective variables on the removal percentage of AMX

This section investigates the impact of the independent variables of the current and the initial antibiotic concentration on antibiotic removal efficiency, as depicted in Figs 5 to 7. As shown in Fig 5, a removal percentage of more than 90% is achieved at an initial AMX concentration

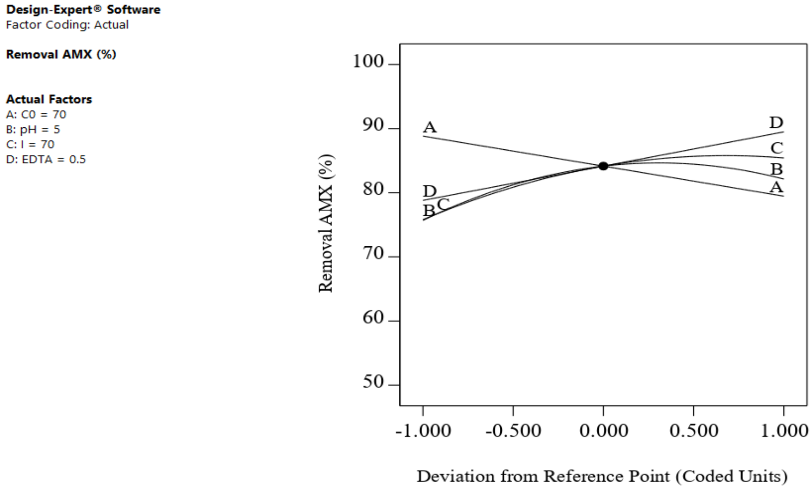


Fig. 4. Perturbation plot showing the effect of all factors on the AMX

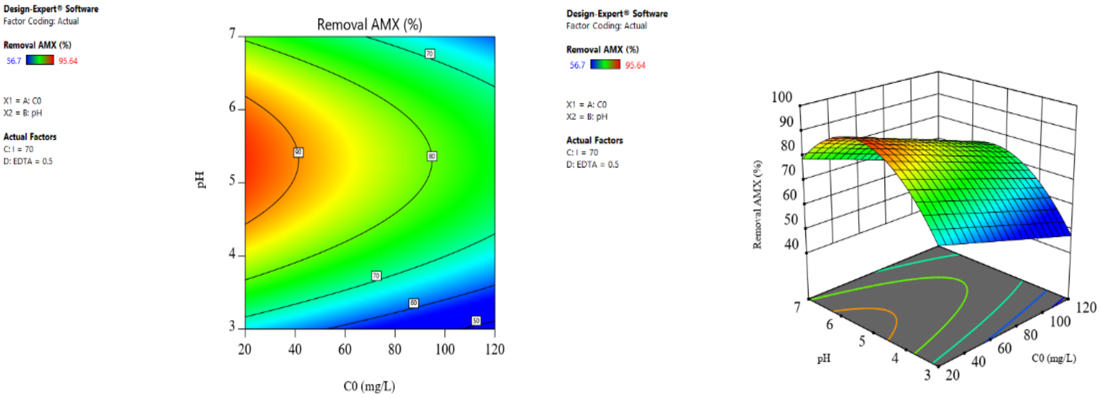


Fig. 5. The effect of AMX concentration and initial pH on AMX removal

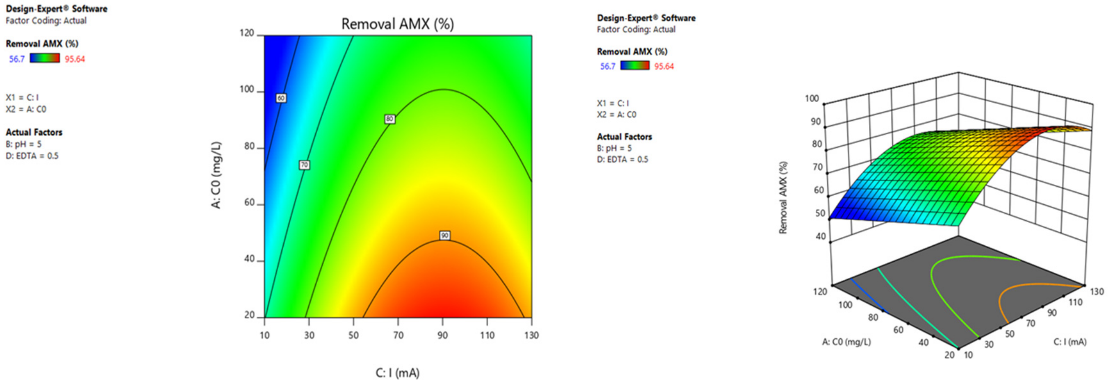


Fig. 6. The effect of AMX concentration and current intensity on AMX removal percentage

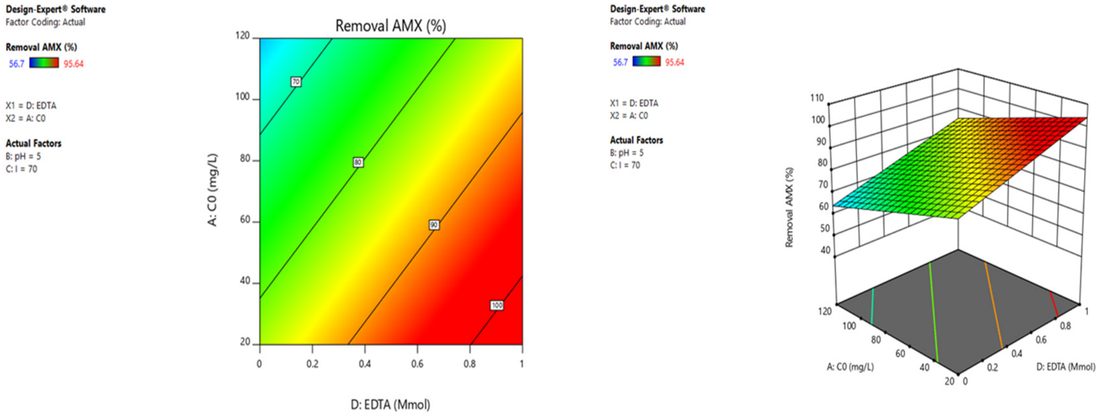


Fig. 7. The effect of AMX and EDTA concentration on AMX removal percentage

Table 4. The analysis of variance (ANOVA) results for adequacy of the quadratic model

Response	**Prediction value**	**Experiment value**	**ARE* (%)**
AMX Removal	$(95.71)^4$‹$(95.90)^3$‹$(95.42)^2$‹$(95.82)^1$	36.96	0.67
First run [1]	Second run [2]	Average of two runs [3]	ARE: Absolute Relative Error *

of less than 40 mg/L and pH between 5 and 6. Similarly, Figs 6 and 7 illustrate that AMX removal rates of over 90% can be achieved at concentrations less than 40 mg/L and current intensities between 70 and 110 mA, as well as EDTA concentrations greater than 0.8 mM and concentrations less than 40 mg/L.

Optimization results and confirmatory trials

This study aimed to optimize variables during the design phase to maximize the removal of AMX. The software generated 100 optimal suggested points, and one of the points with current intensity of 85 mA, pH of 5.6, initial AMX concentration of 24 mg/L, and EDTA amount of 0.6 mM were selected based on Fig S6. Three experiments conducted, and the results are presented in Table 4. As indicated in the table, there is a strong correlation between the experimental and predicted values, suggesting the model's accuracy and reliability.

Electrical energy consumption

Using Eq (9) to calculate the energy consumption in terms of (kWh/m^3), it was discovered that the in optimal point with a voltage of 2.03 V, current intensity of 85 mA, duration of 30 min, and volume of 0.1 liters, for electrical energy consumption is achieved 0.386 kWh/m^3. This finding was compared with data other studies, as presented in Table 5, and it was determined that the optimal energy consumption level is comparable to values reported in previous research. It is clear that these optimal conditions should be used to help reduce overall electrical energy consumption.

Consumable electrode

The optimal electrode consumption has been calculated using Eq (6), where this denotes the current intensity of 0.085 mA, it is the reaction time of 1800 seconds, M represents the molar mass of divalent iron of 55.85 g/mol, n is the number of transition electrons equal to 2, F denotes Faraday's constant of 96485 mol/Coulomb. The amount of electrode used for each experiment was found to be 0.044 g. In previous study by Yousefian et al. (2017) on CIP, the amount of electrode used for each experiment was 0.0625 g (Yousefian et al., 2017). The electrodes used in this study showed no corrosion after experimenting and are deemed suitable for this process.

CONCLUSION

The aim of the present study was to investigate and optimize the electro Fenton process in the removal of AMX antibiotic from aqueous solution using EDTA chelating agents at high pH. The results showed that among the examined parameters, the amount of EDTA had the more effect and instead, the pH had the least effect on the antibiotic removal efficiency, which indicates the effectiveness of the chelating agent in maintaining iron at high pH. In optimal conditions, the initial concentration of AMX 24 mg/L, the current intensity 85 mA, initial pH 6.5, and the concentration of EDTA were 0.6 mM in a period of 30 min, which removed 95.71% of AMX. The use of chelating agent showed an important role in increasing the pH and increasing the pollutant removal percentage. Also, the results of energy consumption at the optimal point according to the voltage of 2.03 V, the current intensity of 85 mA the duration of 30 min and the volume of 0.1 liter were obtained equal to 0.386 kWh/m^3. The findings of the current research will benefit others in several ways:

Environmental Impact: The optimized Electro-Fenton process offers a more efficient and sustainable method for removing amoxicillin antibiotic from aqueous solutions. This can contribute to reducing the environmental impact of pharmaceutical pollutants in water bodies.

Resource Efficiency: By using chelating agents at high pH values, the process may lead to improved resource efficiency and cost-effectiveness in the treatment of antibiotic-contaminated water.

Health and Safety: The research findings can potentially enhance water treatment processes, thereby ensuring cleaner and safer drinking water for communities, ultimately benefiting public health.

Technological Advancements: The optimization strategies and insights gained from this research can be applied in the development of enhanced water treatment technologies for pharmaceutical pollutant removal, benefiting researchers and practitioners in the field.

In conclusion, the outcomes of this study have the potential to advance the field of water treatment, offering practical solutions for addressing antibiotic contamination in aqueous environments and benefiting various stakeholders concerned with water quality and environmental sustainability.

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The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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