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A feasible treatment of carpet-washing wastewater through the electro-fenton process: optimization using response surface methodology

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ABSTRACT

Wastewater from carpet washing has yet to be extensively studied despite its high volume and pollution load due to complex hazardous materials and high organic content. This study examined the feasibility of using Electro-Fenton (EF) processes to remove chemical oxygen demand (COD) from carpet washing wastewater (CWW) generated by the Nickpour Industry in Iran. COD removal was used to assess the total organic content. Anionic surfactants, especially Linear Alkylbenzene Sulfonate (LAS), are widely used in household and industrial detergents. The study utilized an experimental design using response surface methodology (RSM) based on the Box–Behnken method to evaluate the setup's effectiveness and optimize the conditions. Five independent factors, including H2O2/COD ratio, reaction time, effective surface area, pH, and applied voltage, were used as indicators to optimize the reaction parameters. Although LAS elimination reduced turbidity, the results showed that effective surface area and pH had a more significant impact on COD removal than other variables. The interactions between different parameters also had a significant effect on the results. Under the optimal conditions of a 1.8 H2O2/COD ratio, 3 cm2 electrode surface area, 30 minutes of reaction time, 23 V applied voltage, and an initial pH of 3, the study achieved a COD removal rate of 97.99%, a 99.92% removal of LAS, and a reduction of turbidity by up to 99.99%.

1. Introduction

Water is essential for the existence of all living organisms and is a resource that requires careful protection. Contaminating water sources poses a severe risk to human health by causing waterborne diseases and environmental degradation. As a result, there is a growing emphasis on waste reduction and wastewater recycling as strategies to mitigate the consequences of water contamination [1, 2]. Carpets are a typical floor covering in Iranian houses and reflect the region's cultural heritage and skilled craftsmanship. However, they are washed in factories that release a significant amount of wastewater into the environment. Properly managing CWW is crucial due to its toxic and hazardous contaminants, such as grease, oil, chlorinated solvents, and heavy

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metals. The wastewater treatment of industrial laundries has been the subject of much research, but CWW could have unique differences; therefore, it is necessary to study it separately. It contains a mixture of grease, oil, chlorinated and aromatic solvents, colors, disinfectants, volatile organic compounds (VOCs), heavy metals, sand, soil dust, and various detergents, which pose a significant environmental threat if not handled correctly. Developing effective strategies for treating and disposing of CWW involves implementing advanced treatment methods and responsible disposal practices to ensure environmental sustainability. This involves minimizing the generation of wastewater and enabling reuse through treatment and reclamation processes [3-5].

Surfactants, builders, and fillers are the primary ingredients of detergents. Surfactants are crucial in cleaning products, enabling them to efficiently remove dirt, grease, and grime from various surfaces. Surfactants containing hydrophilic and hydrophobic groups reduce surface tension between liquid molecules, allowing them to be dispersed and washed away. Anionic, cationic, nonionic, and amphoteric surfactants have different characteristics and uses. Cleaners, disinfectants, laundry detergents, and personal care products use anionic, cationic, nonionic, and amphoteric surfactants, respectively. Formulating with a combination of surfactants can enhance cleaning [6-8]. LAS is the predominant anionic surfactant used in cleaners due to its high efficacy and affordability. Despite its low toxicity to humans, LAS is a harmful component in wastewater that can harm aquatic life, disrupt development and photosynthetic processes, and cause skin and eye irritation, respiratory issues, and lung damage [9]. Various methods exist for removing surfactants: chemical precipitation, membrane technology, adsorption, microwave irradiation, biological methods, electrochemical coagulation, and flocculation. Each method has limitations in practical application, such as sludge generation, high maintenance costs, potential fouling, etc. [10, 11].

Advanced Oxidation Processes (AOPs) employ highly reactive hydroxyl radicals ($OH^{\bullet}$) to chemically transform organic and inorganic contaminants in water and wastewater. These reactive radicals can oxidize and break down a wide range of recalcitrant organic pollutants, such as aromatics, pesticides, petroleum constituents, volatile organic compounds (VOCs), and some inorganic contaminants. Standard AOPs include Hydrogen Peroxide-Ultraviolet Radiation (H_2O_2/UV), Photolytic (O_3/UV), Fenton (Fe^{2+}/H_2O_2), Photo-Fenton (Fe^{2+}/ H_2O_2/UV) and Electrochemical Electro-Fenton (EF). AOPs are highly effective in treating non-biodegradable, hazardous, or refractory pollutants in industrial wastewater. Advantages of AOPs include the ability to handle varying flows/compositions and the potential for complete mineralization of organics to CO_2 and H_2O [12, 13].

Electro-Fenton is an eco-friendly and cost-effective technique that combines the advantages of Fenton and electrochemical methods; however, it has sludge production, pH sensitivity, and energy consumption. This technology enhances the breakdown of organic pollutants by continuously regenerating ferrous ions (Fe^{2+}) at the cathode electrode and producing hydrogen peroxide (H_2O_2) electrochemically by reducing dissolved oxygen on the cathode surface. In the EF approach, hydroxyl radicals ($OH^{\bullet}$), the most potent oxidizing species, can non-selectively oxidize any organic pollutant into CO_2 and H_2O (Eqs. 1 and 2).

$$\mathrm{OH^{\bullet} + Organic\ pollutant \rightarrow Primary\ intermediates} \quad (1)$$

$$\mathrm{Primary\ intermediates + OH^{\bullet} \rightarrow CO_2 + H_2O + Inorganic\ ions} \quad (2)$$

The EF process progresses through the following chain reactions. Eq. 3 is sustained by the continuous regeneration of Fe2+ at the cathode. In order to prevent the buildup of ferric ion (Fe^{3+}), Eq. 4 is initiated, and the resulting Fe^{3+} from Eq. 3 can be reduced to Fe^{2+} on the cathode surface [14-16].

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

$$Fe^{3+} + e^{-} \rightarrow Fe^{2+} \quad (4)$$

This research extensively examined the impact of different factors, including the H_2O_2/COD ratio, reaction time, effective surface area, pH, and applied voltage on LAS, COD, and turbidity, from genuine CWW using the EF approach. Moreover, a cost-efficient electrolytic solution employing two iron electrodes was employed to accomplish this objective.

2. Materials and Methods

2.1 *Chemical and analytical procedures*

The chemicals used in this study were of analytical grade and procured from Merck, Germany: hydrogen peroxide (H_2O_2), methylene blue, chloroform, linear alkylbenzene sulfonate, phenolphthalein, and methanol. Sodium hydroxide (NaOH) and sulfuric acid (H_2SO_4) were employed to adjust the initial pH of the solutions. The CWW samples were collected from the Nickpour Carpet Washing Industry located in Rostamabad, Guilan Province, Iran, and transported immediately to the laboratory for analysis. All experiments were conducted under controlled conditions at room temperature (20 ± 2°C) and standard atmospheric pressure.

The key parameters of LAS, COD, and turbidity were quantified using a spectrophotometer (MACHEREY-NAGEL) at 652 nm, a turbidity meter (LUTRON TU), and a COD heat reactor at 600 nm and 150°C (AQUA LYTIC AL 100), respectively. The initial characteristics of the wastewater, including pH, temperature, and composition, are provided in Table 1.

2.2 *Electro-Fenton process*

The electrochemical cell utilized in this study was meticulously designed to optimize the performance of the Electro-Fenton (EF) process for treating CWW. This cell included two parallel iron electrodes made of iron, which were immersed in a 400 ml glass container. The strategic positioning of these electrodes at varying depths allowed for the modification of the effective surface area, a critical factor influencing the rate of iron ion (Fe^{2+}) production. By adjusting the effective surface area, the study aimed to maximize Fe^{2+} ion generation, which was essential for the continuous production of hydroxyl radicals (OH•) through the Fenton reaction. These radicals played a pivotal role in breaking down complex organic contaminants present in the wastewater.

In each test, specific amounts of CWW and H_2O_2 were initially added to the electrolytic cell, and the pH was adjusted. The added H_2O_2 served as a reactant, which was consumed during the oxidation process but was continuously regenerated at the cathode as per Eq. 5. This regeneration process was crucial for sustaining the chain reactions necessary for the efficient degradation of pollutants.

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \tag{5}$$

The current density was precisely set to 2.5 A using a digital power supply (DAZHENG P-S305D) to initiate the EF process (Fig.1), ensuring optimal conditions for the electrocoagulation process. This precise control of current density is vital for maintaining the desired electrochemical reactions, particularly the production and regeneration of Fe^{2+} ions. The electrodes were then positioned 3 cm apart within the sample. Following each run, the samples were allowed to settle for 30 minutes to facilitate particle settling before being separated using filter paper. Finally, the results were analyzed after performing LAS, COD, and turbidity measurements on the filtered samples using the previously mentioned methods and calculating the removal percentage with Eq. 6.

Table 1. Initial characteristics of used wastewater.

Parameters	Units	Values
Chemical oxygen demand (COD)	mg/l	765
Biochemical oxygen demand (BOD5)	mg/l	163
Linear Alkylbenzene Sulfonate (LAS)	mg/l	50.81
Turbidity	NTU	142.7
Color	TCU	18.7
pH	-	9
Temperature	°C	19
Total Suspended Solids (TSS)	mg/l	578
Total Dissolved Solids (TDS)	mg/l	85.1
Electrical Conductivity (EC)	ms/cm	170.2
TDS/EC ratio	-	0.5

$$Removal(\%) = (C_{in} - C_{out})/C_{in} \times 100 \quad (6)$$

where C_{in} and C_{out} are the initial and final concentration of LAS, COD, or turbidity values.

This approach allowed for a precise evaluation of the efficiency of the EF process, demonstrating the significant reduction in contaminants achieved under the optimized experimental conditions.

2.3 *Experimental Design*

The Box-Behnken design, guided by Minitab software (Version 11), was used to assess the impact of five independent variables: H_2O_2/COD ratio (X1), reaction time (X2), effective surface area (X3), pH (X4), and applied voltage (X5). This experimental design is particularly effective for studying the interactions between multiple factors with a limited number of experiments, allowing for the optimization of complex processes like the Electro-Fenton treatment. Each variable was examined at three levels: low, intermediate, and high. These levels were selected based on preliminary studies and the operational ranges of the variables, ensuring that the design space effectively captured the behavior of the system under different conditions. The efficiency of the current treatment is documented in Table 2.

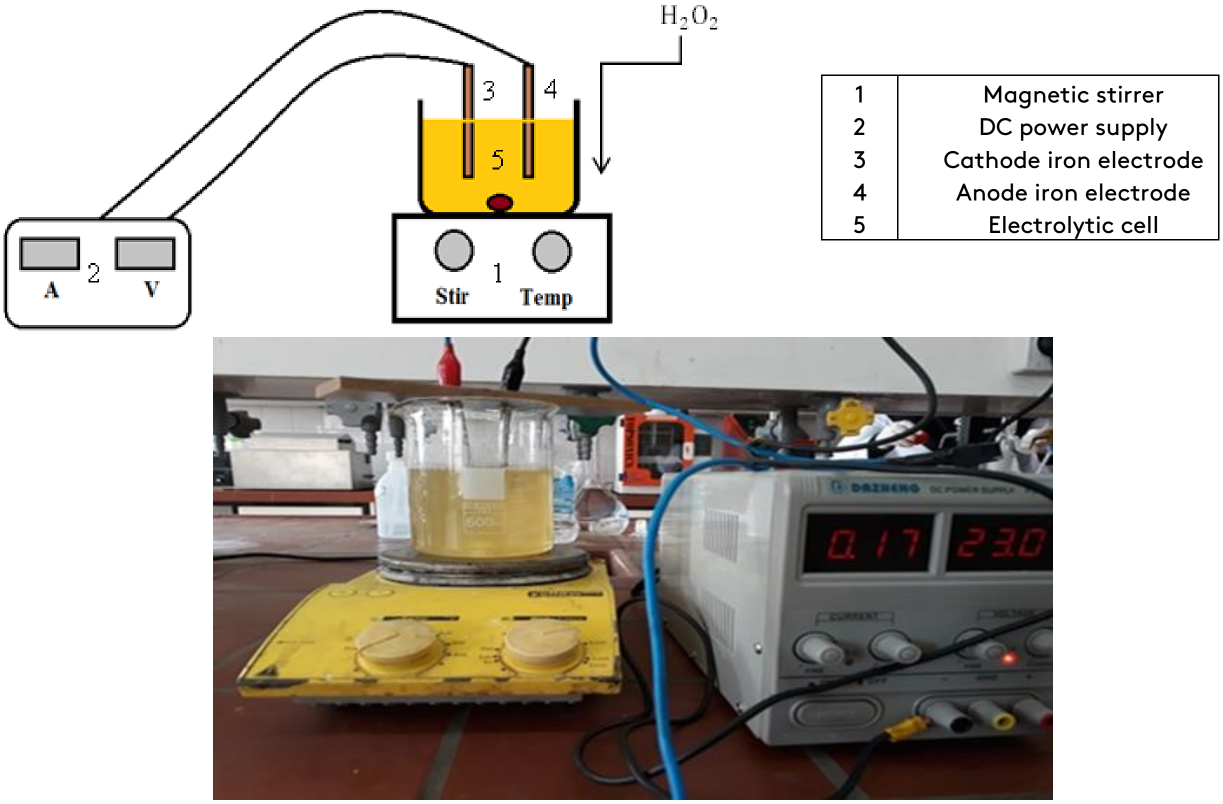


Fig. 1. Electro-Fenton process reactor set-up.

Table 2. Independent factors and their levels.

Symbol	Factors	Units	Coded levels for variables	
			-1	+1
X1	H_2O_2/COD	ml/l	1.80	2.30
X2	Reaction time	min	30.00	70.00
X3	Effective surface area	cm^2	1.0000	3.00
X4	pH		3.00	4.00
X5	Voltage	V	23.00	25.00

As shown in Table 3, 31 experiments were designed, independent factors were optimized, and the data were analyzed using analysis of variance (ANOVA). The use of ANOVA allowed for the evaluation of both individual and interactive effects of the independent variables, providing a comprehensive understanding of their influence on the process outcomes. Individual and interactive effects were expressed through variables [17].

This study used the second-order model equation, represented by Eq. 7, to predict the optimal conditions and express the correlation between the responses and variables. This quadratic model is particularly useful in capturing non-linear relationships and interactions between variables, which are common in complex processes like the Electro-Fenton treatment.

$$Y= \beta_0 + \sum_{i=1}^{k} \beta i.Xi + \sum_{i<j}^{k} \beta_{ij}.X_iX_j + \sum_{i=1}^{k} \beta_{ii}.{X_i}^2 .... + \varepsilon \qquad (7)$$

In Eq. 7, Y represents the response variable (e.g., COD removal), i, j, and β are the linear, second-order, and regression constants, respectively, and ε denotes the random error term accounting for the variability not explained by the model. The number of factors is denoted by k, and in this case, corresponds to the five independent variables examined in the study.

Table 3. Experimental matrix, Box Behnken design for the optimization of the Fenton process.

Run	H_2O_2/COD	Time (min)	Effective Surface Area (cm^2)	pH	Voltage
1	1.8	40	2	3	24
2	2.1	30	2	3	23
3	2.3	70	1	3	25
4	1.8	30	1	4	23
5	1.8	70	3	4	23
6	2.3	30	3	4	23
7	2.3	30	1	3	25
8	1.8	70	1	4	25
9	2.3	70	1	4	23
10	2.1	50	3	4	24
11	1.8	30	3	3	23
12	2.3	70	3	4	25
13	2.3	70	3	3.5	23
14	2.1	50	2	3.5	24
15	1.8	40	2	3	24
16	1.8	30	1	3	23
17	1.8	70	1	3	23
18	2.3	40	1	3	23
19	2.3	70	1	3	25
20	1.8	30	3	4	25
21	1.8	70	3	3	25
22	1.8	50	1	3.5	23
23	2.1	50	3	4	24
24	2.3	70	3	4	25
25	1.8	30	3	4	25
26	2.3	30	3	3.5	24
27	2.1	50	2	3.5	24
28	2.3	30	2	3	23
29	2.1	60	3	4	24
30	2.3	40	3	3	25
31	1.8	50	2	3.5	25

3. Results and discussion

3.1 Model fitting and statistical analysis for COD removal

The ANOVA results, with an F-value of 12.34 and a p-value of <0.001 from Eq. 8, suggested that a quadratic model provided the best fit for the responses. This quadratic model was employed to accurately predict the removal of COD using iron electrodes by excluding the coefficients that were deemed insignificant.

$$\text{COD Removal}(\%)= +58.39-4.07A-1.02B +5.78C-15.35D-2.80E+3.35AB+4.18AC -4.65AD-6.37AE+11.11BD+4.49CD- 2.70CE-6.39DE+6.06C^2+13.79D^2 \quad (8)$$

Fig. 2 depicts the experimental and corresponding predicted values obtained from Eq. 8 for COD removal. The high R^2 value of 0.9922 for COD removal demonstrated that the model was in close agreement with the experimental values, indicating a strong correlation.

The new model was validated using ANOVA, which checked model accuracy, evaluated data fit, and determined parameter significance. To estimate the goodness-of-fit and sufficiency of the model, several statistical measures such as R^2, Adjusted R^2, Predicted R^2, F, and P values, the Coefficient of Variation (C.V. %), and Adequate Precision (A.P.) were evaluated, and the results are presented in Table 4.

The quadratic model shows that R^2, Adjusted R^2, and Predicted R^2 values were 0.9922, 0.9844, and 0.9650, respectively. The closeness of these three coefficients to one indicated a high degree of correlation between observed and predicted data and clearly demonstrated the interactions between the independent parameters and responses. In this study, as shown in Table 4, the F-value and P-value were 127.48 and <0.0001 for COD removal, respectively. The high F-value and low P-value (below 0.05) indicated that the regression model could explain most of the variation in the responses. This confirmed that the obtained model was statistically significant. The Coefficient of Variation (C.V. %) was reported as 2.33%, indicating a low level of relative variability. The low Coefficient value indicated the high precision and reliability of the experimental runs.

Adequate Precision (AP) evaluates the signal-to-noise ratio and the range of estimated values at specific points compared to the average prediction error. The AP ratio for COD removal was calculated as 32.3024. Ratios greater than four suggested sufficient model discrimination and showed that the model could be reliably employed for investigating the intended domain.

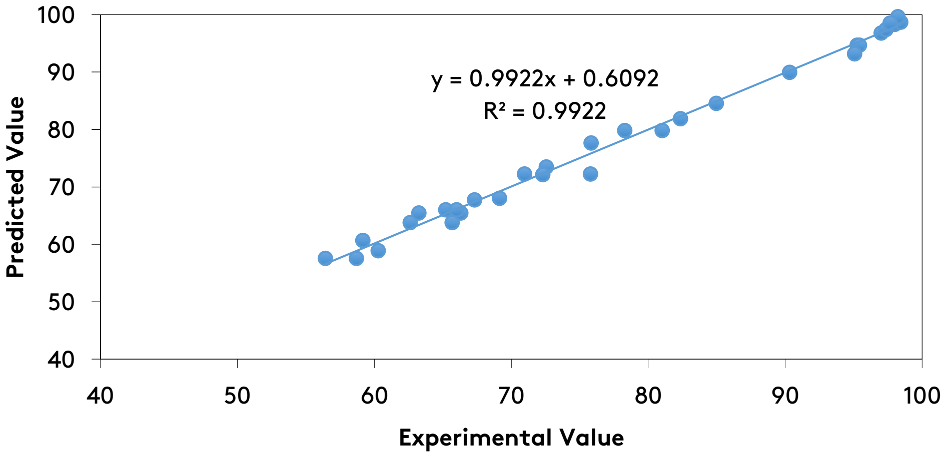


Fig. 2. Experimental vs. predicted values for COD removal.

Table 4. Analysis of variance data.

Source	Sum of Squares	df	Mean Square	F-value	P-value	Remark
Model	6290.45	15	419.36	127.48	< 0.0001	Significant
A- H_2O_2/COD Ratio	252.43	1	252.43	76.73	< 0.0001	Significant
B-Reaction Time	15.30	1	15.30	4.65	0.0477	
C-Effective Surface Area	503.79	1	503.79	153.14	< 0.0001	Significant
D-pH	2672.32	1	2672.32	812.33	< 0.0001	Significant
E-Voltage	132.97	1	132.97	40.42	< 0.0001	Significant
AB	149.36	1	149.36	45.40	< 0.0001	Significant
AC	185.22	1	185.22	56.30	< 0.0001	Significant
AD	266.36	1	266.36	80.97	< 0.0001	Significant
AE	415.22	1	415.22	126.22	< 0.0001	Significant
BD	1193.62	1	1193.62	362.84	< 0.0001	Significant
CD	206.24	1	206.24	62.69	< 0.0001	Significant
CE	105.95	1	105.95	32.21	< 0.0001	Significant
DE	358.73	1	358.73	109.05	< 0.0001	Significant
C^2	115.36	1	115.36	35.07	< 0.0001	Significant
D^2	655.72	1	655.72	199.32	< 0.0001	Significant
Residual	49.35	15	3.29			
Lack of Fit	22.20	9	2.47	0.5454	0.8015	Not Significant
Pure Error	27.14	6	4.52			
Cor Total	6339.79	30				

Coefficient	R^2	R^2_{Adj}	R^2_{Pre}	F-value	P-value	C.V. %	A.P	Press
Value	0.9922	0.9844	0.9650	127.48	<0.0001	2.33	32.3024	221.93

Utilizing perturbation and diagnostic plots can confirm the model's reliability and adequacy, allowing for a discussion of its efficiency and deficiency. The perturbation plot (Fig. 3a) shows COD removal (%) versus coded factors. The effects of each factor under ideal circumstances are compared in the design space under consideration using the perturbation plot. The steep curvature of factors indicated that the responses were highly affected by these factors, and a relatively straight line demonstrated a lower effect on COD removal. As shown in Table 4 and Fig. 3a, the data revealed that effective surface area and pH had a significant impact on results. In contrast, variations in H_2O_2/ratio, reaction time, and applied voltage showed a minimal effect on COD removal.

The predicted versus actual response values for COD removal (%) by process are shown in Fig. 3b. As seen in this plot, the points fell on a relatively straight line, indicating the constancy of variance. The comparison of both data revealed that the predicted and actual COD removal (%) were in good agreement with each other.

Fig. 3 shows the standard plot of residuals for COD removal from CWW by the EF process to approve whether the predicted and actual values were distributed normally or not. As displayed in Fig. 3c, points were aligned on in an almost straight line. Thus, this plot indicated an adequate correlation between the response values.

Figures 3d and 3e show the plots of studentized residuals versus predicted COD removal (%) and studentized residuals versus experimental runs, respectively. These diagrams showed that all points were randomly scattered, and all values were laid within ±3 sigma without any unusual structure. Accordingly, these plots confirmed that the captured model effectively revealed the relationship between the examined variables.

3.2 Effect of operation parameters on COD removal

Fig. 4 presents a three-dimensional (3D) response surface and two-dimensional (2D) contour plot of the effects of various variables on COD removal. The 3D response surface can be utilized to evaluate the removal efficiency, and the 2D contour plot presents the combined effect of any two variables on COD removal percentage. In contrast, the other variables are kept constant at the midpoint values. Determining the optimum H_2O_2 concentration in the EF process is very important for the related removal efficiency. The effect of initial H_2O_2 concentration on the removal efficiencies of industrial laundry wastewater was studied in the range of 0.1–0.5ml/l. The rates at which color and COD were removed were directly related to the amount of H_2O_2 present when the EF process was run with a high enough initial H_2O_2 concentration. This is because of the unwanted OH scavenging reaction effect. This situation was contingent upon Eqs. 9-11, resulting in the generation of less reactive radicals from the hydroxyl radical. Critical operating parameters, such as reaction time, temperature, and concentration, significantly influence the process by determining the optimum conditions and response measures. The data from Fig. 4 (a, b) highlights that the influence of reaction time on COD removal may be lower than the other factors, but it remains a crucial parameter that demands attention. Increasing the reaction time impacted the EF process negatively, as it led to the completion of the oxidation reaction over time. This was due to the reactant's reduction and undesirable side reactions simultaneously. Hydroperoxyl radicals($HO_2^\bullet$) have lower oxidation power during formation in the electrolytic cell than hydroxyl radicals in the degradation of organic pollutants [20].

$$Fe^{2+} + OH^\bullet \rightarrow Fe^{3+} + OH^- \qquad (9)$$

$$Fe^{3+} + H_2O_2 \leftrightarrow Fe - OOH^{2+} + H^+ \qquad (10)$$

$$Fe + OOH^{2+} \rightarrow HO_2^\bullet + Fe^{2+} \qquad (11)$$

The experiments showed that the reaction between Fe^{2+} ions and H_2O_2 was completed by producing $OH^\bullet$radicals.

The maximum COD removal occurred at 30 min and decreased after the optimum reaction time due to the depletion of reactants [21]. Achieving 79% COD removal and 63% BOD removal within a 30-minute EF process was considered successful. These outcomes aligned with expectations and 30 minutes was established as the ideal duration for the research. Another significant operational factor in the EF process is the effective surface area of electrodes, which influences the efficiency of COD removal by facilitating more excellent contact between the electrodes and the wastewater. As observed in Fig. 4(c, d, and e), increasing the effective surface area from 1 to 3 cm^2 by adjusting the depth of immersion in each run resulted in enhanced COD removal due to improved electrode-wastewater interaction. This trend could be attributed to more excellent production of H_2O_2 and °OH radicals at the place of the cathode and anode electrodes, respectively, and also maximizing the process efficiency by higher electro-regeneration of Fe^{2+} ions from Fe^{3+} ions at 2.5A current density. Furthermore, the higher effective surface area led to higher COD removal from the carpet wastewater. The applied voltage was a crucial operational parameter that significantly influenced the efficiency of the EF system by controlling the generation of reactive species and the electro-regeneration of Fe^{2+} ions. According to the data in Fig. 4(e, f, and g), the optimal voltage of 23 volts was determined to maximize COD removal in the EF method, indicating a direct correlation between applied voltage and treatment efficiency. As expressed under applying a relatively high voltage, the removal efficiency was enhanced due to the higher quantity of radicals formed from Fenton's reactions (Reaction 1).

Also, releasing ferrous ions (Fe^{2+}) (Reaction 4), which produce $OH^\bullet$ radicals in reaction with hydrogen peroxide (Reaction 3), resulted in an improvement in the COD target rate. The removal percentage declined by further increasing the applied voltage up to 25 V. Based on our understanding and Faraday's law of electrolysis, an increase in electrical current results in the enhanced generation of Fe^{2+} ions. It was expected that the Fe^{2+} ions would increase in proportion to the necessary amount for reacting with H_2O_2 as the applied voltage increased.

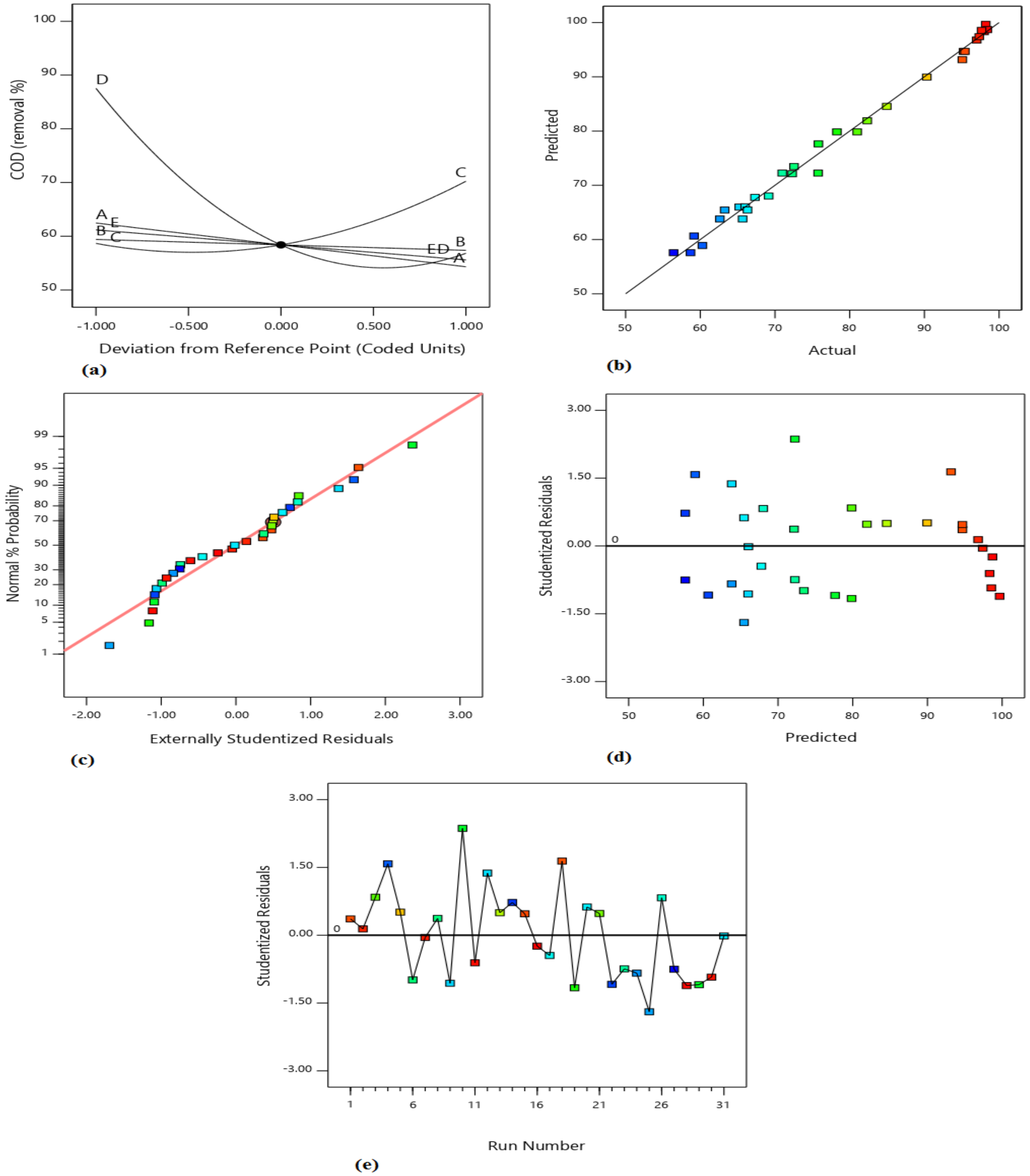


Fig. 3. (a) perturbation plot of COD removal (%), (b) Predicted vs. actual plot of COD removal (%), (c) Normal probability of the studentized residuals, (d) Studentized residuals vs. predicted COD removal (%), and (e) Studentized residuals vs. run number.

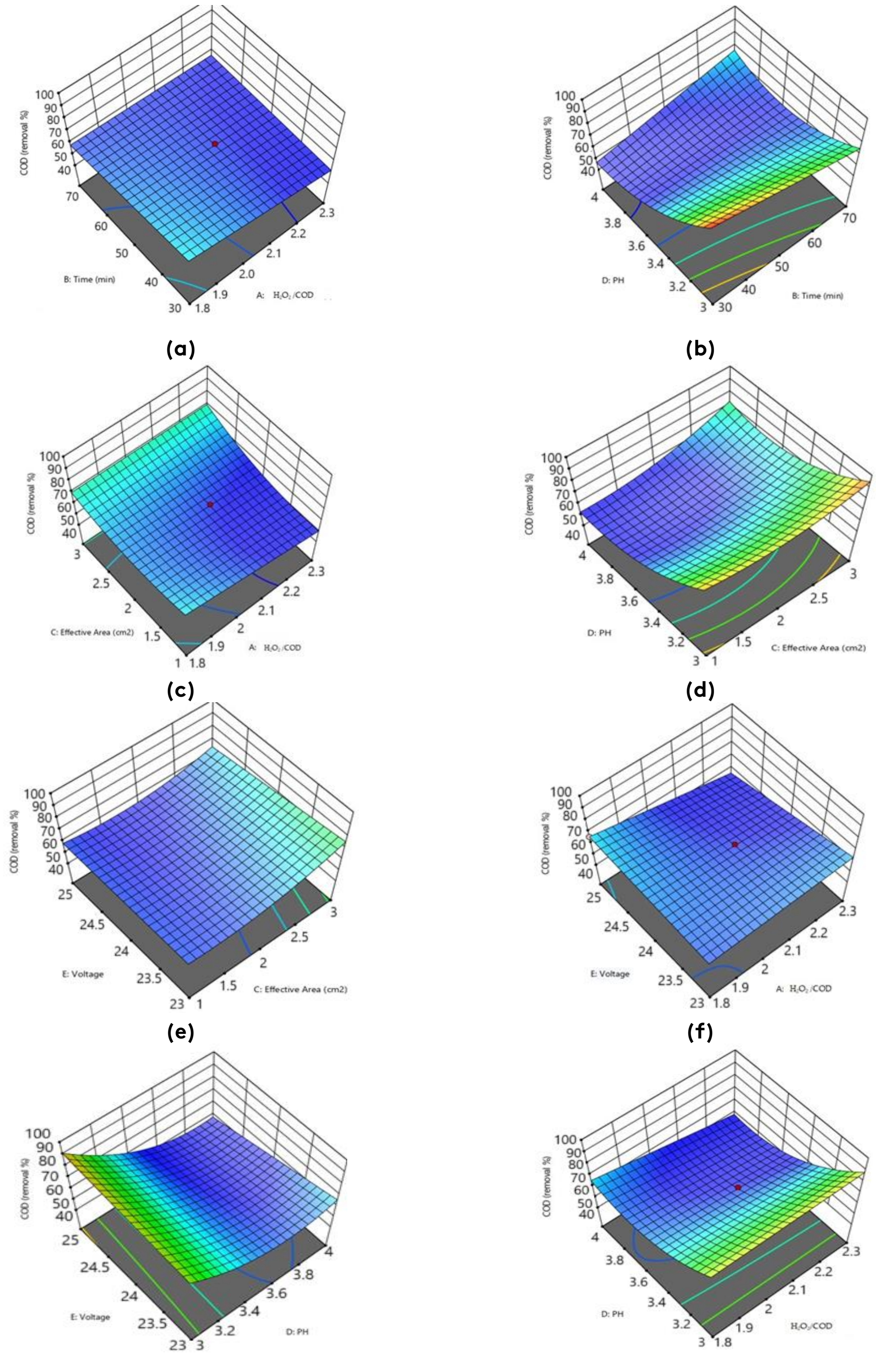
However, increasing the voltage beyond the optimal value causes excessive production of ferrous ions, and according to Reaction 6, the extra amount might react with H_2O_2 and diminish the EF process efficiency [22]. Experiments were carried out at various pH levels to investigate the impact of pH on COD removal during COD degradation (3, 3.5, and 4). It was found that the pH variation significantly affected the COD removal of CWW, as shown in Fig. 4(b, d, g, and h). As observed in Fig. 4, an increase in pH resulted in a notable decrease in COD removal, with improvement noted at a pH of 3. Thus, the optimum EF application was reached at pH=3 based on the plotted responses. The pH directly or indirectly affected both the amount and type of the Fe species and, thus, the amount of OH● radicals produced. It can be stated that pH could control the production of free radicals and the transformation of Fe^{3+}+ ions to Fe^{2+} ions in the EF process. The acidic pH range was mainly used as the optimum value for the EF process. The accuracy and precision of this method identified the optimum pH of around 3. At pH 3-4, the ferric species were prone to form precipitates, specifically ferric hydroxide complexes $Fe(OH)_3$. This sludge could have detrimental effects on the EF process, potentially diminishing the electrolytic cell's catalyst and impeding hydrogen peroxide's breakdown into oxygen and water. An important impact of the H_2O_2/COD ratio on the experiments aimed at decreasing COD concentration is illustrated in Fig. 4(a, c, f, and h), with an optimal H_2O_2/COD ratio achieved at 1.8. It is evident from the figures that an increase in the H_2O_2 ratio resulted in reduced COD removal. The excessive H_2O_2 quantity may lead to the generation of hydroperoxyl radicals ($OH^{\bullet}$) by reaction 8, which possess lower oxidizing power compared to hydroxyl radicals ($OH^{\bullet}$), resulting in a decrease in COD concentration.

The Design of Experiments (DOE) software identified the optimum conditions for COD removal from CWW. Statistical optimization was conducted to achieve the maximum COD elimination. The optimized conditions for 98.33% COD removal from wastewater were an H_2O_2/COD ratio of 1.8, a reaction time of 30 min, an effective surface area of 3 cm^2, pH of 3, and a voltage of 23 V. An experiment was conducted at the derived optimum conditions to confirm this result. In this case, a high COD removal rate of 97.99% was successfully achieved.

3.3 LAS and turbidity removal

LAS, an anionic surfactant, is the main component of most commercial detergents used in carpet cleaning facilities. Consequently, these substances are carried into the wastewater during the washing process. The removal of the surfactant during the EF process is presented in Table 5, and a significant elimination of LAS was observed in all experiments. The removal of LAS in the EF technique depends on generating °OH radicals through Fenton chain reactions. The applied voltage influences the production rate of H_2O_2 and the regeneration of Fe^{2+}. Therefore, LAS removal was achieved under the designed test conditions (relatively high applied voltage), ranging from 76.99% to 99.96% in all runs. Panizza et al. (2013) achieved a higher oxidation rate of anionic surfactants with an applied current of 2A and a pH of 3, almost identical to the applied current of 2.5A in the present study [23].

Turbidity is defined as the measure of clarity or cloudiness of a solution due to the presence of suspended particles that are generally visible. The unit of turbidity is the nephelometric turbidity unit (NTU). Generally, turbidity increases significantly when the solution's pH is acidic, and a shorter processing time is more suitable for turbidity elimination. As shown in Table 5-b, all process conditions were suitable for turbidity degradation.



(a) (b)

(c) (d)

(e) (f)

(g) (h)

Fig. 4. Effects of the variables on COD removal by EF process: (a) reaction time and H_2O_2/COD ratio, (b) reaction time and effective surface area, (c) reaction time and pH, (d) effective surface area and H_2O_2/COD ratio, (e) effective surface area and voltage, (f) effective surface area and pH, (g) voltage and H_2O_2/COD ratio (h) voltage and pH, and (i) pH and H_2O_2/COD ratio.

Table 5. LAS removal (%) and turbidity removal (%) by EF process.

Runs	(a) LAS removal (%)	(b) Turbidity removal (%)
1	99.92	99.99
2	99.96	99.71
3	99.46	96.75
4	88.49	99.99
5	97.89	99.11
6	96.14	99.99
7	99.94	99.99
8	99.78	99.49
9	99.86	96.69
10	99.90	98.83
11	99.92	99.99
12	99.92	99.41
13	99.94	98.76
14	98.28	99.99
15	99.92	99.83
16	99.84	99.72
17	99.88	99.91
18	99.94	99.41
19	98.64	99.99
20	98.24	99.99
21	99.92	98.64
22	99.80	99.99
23	99.92	99.67
24	99.92	99.99
25	99.27	99.99
26	88.49	99.99
27	99.92	99.99
28	99.98	99.99
29	99.76	99.99
30	99.92	91.99
31	98.85	99.99

Other additional factors, including a BOD5 value of 5 mg/l, a pH level of 6.3, a color measurement of 0.3 TCU, a TSS level of 75.5 mg/l, and a TDS/EC ratio of 0.57, were verified using established procedures outlined in the Standard Methods for the Examination of Water and Wastewater publication. These parameters were found to be within the standard range of wastewater discharge [24].

4. Conclusions

In this study, the Electro-Fenton process disposed of carpet-washing wastewater to remove COD, turbidity, and LAS surfactant. Effects of various parameters, including H2O2/COD ratio (1.8 – 2.3), reaction time (30–70 min), effective surface area (1-3 cm^2), pH (3–4), and voltage (23–25 volt), were studied to evaluate the efficiency of the treatment. The application of Box–Behnken experimental design and RSM in producing quadratic models for the EF process with high significance and high R^2 coefficients was successful. The findings indicated that the response surface models were reliable in forecasting experimental outcomes and demonstrated the ability to represent both the individual and combined impacts of important factors concerning COD and LAS elimination. An H_2O_2/COD ratio of 1.8, a reaction time of 30 min, an effective surface area of 3 cm^2, a pH of 3, and a voltage of 23 volts were the optimum conditions for achieving 97.99% COD removal, 99.92 % LAS removal, and 99.99 % turbidity removal. At the optimal condition process, the effluent had a BOD5= 5 mg/l, a pH=6.3, color= 0.3 TCU, TSS= 75.5 mg/l, and a TDS/EC ratio= 0.57. This study aimed to evaluate using EF technology for carpet washing wastewater. As a future study, the authors recommend conducting pilot-scale experiments to check the amenability of the oxidation system to large fluctuations in organic loading. They also suggest studying energy and recovery processes to make the system more sustainable.

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References

[1] Maryam, B., & Büyükgüngör, H. (2019). Wastewater reclamation and reuse trends in Turkey: Opportunities and challenges. *Journal of Water Process Engineering, 30* , 100501. https://doi.org/10.1016/j.jwpe.2017.10.001

[2] Chojnacka, K., Witek-Krowiak, A., Moustakas K., Skrzypczak, D., Mikula, K., & Loizidou, M. (2020). A transition from conventional irrigation to fertigation with reclaimed wastewater: Prospects and challenges. *Renewable and Sustainable Energy Reviews, 130* , 109959. https://doi.org/10.1016/j.rser.2020.109959

[3] Ajiboye, T. O., Oyewo, O. A., & Onwudiwe, D. C. (2021). Simultaneous removal of organics and heavy metals from industrial wastewater: A review. *Chemosphere, 262*,128379. https://doi.org/10.1016/j.chemosphere.2020.128379

[4] Azanaw, A.,Birlie, B., Teshome, B., & Jemberie, M.(2022) Textile effluent treatment methods and eco-friendly resolution of textile wastewater. *Case Studies in Chemical and Environmental Engineering,6*, 100230. https://doi.org/10.1016/j.cscee.2022.100230

[5] Babuponnusami, A., Sinha, S., Ashokan, H., Paul, M.V., Hariharan, S.P., Arun, J., Gopinath, K., Le, Q. H., & Pugazhendhi, A.(2023). Advanced oxidation process (AOP) combined biological process for wastewater treatment: A review on advancements, feasibility and practicability of combined techniques. *Environmental research, 237(1),* 116944 . https://doi.org/10.1016/j.envres.2023.116944

[6] Sarkar, R., Pal, A., Rakshit, A., & Saha, B.(2021). Properties and applications of amphoteric surfactant: A concise review. *Journal of Surfactants and Detergents, 24* (5) 709-730. https://doi.org/10.1002/jsde.12542

[7] Mustan, F., Politova-Brinkova, N.,Vinarov, Z., Rossetti, D., Rayment, P., &Tcholakova. S. (2022). Interplay between bulk aggregates, surface properties and foam stability of nonionic surfactants. *Advances in colloid and interface science, 302* 102618. https://doi.org/10.1016/j.cis.2022.102618

[8] Inès. M., Mouna, B., Marwa, E.,& Dhouha, G. (2023). Biosurfactants as emerging substitutes of their synthetic counterpart in detergent formula: efficiency and environmental friendly. *Journal of Polymers and the Environment, 31* (7) 2779-2791. https://doi.org/10.1007/s10924-023-02778-1

[9] Ahmad, I., Abdullah, N., Koji, I., Yuzir, A., Mohamad, S. E., Show, P. L., Cheah, W. Y., & Khoo, K.S. (2022). The role of restaurant wastewater for producing bioenergy towards a circular bioeconomy: A review on composition, environmental impacts, and sustainable integrated management. *Environmental Research, 214* , 113854. https://doi.org/10.1016/j.envres.2022.113854

[10] Saleh, T. A., Mustaqeem, M.,& Khaled, M. (2022). Water treatment technologies in removing heavy metal ions from wastewater: A review. *Environmental Nanotechnology, Monitoring & Management, 17*, 100617. https://doi.org/10.1016/j.enmm.2021.100617

[11] Kumar, A., Thakur, A., Panesar, P. S. (2023). A review on the industrial wastewater with the efficient treatment techniques. *Chemical Papers, 77* (8), 4131-4163. https://doi.org/10.1007/s11696-023-02779-3

[12] Domingues, E., Silva, M. J., Vaz, T., Gomes, J.,& Martins, R. C.(2022). Sulfate radical based advanced oxidation processes for agro-industrial effluents treatment: A comparative review with Fenton's peroxidation. *Science of the Total Environment, 832* , 155029. https://doi.org/10.1016/j.scitotenv.2022.155029

[13] Khan, Z.U.H., Gul, N.S., Sabahat, S., Sun, J., Tahir, K., Shah, N. S., Muhammad, N., Rahim, A., Imran, M., & Iqbal J.(2023). Removal of organic pollutants through hydroxyl radical-based advanced oxidation processes. *Ecotoxicology and Environmental Safety, 267* , 115564. https://doi.org/10.1016/j.ecoenv.2023.115564

[14] Divyapriya, G.,& Nidheesh, P.V.(2020). Importance of graphene in the electro-Fenton process. *ACS omega, 5* (10), 4725-4732. https://doi.org/10.1021/acsomega.9b04201

[15] Deng, F., Olvera-Vargas, H., Zhou, M., Qiu, S., Sirés, I.,& Brillas, E.(2023). Critical review on the mechanisms of Fe2+ regeneration in the electro-Fenton process: Fundamentals and boosting strategies. *Chemical Reviews, 123* (8) 4635-4662. https://doi.org/10.1021/acs.chemrev.2c00684

[16] Xia, P., Zhang, H.,&Ye, Z. (2024). Recent advances in the application of natural iron and clay minerals in heterogeneous electro-Fenton process. *Current Opinion in Electrochemistry,* 101495. https://doi.org/10.1016/j.coelec.2024.101495

[17] Lee, B., Mahtab, M., Neo, T., Farooqi, I., & Khursheed, A. (2022). A comprehensive review of Design of experiment (DOE) for water and wastewater treatment application-Key concepts, methodology and contextualized application. *Journal of Water Process Engineering, 47*, 102673. https://doi.org/10.1016/j.jwpe.2022.102673

[18] Mariah, G.K.,& Pak, K.S.(2020). Removal of brilliant green dye from aqueous solution by electrocoagulation using response surface methodology. *Materials today: proceedings, 20* 488-492. https://doi.org/10.1016/j.matpr.2019.09.175

[19] Ahmad, A., Yadav, A., & Singh, A. (2024). Process optimization of spirulina microalgae biodiesel synthesis using RSM coupled GA technique: a performance study of a biogas-powered dual-fuel engine. *International Journal of Environmental Science and Technology, 21* (1) 169-188. https://doi.org/10.1007/s13762-023-04948-z

[20] Varank, G., Guvenc, S.Y., Can-Güven, E., Yokus, S., & Bilgin, O.N. (2024). Hydrogen peroxide assisted advanced electrocoagulation for reducing the organic content of leachate nanofiltration concentrate. *Journal of Industrial and Engineering Chemistry, 132* 369-382. https://doi.org/10.1016/j.jiec.2023.11.031

[21] Dehboudeh, M., Dehghan, P., Azari, A.,& Abbasi, M.(2020). Experimental investigation of petrochemical industrial wastewater treatment by a combination of integrated fixed-film activated sludge (IFAS) and electro-Fenton methods. *Journal of Environmental Chemical Engineering, 8* (6), 104537. https://doi.org/10.1016/j.jece.2020.104537

[22] Seibert, D., Zorzo, C. F., Borba, F. H., de Souza, R. M.,Quesada, H. B., Bergamasco, R., Baptista, A. T., & Inticher, J. J. (2020). Occurrence, statutory guideline values and removal of contaminants of emerging concern by Electrochemical Advanced Oxidation Processes: A review. *Science of The Total Environment, 748*, 141527. https://doi.org/10.1016/j.scitotenv.2020.141527

[23] Panizza, M., Barbucci, A., Delucchi, M., Carpanese, M., Giuliano, A., Cataldo-Hernández, M.,& Cerisola, G. (2013). Electro-Fenton degradation of anionic surfactants. *Separation and Purification Technology, 118* 394-398. https://doi.org/10.1016/j.seppur.2013.07.023

[24] Rice E. W.,Baird R. B.,Eaton A. D. *Standard methods for the examination of water and wastewater;* American Public Health Association(APHA), American Water Works Association(AWWA), Water Environment Federation(WEF), 2017. https://doi.org/10.2105/SMWW.2882.216.

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