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Multi-objective optimization of electrocoagulation-flotation (ECF) process for treatment of real dairy wastewater

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ABSTRACT

The present work reports treatment of real dairy products wastewater by electrocoagulation–flotation (ECF). The process was performed in a batch stainless steel reactor equipped with aluminum electrodes. The model attributed to batch ECF process was optimized employing response surface methodology (RSM) via operating variables viz. pH, current intensity, and electrolysis time in view of chemical oxygen demand (COD), total kjeldahl nitrogen (TKN), total phosphorus (TP), and turbidity, as well as fat-oil-grease (FOG) removal from wastewater. Sludge settling characteristics, specific electric energy consumption (SEEC), and biodegradability of treated effluent were also analyzed. The *p*-values with low probability (<0.05), determination coefficient (R^2 = 0.61–0.97) and the non-significant lack of fit (p > 0.05) showed quadratic models with a good fit with experimental terms. The process was favored at pH, 8; current intensity, 3A; and electrolysis time, 45 min. Under optimized conditions, removal of COD, TKN, TP, and FOG were 66%, 73%, 98%, and 97%, respectively. The results further indicated an improved biodegradability of treated effluent in terms of BOD_5/COD ratio (0.79 as compared to 0.41 in raw wastewater) under optimum conditions. Moreover, sludge volume index and SEEC were ~86 mL/g and 0.069 kWh/kg COD, respectively. The enhanced performance of this promising electrochemical process in the treatment of real dairy wastewater might be explained by a combination of direct coagulation/floatation of suspended organics/inorganics and indirect oxidation–reduction of dissolved chemicals by oxygen bubbles in the anode and hydrogen in the cathode, rendering it amenable to the final aerobic treatment.

Keywords: Dairy wastewater; Electrocoagulation–flotation (ECF); Energy consumption; Biodegradability; Response surface methodology (RSM)

1. Introduction

Dairy product wastewaters are characterized by high organic content, fat, nutrients, and variations in pH (4.2–11), as well as a high load of suspended solids (0.024–4.5 g/L) [1–4]. Treatment of such wastewaters to an acceptable level before discharging into the natural environment represents a challenge for dairy industries [5,6]. To date, several technologies have been developed to suppress dairy effluents' pollution. Given the high organic load of such wastewaters,

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anaerobic biological processes have been widely preferred for dairy wastewater treatment [7], while additional treatment is requited before final discharge into the environment. Meanwhile, except for proteins and fats which are usually persistent to biological treatment, all other components of dairy wastewater are easily biodegradable causing aesthetic concerns particularly in the absence of sufficient oxygen. On the other hand, the aforementioned constituents act as inhibitory compounds during anaerobic treatment of dairy wastewaters. Given the relatively low biochemical oxygen demand/chemical oxygen demand (BOD_5/COD) ratios of dairy product wastewaters (0.2–0.5), it can be inferred that such wastewaters are likely to be dealt with biological remediation. The treatment plant design may then focus on the fat-oil-grease (FOG) and organics loading rate, which in turn can become sources of not scent strong odor. Notwithstanding the organic strength of dairy products wastewater, choose of an anaerobic biological process for organics reduction before aerobic treatment would probably not be an appropriate strategy.

Electrocoagulation/floatation (ECF), as an attractive and favorable electrochemical technique, has been effectively used to remove various environmental contaminants including synthetic dyes [8,9], nutrients [10], organics [11], and heavy metals [12]. Environmental compatibility, versatility, energy efficiency, safety, selectivity, amenability to automation, easy applicability, and cost-effectiveness are the benefits of such techniques.

During the ECF process, metallic hydroxides are formed within the water/wastewater by electro dissolution of anode(s) which are usually made of iron or aluminum. The dominate path of pollutants' removal, sedimentation, or floatation, during the electrocoagulation process is governed by the magnitude of the applied current, where the floatation is the prevailing path at high currents, while sedimentation is dominated at low currents [10]. The main reactions governing ECF process for aluminum or iron electrodes have been previously described [13].

The amorphous metallic hydroxides (e.g., $Al(OH)_3$ (s), $Fe(OH)_3$ (s)), having great surface area, contribute to quick adsorption of soluble organic compounds and also trap the colloidal particles. The flocs can be easily separated from aqueous medium through H_2 flotation or sedimentation [14]. Consequently, the removal mechanism of pollutants from wastewater by ECF is attributed to the formation of metallic hydroxides, as well as monomeric and polymeric metal species as a result of coagulation, precipitation, co-precipitation, and electro-oxidation. Electrocoagulation may also assist with oxidation–reduction reactions; some species may be oxidized on the anode and other species may be reduced on the cathode [4].

The success of ECF has been proved in the treatment of urban, industrial, and agricultural wastewaters, as well as land fill leachate and ground water polluted by heavy metals [15–25]. To date, considerable efforts have also been devoted to remediate industrial synthetic wastewaters by ECF (including dairy wastewater [26]); however, there is still a dearth in investigating real dairy industry wastewater treatment using ECF process. Also, there are few studies in the literature utilizing EC treatment successfully for dairy wastewaters. For example, in the recent years, Bruguera-Casamada et al. [27] studied the EC treatment with iron electrodes whereas in several studies aluminum electrodes have been used for the same process [3,15,28,29]. Valente et al. [30] studied the effects of EC process on the treatment of dairy plant wastewater using iron electrodes. From the battery of performance criteria, the removal of turbidity as well as total, and volatile suspended solids were marked as the most susceptible to the process; other parameters including COD removal did not show clear patterns of change. Sengil and Oscazar [4] also investigated the treatment of dairy wastewater using parallel mild steel electrodes.

In the present study, we attempted to make use of ECF effect to alleviate the strength and also to improve the biodegradability of real dairy product wastewater. The effectiveness of this new cost-effective electrochemical process was determined while aluminum and steel (the body of reactor) were used as electrodes. To go further in our understanding of the ECF effectiveness, the COD, turbidity, total Kjeldahl nitrogen (TKN), total phosphorous (TP), and FOG removals were determined as a function of operating variables (i.e., pH, current intensity, electrolysis time, and the content of NaCl). To our best knowledge, the report about assistance of ECF in improving biodegradability of real dairy product wastewater is presently lacking in the literature. Hence, it is necessary to optimize ECF in order to achieve maximum process efficiency, taking simultaneously into account the value and characteristics of affecting parameters. For this purpose, optimization of desired responses, estimation of the impact of input variables on response(s), as well as formulation of regression models are usually carried out using response surface methodology (RSM) [14,31,32]. RSM has been proved to be a powerful statistical technique in designing the experiments and optimizing different environmental processes in which a response of interest is influenced by several variables and the objective is to optimize this response [33,34]. Likewise, among many experimental design methods, central composite designs (CCD) provides high quality predictions in studying linear, quadratic and interaction effects of factors influencing a system and is the most-used design method for the creation of second-order response surface model in environmental processes [35].

2. Materials and methods

2.1. Wastewater sampling and characterization

The present study was carried out on real wastewater samples obtained from a milk-processing factory in Tehran, Iran. This factory rejects an average daily volume of 80,000 L of wastewater. It produces milk, cheese, cream, and yogurt. Table 1 shows the characteristics of samples determined according to the standard methods [36] after preservation at 4°C.

2.2. Chemicals and experimental setup

All the chemicals used in the experiments were of analytical-reagent grade. HCl or NaOH 1 N were used to adjust pH of samples. NaCl was used as the supporting electrolyte to set the conductivity and avoid passivation of the electrodes.

ECF batch experiments were conducted using a bench-scale non-magnetic stainless steel cylindrical reactor having a total volume of 3.52 L with 25 cm height and 13.4 cm diameter. Three parallel plates of unalloyed aluminum as anode and two plates of steel as cathode electrodes, with dimensions of 22 cm × 8 cm × 2 mm were used. The electrodes were connected to each other in a bipolar-serial (BP-S) way with an inner distance of 2 mm (Fig. 1). The body of reactor was also considered to as cathode. For each test, 2 L wastewater sample was used. Before each run, electrodes were washed with HCl 1 N and the impurities on electrode surfaces were removed. Electrodes were connected to a regulated (RXN-605D) DC power supply. The reactor was stirred by a magnetic stirrer (JENWAY 1000-England) at 500 rpm for mixing reactor contents. Treated samples were allowed to settle for 30 min, followed by supernatant collection for analyzing. Standard methods were employed for the measurement of desired analyses. The determination procedures are detailed in the Standard Methods for the examination of water and wastewater [36]. All measurements were tripled and the arithmetic average of the three measurement results was reported here. The process efficiency (removal percentage) was calculated using Eq. (1).

Table 1
Characteristics of the primary sedimentation tank effluent

Parameters	Value
pH	6.5
Turbidity, NTU	1,277
Total COD, mg/L	3,060
(COD soluble), %	65.1
(COD particulate), %	34.9
BOD_5, mg/L	1,260
BOD_5/COD ratio	0.41
Chlorides, mg/L	380
Electric conductivity, µs/cm	2,850
TKN, mg/L	200.75
Total alkalinity, mg/L as $CaCO_3$	1,400
Total phosphorus, mg/L	25.6
FOG, mg/L	2.63
TSS, mg/L	618
TDS, mg/L	2,574
TS, mg/L	3,192
Color, platinum cobalt	3,284

$$R\,\% = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

where C_0 and C_e denote the initial and final COD, TP, FOG, TKN concentration (mg/L) or turbidity, respectively [14].

2.3. Response surface modeling

Experiments were launched as a preliminary assessment through the one factor at a time (OFAT) method for determining a narrower range of operating variables before designing the experimental runs. Accordingly, a wide range of pH (4, actual wastewater, 9), electrolysis time (10, 30, and 60 min), electrical conductivity (that was achieved by adding 0.2, 0.5, 1, and 1.5 g NaCl/L to the sample of the real

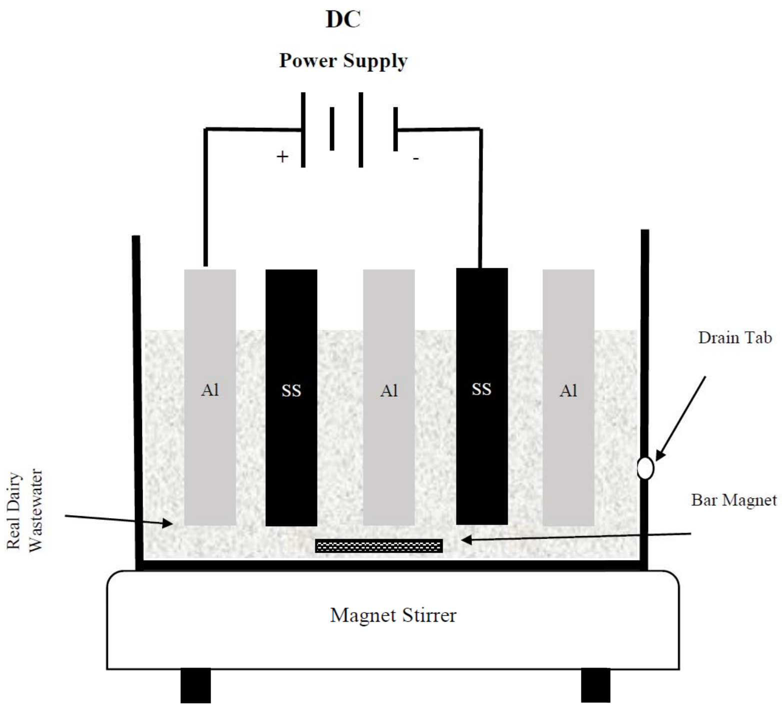

Fig. 1. Schematic diagram of the electrocoagulation–flotation reactor.

wastewater) and current intensity (0.25, 0.5, 0.75, 1, and 5 A) were tried to search for a narrower and more effective range, considering appreciable achievements in the process responses. Accordingly, experimental runs were confined to pH, electrolysis time, and current intensity, aside from electrical conductivity due to its trivial effect on process performance. Designing of experiments in the second phase was conducted using Design-Expert software, version 8 (Stat-Ease Inc., Minneapolis, USA) based on RSM/CCD. Each factor was studied at two different factorial levels (–1,+1), two axial points (–2,+2), and a center point (0) which is the midpoint of each variable range. The ranges of variables and experimental conditions are depicted in Table 2. Actually, CCD consists of $2^k + 2k + n$ runs, where k denotes the number of factors, 2^k is the number of the factorial points at the corners of the cube, $2k$ is the number of the axial points on the axis of each design factor, and n stands for the number of the replication of center points at the center of the cube. Accordingly, the experiments consisted of eight factorial, six axial, and six center points. The total number of experiments in our study was therefore 8 + 6 + 6 = 20 runs.

Considering the general function expressing the interaction between the independent and dependent variables, the response surface procedure was employed to fit the following second-order model [Eq. (2)]:

$$Y = b_0 + \sum b_i X_i + \sum b_{ii} X_i^2 + \sum b_{ij} X_i X_j + \varepsilon \quad (2)$$

where Y_i is the value of the pollutant removal, b_0 the constant coefficient, b_i the linear coefficients, b_{ii} the quadratic coefficients, b_{ij} the interaction coefficients, and X_i, X_j the coded values of the factors [26].

The experimental data was fitted using sequential analysis of variance (ANOVA) modelling and the best model was developed based on the p-value ($p < 0.05$) and non-significant ($p > 0.05$) lack-of-fit (LOF). The final model was selected based on coefficient of determination (R^2), predicted residual error sum of squares (PRESS), and coefficient of variation (CV%).

2.4. Energy consumption

Cost is an important parameter that greatly determines the feasibility of any process of wastewater treatment. In addition to aluminum electrodes consumed in the EC process, the major operating cost is for electric energy consumption (EEC). The specific electric energy consumption (SEEC) is defined as the amount of energy consumed per unit mass of pollutant removed [37]. SEEC was calculated using Eq. (3):

$$\text{SEEC} = \frac{I \cdot U \cdot t}{(X_0 - X_t)V} \quad (3)$$

where I, U, t, V are the current intensity (A), the average cell voltage (V), the electrolysis duration time (hour) and the volume of treated sample (L), respectively. Likewise, X_0 and X_t are the initial and at desired time (t) concentrations (mg/L) of COD.

3. Results and discussion

3.1. Response analysis by central composite design

The CCD matrix coupled with the experimental results of removal efficiencies using the ECF are shown in Table 3. According to the obtained results, the highest removal efficiencies for COD, TKN, TP, and FOG were 67.61%, 73.47%, 98.55%, and 97.46%, respectively. The experimental data were fitted to various mathematical models (linear, interactive, quadratic, and cubic) in order to obtain regression equations. Model summary statistics were performed to decide about the most suitable model [38]. Accordingly, the quadratic models were selected for further analysis to represent the ECF process, eliminating the insignificant model terms to improve the model. The final quadratic equations considering study parameters were obtained, as shown in Eqs. (4)–(7).

$$\text{COD removal (\%) } Y_1 = -206.61 + 70.35\,[\text{pH}] - 4.93\,[\text{Time}] + 70.54\,[I] + 0.042\,[\text{pH}][\text{Time}] - 1.66\,[\text{pH}][I] - 0.05\,[\text{Time}][I] - 3.95\,[\text{pH}]^2 + 0.037\,[\text{Time}]^2 - 9.59\,[I]^2 + 0.01\,[\text{pH}][\text{Time}][I] \quad (4)$$

$$\text{TKN removal (\%) } Y_2 = -81.15 + 18.19\,[\text{pH}] + 1.32\,[\text{Time}] + 20.58\,[I] - 0.17\,[\text{pH}][\text{Time}] - 2.44\,[\text{pH}][I] + 0.016\,[\text{Time}][I] \quad (5)$$

$$\text{TP removal (\%) } Y_3 = -91.03 + 52.09\,[\text{pH}] - 1.05\,[\text{Time}] - 5.73\,[I] + 0.07\,[\text{pH}][\text{Time}] + 0.95\,\text{pH}][I] - 0.02\,[\text{Time}][I] - 3.42\,[\text{pH}]^2 + 4.24\text{E-}003[\text{Time}]^2 - 2.00\text{E-}003\,[I]^2 \quad (6)$$

$$\text{FOG removal (\%) } Y_4 = -750.81 + 222.2\,[\text{pH}] - 6.18\,[\text{Time}] + 27.93\,[I] + 0.14\,[\text{pH}][\text{Time}] - 0.61\,[\text{pH}][I] - 0.11\,[\text{Time}][I] - 13\,[\text{pH}]^2 + 0.05\,[\text{Time}]^2 - 2.22\,[I]^2 \quad (7)$$

The data demonstrate excellent fits to the second-order model with high R^2. Good fits can also be confirmed by large F-values and small p-values (<0.05).

Adequate precision (AP) is a measure of predicted response value range at the design points relative to associated

Table 2
Independent variables and limit level for response surface study

Variables	Coded symbols	Levels				
		–2	–1	0	+1	+2
pH	*A*	8.0	8.5	9.0	9.5	10.0
Reaction time, min	*B*	45.0	52.5	60.0	67.5	75.0
Intensity of current, A	*C*	1.50	2.25	3.00	3.75	4.5

Table 3
CCD for the experimental variables

Run no.	Independent variables			Removal efficiency (%)			
	A	*B*	*C*	COD	TKN	TP	FOG
1	10	75	1.5	44.25	69.17	92.93	88.59
2	9.5	60	3	61.76	70.01	94.22	90.19
3	8	75	4.5	54.9	72.75	98.2	97.46
4	10	75	4.5	54.8	54.85	96.72	88.59
5	9	60	3.75	56.5	66.54	97.3	92.59
6	10	45	1.5	40.67	73.47	85.2	69.58
7	9	60	3	65.03	69.43	98.55	96.19
8	9	60	3	65.1	65.73	97.84	95.9
9	9	60	3	62.9	67.7	96.35	94.72
10	9	67.5	3	67.61	64.06	96.72	96.88
11	8	75	1.5	41.17	59.27	95.43	92.39
12	9	52.5	3	60.53	72.21	97.85	95.77
13	9	60	3	64.1	66.61	97.9	96.57
14	8.5	60	3	60.18	70.61	98.16	90.79
15	8	45	1.5	41.5	66.38	96.87	83.38
16	9	60	3	64.03	64.38	96.34	96.68
17	10	45	4.5	42.81	70.83	95.86	80.98
18	9	60	2.25	56.62	66.68	96.79	92.39
19	8	45	4.5	49.54	65.2	97.11	97.09
20	9	60	3	65.03	69.22	97.65	95.16

mean prediction error; as the other words, a signal-to-noise ratio. The desired ratio is 4 or more [39]. AP values for the removal of COD, TKN, TP, and FOG were all greater than 4, exploring the existence of adequate signal and high capability of developed models in predicting the results.

Meanwhile, coefficient of variance (CV), also known as relative standard deviation (RSD), describes the precision and reproducibility of a model. Normally, we can consider a model to be reproducible when its CV is not greater than 10% [40]. In our experiments, CV was within the acceptable range of 10% which proves the reproducibility of the model and high precision of obtained results.

The lack of fit *F*-test describes the variation of data around the fitted model. If the model does not fit the data well, the LOF test will be significant [40]. Lack of fit test for TKN and TP responses were not statistically significant, reinforcing a good fitting of the data to the model. Nevertheless, it was significant with regard to COD and FOG, indicating that the points were not properly distributed around the model. Thus, these models are not assumed to predict further values of relevant variables [41].

The response surface plots shows that optimal conditions for the maximum responses depend on the pH, electrolysis time and current intensity in the design space. The interaction effects of current intensity and pH on responses demonstrate concentrically closed curves as depicted in Fig. 2. The optimum conditions indicate that experimental design has been well conducted. The experimental results of the optimum conditions could be assigned to the precision of the data and models. Accordingly, the suitable model was selected for further analysis to represent the ECF process.

The results obtained from variance analysis (ANOVA) are shown in Table 4. As can be seen, all the terms in the statistical quadratic model were significant ($p \leq 0.0001$). Lack of fit tests for TKN and TP removals ($p = 0.0697$, 0.0738, respectively) were not significant, but it was significant for COD and FOG removal ($p = 0.007$, 0.0084, respectively). The model equations were also employed to generate 3D contour plots. The 3D contour plots for the effective factors in eliminating COD, TKN, TP, and FOG are also plotted in Fig. 2. As shown, all responses have an optimum removal point.

3.2. Effect of operating variables

The impact of the nature and concentration of electrolyte in ECF process efficiency have been widely investigated [42]. Using sodium chloride to increase the electric conductivity has often outcompete other compounds such as potassium chloride, sodium nitrate, etc. [43]. Chloride ions have been shown to decline undesirable effects compared with other ions such as HCO_3^- by preventing calcium settlement in wastewater. In addition, sedimentation of sulfate and carbonate on the electrode surface can reduce the process efficiency. Actually, sedimentation as a layer of isolation on the surface of electrodes can restrain metal from dissolving.

In a constant current intensity, the increase of the electrolyte concentration declines voltage between electrodes. According to Hung et al., chloride ions in the solution containing $Al(OH)_3$ can form some types of $Al(OH)_2Cl$, $Al(OH)Cl_2$, and $AlCl_3$ [37,44]. Formation of $AlCl_3$ in the presence of extra chloride ion contributes to decreasing efficiency of

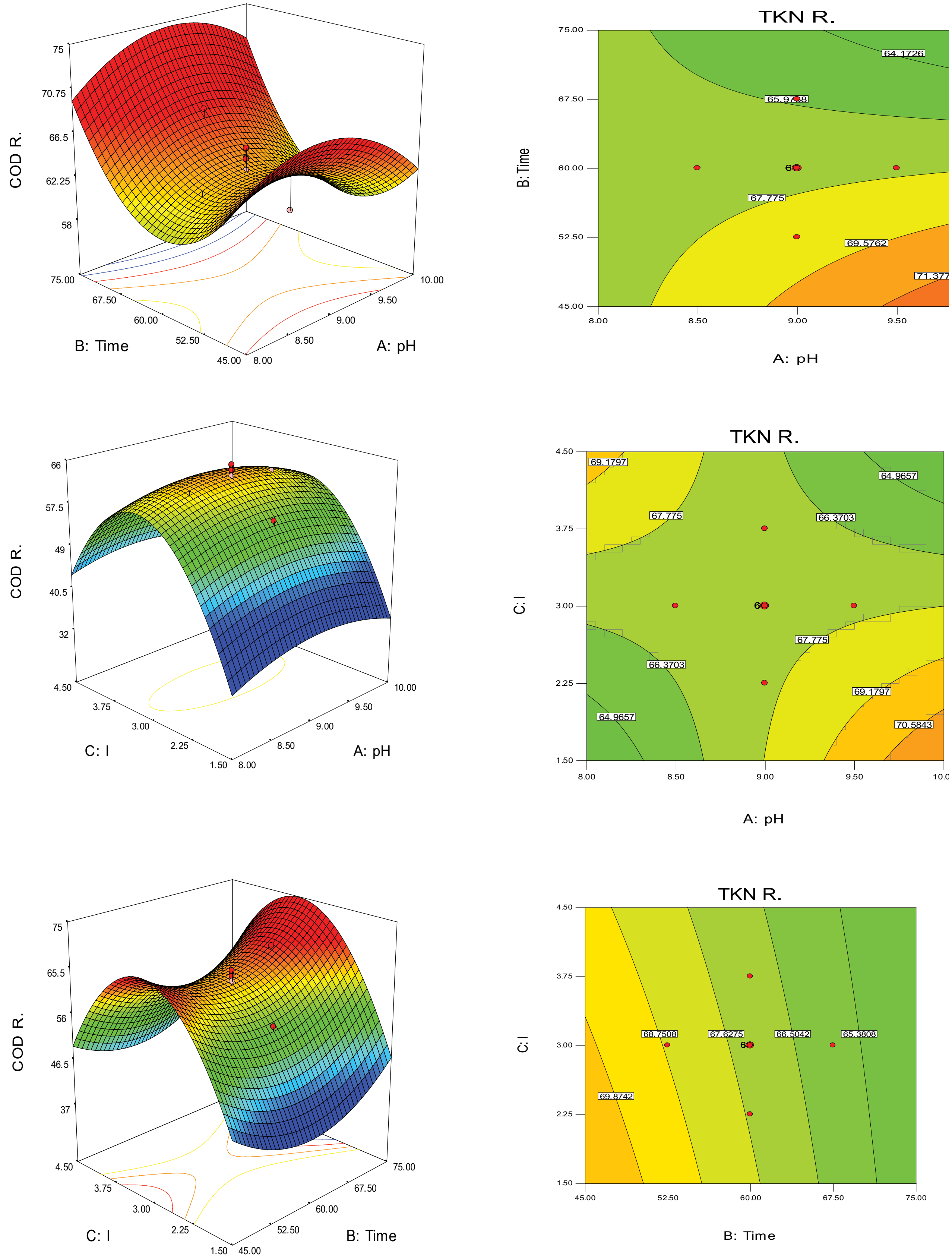

Fig. 2. *Continued*

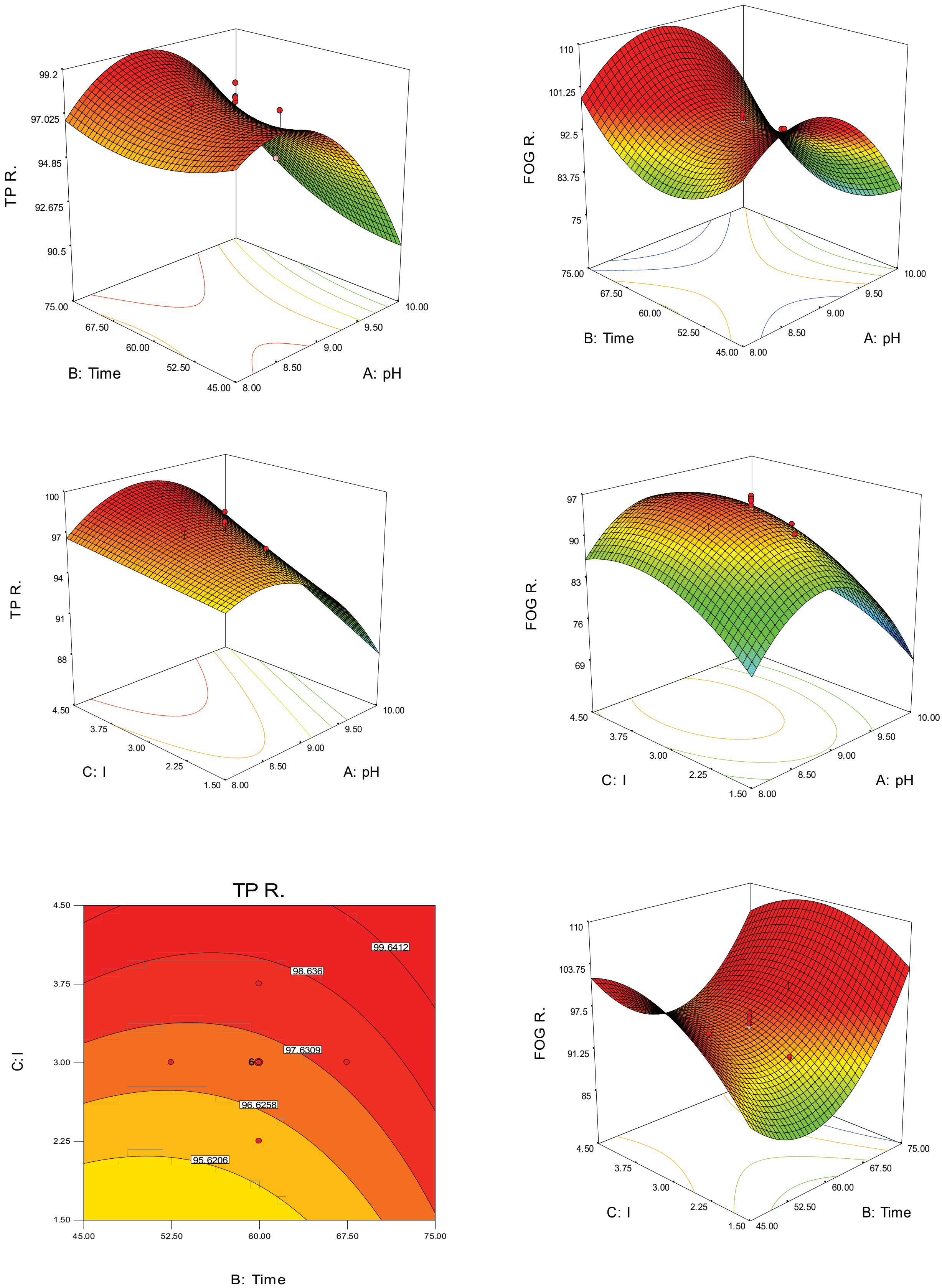

Fig. 2. 3D and contour plots for the removal of COD, TKN, TP, and FOG by ECF process.

Table 4
ANOVA results for the removal of COD, TKN, TP, and FOG using ECF process

Source of variations	Sum of squares	Mean square	Coefficient estimate	Standard error	*F*-value	*p*-value
			COD removal (%)			
Model	1,609.57	160.96			34.34	<0.0001
Intercept	–	–	63.21	0.65	–	–
[pH]	1.69	1.69	–0.45	0.74	0.36	0.5630
[Time]	68.56	68.56	2.84	0.74	14.63	0.0041
[*I*]	139.22	139.22	4.05	0.74	29.70	0.0004
[pH][Time]	13.89	13.89	1.32	0.77	2.96	0.1193
[pH][*I*]	10.31	10.31	–1.14	0.77	2.20	0.1723
[Time][*I*]	24.85	24.85	1.76	0.77	5.30	0.0468
$[pH]^2$	2.91	2.91	–3.95	5.01	0.62	0.4510
$[Time]^2$	13.33	13.33	8.45	5.01	2.84	0.1260
$[I]^2$	86.98	86.98	–21.59	5.01	18.56	0.0020
[pH][Time][*I*]	0.92	0.92	0.34	0.77	0.20	0.6674
Residual	42.18	4.69	–	–	–	–
Lack of fit	38.43	9.61	–	–	12.80	0.007
Pure error	3.75	0.75	–	–	–	–
			TKN removal (%)			
Model	234.01	39.00			3.42	0.0298
Intercept	–	–	67.26	0.75	–	–
[pH]	2.30	2.30	0.52	1.16	0.20	0.6607
[Time]	67.29	67.29	–2.81	1.16	5.90	0.0303
[*I*]	2.63	2.63	–0.56	1.16	0.23	0.6388
[pH][Time]	53.66	53.66	–2.59	1.19	4.71	0.0491
[pH][*I*]	107.02	107.02	–3.66	1.19	9.39	0.0090
[Time][*I*]	1.11	1.11	0.37	1.19	0.097	0.7599
Residual	148.14	11.40	–	–	–	–
Lack of fit	128.38	16.05	–	–	4.06	0.0697
Pure error	19.76	3.95	–	–	–	–
			TP removal (%)			
Model	142.87	15.87	–	–	7.69	0.0019
Intercept	–	–	97.25	0.43	–	–
[pH]	41.89	41.89	–2.22	0.49	20.30	0.0011
[Time]	6.93	6.93	0.90	0.49	3.36	0.0968
[*I*]	36.92	36.92	2.08	0.49	17.89	0.0017
[pH][Time]	9.99	9.99	1.12	0.51	4.84	0.0524
[pH][*I*]	16.36	16.35	1.43	0.51	7.93	0.0183
[Time][*I*]	2.35	2.35	–0.54	0.51	1.14	0.3106
$[pH]^2$	2.19	2.19	–3.42	3.33	1.06	0.3274
$[Time]^2$	0.17	0.17	0.96	3.33	0.083	0.7797
$[I]^2$	3.805E-006	3.805E-006	–4.51E-003	3.33	1.84E-006	0.9989
Residual	20.64	2.06	–	–	–	–
Lack of fit	16.59	3.32	–	–	4.10	0.0738
Pure error	4.05	0.81	–	–	–	–

(Continued)

Table 4 Continued

Source of variations	Sum of squares	Mean square	Coefficient estimate	Standard error	*F*-value	*p*-value
			FOG removal (%)			
Model	870.84	96.76	–	–	24.43	<0.0001
Intercept	–	–	94.85	0.60	–	–
[pH]	216.32	216.32	–5.04	0.68	54.61	<0.0001
[Time]	157.21	157.21	4.30	0.68	39.69	<0.0001
[*I*]	107.87	107.87	3.56	0.68	27.23	0.0004
[pH][Time]	37.15	37.15	2.16	0.70	9.38	0.0120
[pH][*I*]	6.81	6.81	–0.92	0.70	1.72	0.2192
[Time][*I*]	50.20	50.20	–2.51	0.70	12.67	0.0052
$[pH]^2$	31.54	31.54	–13.00	4.61	7.96	0.0181
$[Time]^2$	19.95	19.95	10.34	4.607	5.04	0.0487
$[I]^2$	4.67	4.67	–5.00	4.61	1.18	0.3031
Residual	39.61	3.96	–	–	–	
Lack of fit	36.53	7.31	–	–	11.88	0.0084
Pure error	3.08	0.62	–	–	–	–
			Other statistical parameters			
	Standard Dev.	Mean	CV%	*R*-squared	Adj. *R*-squared	Adeq. precision
COD	2.16	55.95	3.87	0.9745	0.9461	16.214
TKN	3.38	67.26	5.02	0.6124	0.4334	9.632
TP	1.44	96.20	1.49	0.87	0.76	11.74
FOG	1.99	91.59	2.17	0.9565	0.9173	20.638

treatment [45]. In the first phase of present study, it was distinguished that addition of sodium chloride did not considerably improved the process performance, which could be explained by the already presence of required salts in real effluents.

The Pareto's chart (Fig. 3) represents the effect of each operational variable, isolated or combined, on COD, TKN, TP, and FOG removal by ECF process. The positive effect (higher than the significance level) of the variables time, current intensity, and pH shows that the use of their higher levels is necessary to attain a high efficiency in COD, TKN, TP, and FOG removal. On the other hand, the negative values suggest that low levels might be used to reach a high efficiency. In relation to the effects estimated for the combined variables, a negative value indicates the need to reverse the trend of one of the variables to get a combined positive effect [46,47]. Given the nature of contaminants, it is worth noting here that the fate of each substance, or group of substances, in ECF would be largely driven by its initial concentration as well as the physicochemical characteristics of the compounds under process, which in turn dictated largely by their size (soluble, colloidal, or suspended), molecular weight, aqueous solubility, the affinity of compounds to the aqueous phase (logKow and pKa values), among others. These processes are also vulnerable to operating parameters. However, direct evidence is rare in the available literature whether these intrinsic properties of chemicals are governing ECF [45,48].

Fig. 2 represents response surface plots for COD, TKN, TP, and FOG removal as a function of time, current intensity and pH. To examine the effect of initial pH, its value was adjusted to the desired level for each experiment by adding sodium hydroxide or hydrochloric acid aliquots. It is evident that the optimum pH for the removal of COD and TP was 9, while it was 10 for TKN and 8 for FOG removal. The process of pollutants removal through ECF process in view of pH does not corroborate previous works. This inconsistency is probably because of using synthetic wastewater in these studies, as well as ignoring properties of settled sludge and applying effluent. In summary, Table 5 presents wastewater by EC. In contrast with our study, also Bensadok et al. [37] reported that the efficiency of pollutants removal from synthetic dairy effluent using aluminum and titanium electrodes in pH 6.6 and sodium chloride concentration of 1.5 g/L was 80%, 59%, and 96% for turbidity, COD, and phosphate, respectively. Moreover, Phalakornkule et al. [49] using a combined process of coagulation–flotation in the pretreatment of a real wastewater from a factory producing date oil reported removal efficiencies as 72% for oil and 64% for COD at pH 5. Similarly, Sangal et al. [50] reported 99% of turbidity removal of the petroleum effluent at pH 6.5. Similar to experiments in our study, Sengil [4] employed steel electrodes to treat real dairy wastewater through ECF process at neutral pH. There observed 99% and 98% respective removal efficiencies for FOG and COD, which might be attributed to the formation of iron chloride complexes as absorption intermediator, resulting in increased efficiency.

When the initial pH tends toward acidic or basic values, the removal efficiency decreases; which is ascribed to the

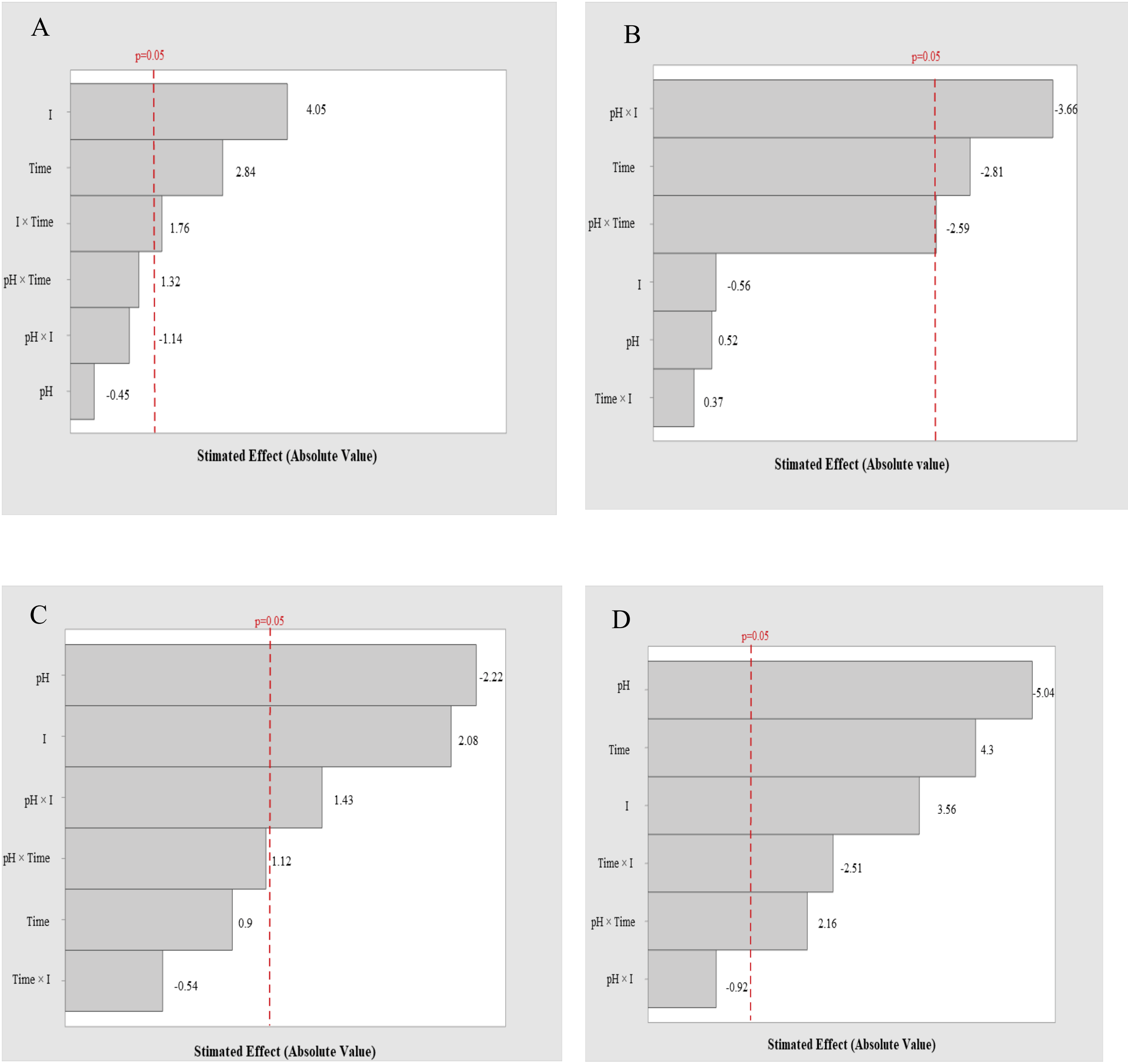

Fig. 3. Pareto chart for treatment of real dairy wastewater by electrocoagulation–flotation (ECF) process. Figure obtained according to the experimental data presented in Table 3. (A) COD removal (%), (B) TKN removal (%), (C) TP removal (%), and (D) FOG removal (%).

amphoteric character of aluminum hydroxide $Al(OH)_3$ that precipitates at pH 6–7 and its solubility increases when the solution becomes either more acidic or alkaline [51]. As a result, the flocs of aluminum hydroxide are less reactive and the flocculation is less effective, because of the formation of flocs of smaller size.

By the same taken, the optimum electrolysis time for COD and TP removal in our study was 60 min, while it was 45 min for TKN and 75 min for FOG removal (Fig. 2). According to the Faraday's law, the produced ions concentration increases by expanding electrolysis time which in turn results in coagulation through the formation of hydroxide flocs [52]. In the initial steps of reactions, especially in low current intensity, the amount of produced cations in anode as well as the amount of flocs and thence efficiency is low. On the other hand, with the increase of time, the concentration of hydroxide ions appears to grow and the chance of floc formation increases. Also, this was in line with a generally assumed pattern where an increase in time and current intensity is associated with enhanced secondary reactions such as indirect oxidation of organic compounds in the presence of chloride ions, which may further result in improved removal efficiencies [53]. Exceeding the optimal value, the removal efficiencies remain almost constant; this may be assigned to the saturation of ions released from the electrodes and the formation of new flocs. The findings observed in this study mirror those of the previous studies investigating the application of ECF

Table 5
Summary of pollutants removed by electrocoagulation in wastewater sources

Reference	Pollutants	pH	Time	Current or current density	Electrode materials, electrode connections	Phosphate removal (%)	COD removal (%)	Turbidity removal (%)	FOG removal (%)	Energy consumption
Bensadok et al. [37]	Dairy effluent	6.6	2 min	0.5 mA/cm^2	Al–Al	59	80	96	–	0.03 kWh/kg COD
Phalakornkule et al. [48]	Palm oil mill effluent	5	5 min	20 A/m^2	Al–Al monopolar	–	72	64	–	0.10 kWh/m^3
Sangal et al. [50]	Oil Wastewater	6.5	–	138.8 A/m^2	Al–Al	–	-	–	99	–
Mirji and Kalburgi [54]	Dairy wastewater	7	30 min	–	Al–Fe	–	88.54	–	–	–
Jagadal et al. [70]	Dairy wastewater	8	40 min	–	Al–Al monopolar	–	88	93.1	–	–
Yazdanbakhsh et al. [59]	Olive oil mill natural wastewater	5.2	60 min	117.187 A/m^2	Ti–Fe monopolar	–	96.14	99.89	–	–

in dairy wastewater treatment [15]. Mirji and Kalburgi [54] studied COD removal from real dairy wastewater in an electrocoagulation system (5–30 min) and produced treated waters with 30% to about 88% removal efficiencies. Also, Shivayoghimath and Meti [55] pointed out that the turbidity removal efficiency increased over time until it reached 92.2%.

The optimum current intensity for COD and TP removals was 3 A, while it was 1.5 A for TKN and 4.5 A for FOG removal. The release of Al^{+3} ions are dependent on current intensity and electrolysis time. The intensity of current, as an important operational parameter, determines the amount of electrochemical metal dosage in the water as well as the density of electrolyte bubbles. Consequently, a greater ascending flow and better removal of pollutants and sludge by flotation are obtained [56,57]. Shiny surfaces, such as stainless steel in our study, generate bubbles of appropriate size and flux and play a pivotal role in floating particles with various sizes, so that gas bubbles shrink by increasing current intensity [41]. It was ever reported that shrinking size of bubbles coupled with increased density of gas bubbles may cause efficiency improvements achieved in EF process [58]. It is clear that the higher current intensities the less required time for obtaining similar efficiencies, which could be assigned to increased process rate at higher current intensity. On the other hand, with the increase of electrolysis time, further improvements in the removal efficiencies of COD, TKN, and FOG were observed ($p < 0.05$), which is corresponding to increased reaction time. Moreover, it was evident that more hydrogen bubbles were generated by time at the cathode surface. It has been widely accepted that not only these bubbles enhance the degree of mixing but could also improve the flotation ability of the cell with a consequent increase in the percentage removal. The findings observed in this study mirror those of Emamjomeh and Sivakumar [45] who explored pollutants removal by EF and ECF processes. During the ECF experiments, very high turbidity removal, that is, 95.94%–99.91% was also achieved, while color removal was about of 94.61%–99.81%. In accordance with the present results, Yazdanbakhsh et al. [59] who examined the wastewater treatment of olive oil industry through ECF process, found that the efficiency for turbidity, COD and phenolic compounds were 99.89%, 96.14%, and 89.97%, respectively.

Under optimum conditions, the effluent pH increased by 1.15 units. The increase in effluent pH (even minor) might be resulted from either $Al(OH)_3$ formation, hydroxide sediments of other cations around the anode electrodes, or generation of hydroxide ions, as has been previously pointed out [60]. These results match those observed in several earlier studies in which the effluent pH of dairy industry wastewater increased by 1.5–2 units through EC process [61,62].

In addition to electrodes consumed, major operating cost of ECF is associated with the electrical energy consumption during the electrochemical process [63]. It was clear that when the time increased for a current intensity of 3 A, the energy consumption increased. After 45 min of treatment, all the dependent parameters were constant, but the energy consumption increased significantly. We can, therefore, consider that the electrolysis time of 45 min provides the optimum conditions for maximum removals of

COD, TKN, TP, and turbidity and FOG. Thus, the energy consumption increases with the current intensity and operating time. We can conclude that the optimum current intensity of 3 A leads to an optimal value of the energy consumed by the system with the best removal efficiencies pollutants. The value of the energy consumption for the optimal conditions (electrolysis time of 45 min and current intensity of 3 A) is about 0.069 kWh/kg COD.

3.3. Process optimization

Additional laboratory experiments were carried out to confirm the reliability of the model based on the optimum conditions suggested by the Design-Expert software as shown in Table 6. As can be seen, the model showed a satisfactory correlation between the experimental results and predicted values with regard to removal efficiencies. The optimum conditions were obtained at pH 8, electrolysis time 45 min, and current intensity 3 A. It implies that the role played by ECF in the treatment of dairy wastewater might be explained by a combination of direct coagulation of suspended organics/inorganics and indirect oxidation–reduction of dissolved chemicals by oxygen bubbles in the anode and hydrogen in the cathode. Nevertheless, it is evident that the ECF process may be used as primary treatment for the real dairy products wastewater suggestively in place of anaerobic treatment unit. Another process (which could be conventional biological treatment) has to be followed to deeply remove dissolved COD as well as N and P from effluent in order to meet discharge legislations. This viewpoint is in good agreement with BOD_5/COD ratio of ECF treated effluent (0.79), reinforcing that the effluent could be effectively treated biologically. The findings are well-suited with Roopashree and Lokesh [64] who employed ECF process for pre-treatment of textile industry wastewater with the special focus on BOD_5/COD ratio of effluent. Furthermore, the results are congruent with previous studies exploring the application of EF process in petroleum wastewater with appreciable efficiencies of up to 99% [50].

With regard to solids settling ability, the coagulation induced sludge is described as having good attribute considering its sludge volume index (SVI) value (86.63 mL/g) [64,65]. Such SVI value is normally associated with more rapid thickening and more efficient clarifier performance. Sedimentation velocity of sludge was also calculated according to Eq. (8).

$$V = 28.1\,(\mathrm{SVI})^{-0.2667} \tag{8}$$

where V denotes sedimentation velocity of sludge (m/h) [66]. Clearly, the ECF process with aluminum electrodes and stainless steel reactor body produces sludge with appreciable velocities reaching to 8 m/h.

Aluminum concentration in the treated effluent (14.157 mg/L) and sludge (3.1 mg/L) did not meet the effluent standards promulgated by U.S. EPA, declaring aluminum concentration in the effluent as 0.2–0.5 mg/L and also Indian Standards announcing it as less than 0.2 mg/L. Moreover, our results are in contrast to earlier findings reporting very low (about 0.001 mg/L) aluminum content in treated effluent [50]. The values are indeed beyond the national regulatory standards. To meet a national standard of 5 mg Al/L or less in the effluent, another process (which could be adsorption, ion exchange, etc.) should be followed to attenuate Al concentration in effluent in order to dispose into surface water or wet well, as well as irrigation and agricultural reuses [67]. It is also worth noting here that national criteria and legislations have not been specified for sludge quality in wastewater treatment plants at present. Wastewater treatment plants operators do not monitor the quality of the sludge [68]. Meanwhile, in authentic regulations of other nations, there was still lack of research on aluminum concentration in disposed sludge. However, a bunch of studies have investigated other metals such as chrome, copper, zinc, and lead [69]. The national department of environment is highly recommended to set minimum standards of sludge disposal.

Basically, SEEC was estimated at 0.069 kWh per kg of removed COD (0.069 kWh/kg COD). This result differ from previous estimations of SEEC, probably because of applying mono-polar electrode arrangements in their experiments [59], but is broadly consistent with Bazrafshan et al. [15] who worked on dairy industry wastewater treatment by ECF process. The diverging results between studies may be due to differences in reactor configuration, the employed electrodes, and perhaps most importantly, composition

Table 6
Verification of experimental results at optimum conditions (electrolysis time of 45 min, pH of 8, and current intensity of 3 A)

Optimum condition	Experimental	Predicted
COD removal efficiency (%)	66.44	63.28
Ratio of BOD_5/COD in effluent	0.79	NA
Aluminum concentration in effluent (mg/L)	14.157	NA
Aluminum concentrations in sludge (mg/L)	3.10	NA
Effluent turbidity (NTU)	4.61	4.35
SVI (ml/g)	86.6	149.4
Sedimentation velocity of sludge (m/h)	8.54	NA
SEEC (kWh/kg COD)	0.069	NA
Cost (cent/kg COD)	0.828	NA

NA: not available

of wastewater-especially with regard to synthetic or real effluent.

4. Conclusion

This study explores dairy wastewater treatment of a factory using aluminum (anode) and steel (cathode) electrode by ECF process in an electrochemical reactor with batch flow. In this study, approximate equations were achieved from second-order model. We attempted to examine the main and interactive effects of operational parameters such as pH, current intensity, and electrolysis time on the removal efficiencies of COD, TKN, TP, and FOG. Practical optimum condition limits were discovered to obtain the highest efficiency in pH 8, current intensity 3 A, electrolysis time 45 min, in which removal efficiencies for COD, TKN, TP, and FOG were 66.44%, 73%, 98%, and 97%, respectively. SVI of 86.63 mL/g was estimated as well. Finally, it could be concluded the novel ECF could be a cost-effective and promising process for real dairy wastewater treatment. To go further in our understanding of the influencing variables and upgrading of existent units, in depth optimization protocols should be investigated using high-throughput techniques.

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