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The treatment of printing and packaging wastewater by electrocoagulation–flotation: the simultaneous efficacy of critical parameters and economics

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ABSTRACT

In this work, electrocoagulation–flotation (ECF) following sedimentation was applied as a printing and packaging wastewater treatment using four Al electrodes with a parallel monopolar configuration. A sedimentation process was applied after the ECF as a post-treatment phase to remove large pollutants. The simultaneous efficacy of the operating parameters initial color content (1,843.44–12,156.56 ADMI), initial pH (3.56–10.44), current density (6.02–22.18 mA/cm^2), and treatment time (5.62–74.38 min) on color and chemical oxygen demand (COD) removal efficiencies were evaluated alongside processing costs. Response surface methodology (RSM) and central composite design (CCD) optimized these key parameters to achieve the highest removal efficiencies and lowest operating costs. Based on the results analyzed by RSM-CCD, using initial color content of 5,576.38 ADMI, initial pH of 7.29, the current density of 18.49 mA/cm^2, and treatment time of 59.76 min as optimum operational conditions can result in 97.8% and 92.1% for color and COD removal efficiencies, respectively. At these optimum conditions, operating costs (OPCs), including electrodes material and energy consumption, were 0.07 US$/(kg color removed) and 0.4 US$/(kg COD removed). The results confirm ECF-sedimentation as a promising and cost-effective tool for the treatment of printing and packaging wastewater.

Keywords: Printing and packaging wastewater; Electrocoagulation–flotation; Optimization; Operating cost

1. Introduction

In the present century, population growth and rapid urbanization have led to a drastic rise in industrialization and environmental pollution [1,2]. Printing and packaging industries have grown dramatically in the last decades due to the increased use of paper and cardboard in packaging and ink in printing. The printing and packaging industry is one of those that require a great deal of water to produce their products, consuming water in most parts of

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their production process [3,4]. Printing and packaging wastewaters (PPWWs) can contain resin acids, dyes and pigments, solvents, optional additives, phenols, organic and inorganic substances, heavy metals and other soluble materials [5–7]. Amongst the organic compounds, high values of chemical oxygen demand (COD) and color are also recorded in these wastewaters. The treatment of PPWW is challenging due to the aforementioned hazardous compounds. Prior to serious environmental issues, attention must be paid to the suitable treatment of this type of wastewater to find the best treatment system for pollutants to meet environmental standards.

Various methods including membrane filtration [8], adsorption [9], coagulation–flocculation [10], electrochemistry [11], and catalysts [12] have been used to treat industrial discharge over recent decades. Current treatment methods include biological [13], electrocatalytic [4], flocculation [14], fly-ash coated with chitosan [15], a combined process of coagulation and biosorption [5], membrane bioreactor [16], and a combination of Fenton and coagulation [17]. However, these techniques have limitations such as a high volume of sludge generation, lengthyong treatment time and high operating costs requirement, chemical reagent additives, and the production of secondary pollutants necessitating the need for. Accordingly, efficient and affordable technology for the treatment of this type of wastewater is highly expected. Among the various wastewater treatment methods, the electrocoagulation–flotation (ECF) process has been worked well as a conventional emerging technology based on complex mechanisms, to reduce different organic substances and organic dyes from wastewater due to its environmentally friendly and economically attractive merits [18–20]. According to the literature, this process has many potential benefits such as no production of secondary pollutants, no need for chemical additives, less production of sludge and lower energy consumption leading to reductions in operating costs [20–22]. ECF, like any other method, has some drawbacks, such as the need for high electrical conductivity water/wastewater, the deficiency of reactor design, and corrosion of electrodes because of the oxidation process [23–25]. ECF has been widely employed to treat various effluents. One study of urban wastewater treatment resulted in 84% and 80% of COD removal by Al and Fe electrodes, respectively [26]. A study by Adamovic et al. [27] in the treatment of waste offset printing developer wastewater, showed that in the first 5 min more than 90% of turbidity and copper was removed, the percentage of organic substances removed above 50%. Chawaloesphosiya et al. [28] removed more than 90% of turbidity from oily wastewater, this evidences that the ECF process produced less oil sludge than chemical coagulation. In another study, textile wastewater treatment was carried out by Ghanbari et al. [29], where under optimal operational conditions, 98% color and 87% COD were removed. The ECF process has also been used in the treatment of printing wastewater [30] carwash wastewater [31], laundry wastewater [32], electroplating wastewater [33] and restaurant wastewater [34]. The aforementioned studies, [30–32], show ECF processes to be very promising and cost-effective technology for the treatment of a wide range of pollutants, however, based on the literature review, the application of the ECF process followed by sedimentation has not yet been taken into consideration for the treatment of PPWW.

In this alternative technology, as soon as the electrical current starts through the submerged electrodes, the coagulants produce a sacrificial anode (iron or aluminum) by electro-solubilization [27,35]. Simultaneously, hydroxide (OH^-) ions and hydrogen (H_2) gas are produced at the cathode electrode surface [36,37]. The selection of suitable metallic electrodes depends on critical parameters such as material accessibility, cost-effectiveness, oxidation potential, and target pollutant properties [18,38]. The literature shows that aluminum is a very effective and efficient electrode at removing different pollutants under different operating conditions [23,39,40]. With ECF, when using aluminum (Al) as an electrode material by passing an electrical current through a solution and electrodes, the anode releases aqueous Al^{3+} ions (Eq. (1)), while the cathode generates H_2 gas and OH^- ions (Eq. (2)). These cations then form solid $Al(OH)_3$ precipitate (Eq. (3)). The reactions occurring in the ECF process are as follows [41,42]:

Reactions at the anode (for coagulation):

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^- \quad (1)$$

Reactions at the cathode (for flotation):

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^- \quad (2)$$

Overall reactions at bulk (for precipitation):

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3 + 3H^+ \quad (3)$$

The simultaneous effects of important parameters facilitate better removal efficiency of pollutants. As recent evidence shows, response surface methodology (RSM), a powerful mathematical technique, has overwhelmed the limitations of classical experimental methods including time-consuming and high energy requirements that result in a waste of financial resources [43–45]. RSM optimizes numerical parameters such as current density, pH value and treatment time, not qualitative parameters such as electrode material or application mode. This technique has been used to treat several industries using ECF such as biodiesel wastewater [46], waste offset printing developer [27], restaurant wastewater [47], and waste fountain solution [48]. In summary, the researchers acknowledge the fact that RSM as a statistical tool can be used to analyze the impact of several independent factors on the treatment process to maximize the benefit from the process. Because water or wastewater treatment processes are influenced by several variables, the effects of these parameters must be optimized to attain the best performance of the treatment system. This technique can be widely, efficiently and flexibly adapted for the parameter optimization of various wastewater treatment processes.

The economic implications of the ECF process have been investigated in several recent studies where operating costs, materials and energy costs have been taken into account over other parameters. Elazzouzi et al. [26] applied ECF for urban wastewater treatment using Al and Fe electrodes. The results revealed that COD removal using the Al electrode was as low cost as using Fe electrodes. Ozyonar and

Karagozoglu [49] studied the treatment of domestic wastewater by ECF using aluminum electrodes. The operating cost of 0.86 \$/m^3 under the optimum operating conditions was evaluated. Koyuncu and Arıman [50] treated domestic wastewater including suspended solids (SS), biological oxygen demand (BOD), COD, total nitrogen, total phosphorus, using the ECF process in real-scale electrocoagulation (EC) plant, and determined the operating cost of the process as 0.24–0.28 EUR/m^3. As such, the economic perspective is a crucial factor in the ECF process.

To the best of our knowledge, from the literature [27,30,51,52], many variables have been investigated when evaluating the performance of the ECF process used to remove various pollutants. The commonly studied operating parameters, in the ECF, are the solution pH, treatment time and current density. This work is dedicated to the investigation of color and COD removal from PPWW using the ECF-sedimentation process. RSM and central composite design (CCD) were used as the optimization techniques for this process to explore the following key variables: pH, treatment time, current density and the initial content of color. The operating cost was estimated according to the consumed amounts of materials and energy.

2. Experimental setup

2.1. Printing and packaging wastewater

The PPWW used in this work was obtained from a printing and packaging company in Qazvin, Iran. The PPWW samples were taken from the main effluent outlet and stored in a dark, cool (4°C) place before use. Sulphuric acid (H_2SO_4) 0.1 N and sodium hydroxide (NaOH) 0.1 N were used as pH regulators for the initial PPWW pH. The PPWW comprised COD = 235–1,400 mg/L, color = 1,845–12,156 ADMI, pH = 3.56–10.44 and electrical conductivity = 749–920 μS/cm.

2.2. Analytical procedure

The PPWW was analyzed for COD, color, pH and electrical conductivity. COD was determined using a digestion reactor (LT200, Hach, USA) and direct reading spectrophotometer (DR 6000 UV-Vis, Hach, Germany) at 620 nm according to standard test methods: 5220 D (closed reflux colorimetric) [53]. The pH of the PPWW was recorded by a multi-parameter analyzer (CONSORT C831, Belgium) and corrected before each test. The electrical conductivity of samples was measured with a digital calibrated conductometer (Leybold 666222, Germany) and reported on without any adjustments. The color was measured with a spectrophotometric (DR 6000 UV-Vis, Hach, Germany) as per the ADMI method: APHA 2120 F [53]. In each case, the color and COD removal efficiency (R_e) and the operating cost (OPC) US\$/(kg pollutant removed) are calculated as [54–56]:

$$R_e(\%) = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (4)$$

$$\text{OPC}\left(\text{USD/m}^3\right) = \left[a \times Q_{\text{electrode}}\right] + \left[b \times Q_{\text{energy}}\right] \quad (5)$$

$$Q_{\text{Energy}} = \frac{U \times I \times t}{(C_0 - C_t) \times V_R} \quad (6)$$

$$Q_{\text{Electrode}} = \frac{I \times t \times M \cdot V}{Z \times F \times (C_0 - C_t) \times V_R} \quad (7)$$

where C_0 and C_t are the initial and final pollutant contents (mg/L), respectively; U is the operational electrical potential (volt); I is the applied current (ampere); t is the treatment time (h) and V_R is the PPWW volume (L). The electrical energy consumption (EEC) is specified in terms of electric consumption per 1 mg of color and COD removal. In order to define the economic feasibility of the process, operating costs (OPCs) were calculated. Q_{Energy} (kWh/kg pollutant removed) and $Q_{\text{Electrode}}$ (kg Al/kg pollutant removed) refer to the electricity required for COD and color removal and consumption quantities of the electrode material, respectively. a and b are the 0.08 US\$ per 1 kWh electricity as priced by the Iranian Ministry of Energy and 1.95 US\$ per 1 kg aluminum as priced by the Iranian market, respectively. M.V is the molecular mass of aluminum (26.98 g/mol); Z is the number of electrons transferred (Z = 3) while F is Faraday's constant (96.487 C/mol).

2.3. Set-up of the monopolar batch ECF-sedimentation treatment

The experimental apparatus is shown in Fig. 1. The ECF reactor consisted of four aluminum electrodes connected to the direct current power supply (model JPS303D, Iran) to provide an adjustable voltage (0–30 V) and applied current (0–3 A) using a monopolar configuration. The dimension of the electrode was 30 cm × 10 cm × 0.5 cm, the effective area of all the anodes on both sides 480 cm^2. The ECF cell was subjected to the magnetic stirrer (model SHA R-50, Iran) at a constant stirring speed to provide thorough mixing of the solution throughout. In each run, a net volume of 3.3 L of PPWW was placed into the ECF reactor. After ECF treatment, a sample of 20 mL was removed with a syringe, from the center of the reactor at specific time intervals, and transferred to the settling tank for 20 min. These samples were used to analyze the residual COD and color. At the end of each run to avoid passivation, the electrodes were scrubbed thoroughly with sandpaper and washed with water to remove any solid residues on the surfaces.

2.4. Response surface methodology

RSM and CCD are powerful statistical techniques for modeling and optimizing the simultaneous effects of critical factors within a four-factor, five-level framework. The range of experimental variables were determined based on pretest results and the values given in the literature [51,57–59]. In the current work, the modeling and optimization of critical parameters, namely initial pH (3.56–10.44), treatment time (5.62–74.38 min), current density (6.02–22.18 mA/Cm^2), and initial color content (1,843.44–12,156.56 ADMI) as independent variables, vs. COD and color removal efficiencies as responses, were conducted through CCD under RSM via State-Ease Design-Expert® Software, version 7.0. Thirty trials were designed by employing the parameters within the

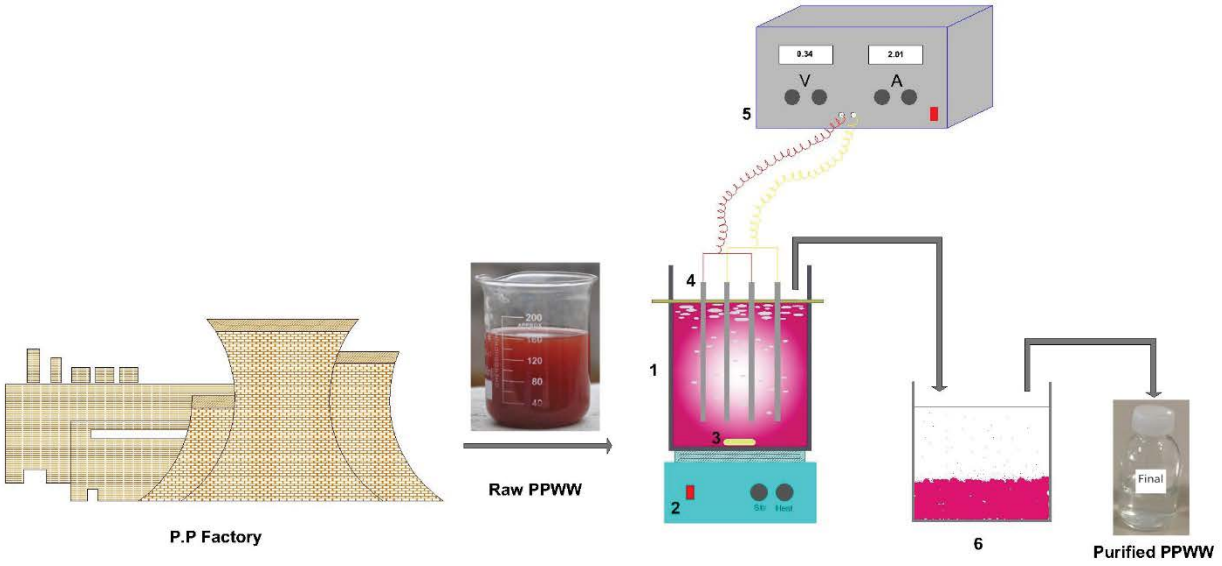

Fig. 1. ECF-sedimentation setup of real printing and packaging wastewater ((1) ECF cell; (2) magnetic stirrer; (3). magnetic bar; (4). aluminum electrodes; (5). DC power supply; (6). sedimentation tank).

framework of four variables and five levels. A second-order polynomial regression represented the empirical model as seen by Eq. (8) which was used to investigate the interaction between y and the independent variables:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{l \le i \le j}^{k} \beta_{ij} x_i x_j + \varepsilon \quad (8)$$

where y is the response (COD and color removal efficiencies, %); i is the linear constant; j is the second-order; β_0, β_i, β_{ii} and β_{ij} are the constant coefficient, the regression constant, the quadratic coefficient, and the interaction coefficient, respectively; and x_i and x_j are the coded independent variables. As shown in Table 1, the critical factors, namely initial pH, treatment time (min), current density (mA/cm^2), and initial color content (ADMI) are coded at five levels, these are evaluated with a CCD.

3. Results and discussion

3.1. Modeling and data analysis

The CCD and RSM analysis were used to determine the individual effects of ECF operational parameters (initial pH, treatment time, current density and initial color content) and their interactions with response variables COD and color removal efficiencies. A total of 30 trials, together with the actual data and model-predicted data, are shown in Table 2. Color removal varied between about 84% and 100% while the COD removal varied between 50% and 95%. The predicted values were fitted with the second-order polynomial functions, where Y_1 and Y_2 are color and COD removal (%), respectively, while A, B, C and D are the values of initial color content, initial pH, current density and treatment time, respectively (Eqs. (9) and (10)).

$$Y_1 \text{ (Color removal, \%)} = 96.77 - 2.98A + 2.10B + 2.61C + 2.72D - 1.22AB + 1.56AC + 1.96AD - 1.25BC + 0.98BD + 1.19CD - 0.96A^2 - 1.85B^2 - 1.64D^2 \quad (9)$$

$$Y_2 \text{ (COD removal, \%)} = 83.88 - 11.28A + 3.88B + 3.47C + 2.60D - 3.37AB + 3.25AC - 3.00BD + 8.13CD - 3.25A^2 - 6.64B^2 - 2.57C^2 - 2.57D^2 \quad (10)$$

In order to verify the adequacy of the model, an analysis of variance (ANOVA) was used as seen in Tables 3 and 4. Table 3 lists the results for color removal which

Table 1
Coded variables for the experimental design

Variables	Levels				
	$-\alpha$	–1	0	+1	$+\alpha$
Initial color content (ADMI)	1,843.44	4,000.00	7,000.00	10,000.00	12,156.56
Initial pH	3.56	5.00	7.00	9.00	10.44
Current density (mA/Cm^2)	6.02	9.40	14.10	18.80	22.18
Treatment time (min)	5.62	20.00	40.00	60.00	74.38

Table 2
Experimental design matrix of factors and their actual and predicted results

Run	Initial color content (ADMI)	Initial pH	Current density (mA/cm^2)	Treatment time (min)	Color removal (%)		COD removal (%)	
					Actual	Predicted	Actual	Predicted
1	7,000.00	7.00	14.10	40.00	99.85	96.77	79	83.88
2	10,000.00	9.00	9.40	60.00	91	91.09	40	42.57
3	7,000.00	7.00	14.10	40.00	96.85	96.77	85	83.88
4	4,000.00	5.00	9.40	60.00	88	87.59	72	70.37
5	7,000.00	7.00	14.10	40.00	98.85	96.77	86	83.88
6	10,000.00	5.00	18.80	60.00	98.5	98.10	77	77.26
7	10,000.00	9.00	18.80	20.00	84	85.60	53	57.32
8	4,000.00	5.00	9.40	20.00	91	90.39	81	74.67
9	7,000.00	10.44	14.10	40.00	94.88	94.92	75	70.94
10	7,000.00	7.00	6.02	40.00	90	90.24	64	70.30
11	4,000.00	5.00	18.80	20.00	91.5	92.60	59	59.12
12	7,000.00	7.00	14.10	5.62	86.9	87.23	69	71.81
13	10,000.00	9.00	18.80	60.00	99.5	99.30	71	72.02
14	7,000.00	7.00	14.10	40.00	95.85	96.77	83	83.88
15	4,000.00	9.00	9.40	60.00	98.8	98.69	80	79.12
16	10,000.00	5.00	9.40	20.00	79.5	79.84	51	53.12
17	7,000.00	7.00	14.10	40.00	94.85	96.77	90	83.88
18	4,000.00	9.00	18.80	20.00	96.5	94.80	79	79.37
19	7,000.00	7.00	14.10	74.38	97.43	96.60	80	80.74
20	10,000.00	5.00	9.40	60.00	84	84.89	53	47.31
21	7,000.00	7.00	14.10	40.00	93.85	96.77	84	83.88
22	10,000.00	5.00	18.80	20.00	89	88.30	55	50.57
23	7,000.00	3.56	14.10	40.00	88.25	87.70	50	57.61
24	7,000.00	7.00	22.18	40.00	99.95	99.20	85	82.25
25	10,000.00	9.00	9.40	20.00	84	82.14	66	60.38
26	4,000.00	9.00	9.40	20.00	96	97.59	93	95.43
27	12,156.56	7.00	14.10	40.00	88.5	88.83	53	54.89
28	4,000.00	9.00	18.80	60.00	99.8	100.65	95	95.57
29	1,843.44	7.00	14.10	40.00	99.9	99.06	92	93.66
30	4,000.00	5.00	18.80	60.00	93.5	94.55	87	87.31

are statistically significant ($p < 0.0001$) for the terms of the model. In this context, terms *A*, *B*, *C*, and *D* were highly significant. The remaining terms of the model were significant except for C^2. Regarding Table 4, a *p*-value of less than 0.0001 indicates that the terms of the model for COD removal are statistically significant. Here, the terms *A*, *CD* and B^2 were highly significant. Factors *B*, *C*, *D*, *AB*, *AC*, *BD*, A^2, C^2, and D^2 also achieved significance while the remaining terms of the model were not significant. According to the, R^2 and R^2_{adj} were 95% and 90% for color removal, while these values were 94% and 90%, respectively for COD removal. These are relatively high values, the correlation coefficients confirming the quadratic equation. Relatively high R^2 values indicated the accuracy of the quadratic equation for initial color content, initial pH, current density and treatment time.

As indicated in Figs. 2a and b, the diagnostic plot of actual vs. predicted values are in good agreement because the points follow an almost straight line, which confirms the suitability of the model for both color and COD removal efficiencies.

3.2. Pareto chart analysis

A Pareto chart is a powerful tool illustrating the level of significance of the effects of variables on responses obtained by the ratio SS_{term} to SS_{Total}. Fig. 3 illustrates the following trend: initial color content > initial pH > current density > treatment time and initial color content > treatment time > current density > initial pH for COD and color removal efficiencies, respectively. Initial color content had the highest effect in both cases of COD and color removal efficiencies with a contribution of 44.45% and 21.07%, respectively. Treatment time and initial pH had the lowest effect on COD and color removal efficiencies, respectively.

3.3. Simultaneous efficacy of critical parameters on the removal of pollutants

The simultaneous efficacy of critical parameters, namely initial color content, initial pH, current density, and treatment time over color and COD removal efficiencies were evaluated, optimized by mathematical models RSM and CCD.

Table 3
ANOVA results of experiment data for color removal

Source of variations	Sum of squares	Degree of freedom	Mean square	*F*-value	Prob. > *F*	Remarks
Model	920.66	14	65.76	21.42	<0.0001	Highly significant
A-Color	194.00	1	194.00	63.20	<0.0001	Highly significant
B-pH	96.56	1	96.56	31.46	<0.0001	Highly significant
C-Current density	148.83	1	148.83	48.49	<0.0001	Highly significant
D-Time	162.68	1	162.68	53.00	<0.0001	Highly significant
AB	24.01	1	24.01	7.82	0.0135	Significant
AC	39.06	1	39.06	12.73	0.0028	Significant
AD	61.62	1	61.62	20.08	0.0004	Significant
BC	25.00	1	25.00	8.14	0.0121	Significant
BD	15.21	1	15.21	4.96	0.0418	Significant
CD	22.56	1	22.56	7.35	0.0161	Significant
A^2	15.94	1	15.94	5.19	0.0377	Significant
B^2	59.58	1	59.58	19.41	0.0005	Significant
C^2	8.39	1	8.39	2.73	0.1191	
D^2	47.20	1	47.20	15.38	0.0014	Significant
Residual	46.04	15	3.07			
Lack of fit	19.21	10	1.92	0.36	0.9216	
Pure error	26.83	5	5.37			
Cor. total	966.70	29				
R^2/R^2_{adj} (%) = 0.95/0.90						

Table 4
ANOVA results of experimental data for COD removal

Source of variations	Sum of squares	Degree of freedom	Mean square	*F*-value	Prob. > *F*	Remarks
Model	6,265.96	14	447.57	18.21	<0.0001	Highly significant
A-Color	2,785.46	1	2,785.46	113.31	<0.0001	Highly significant
B-pH	329.55	1	329.55	13.41	0.0023	Significant
C-Current density	264.30	1	264.30	10.75	0.0051	Significant
D-Time	147.81	1	147.81	6.01	0.0269	Significant
AB	182.25	1	182.25	7.41	0.0157	Significant
AC	169.00	1	169.00	6.87	0.0192	Significant
AD	2.25	1	2.25	0.092	0.7664	
BC	0.25	1	0.25	0.010	0.9210	
BD	144.00	1	144.00	5.86	0.0287	Significant
CD	1,056.25	1	1,056.25	42.97	<0.0001	Highly significant
A^2	184.57	1	184.57	7.51	0.0152	Significant
B^2	768.83	1	768.83	31.27	<0.0001	Highly significant
C^2	115.72	1	115.72	4.71	0.0465	Significant
D^2	115.72	1	115.72	4.71	0.0465	Significant
Residual	368.74	15	24.58			
Lack of fit	303.24	10	30.32	2.31	0.1834	
Pure error	65.50	5	13.10			
Cor. total	6,634.70	29				
R^2/R^2_{adj} (%) = 0.94/0.89						

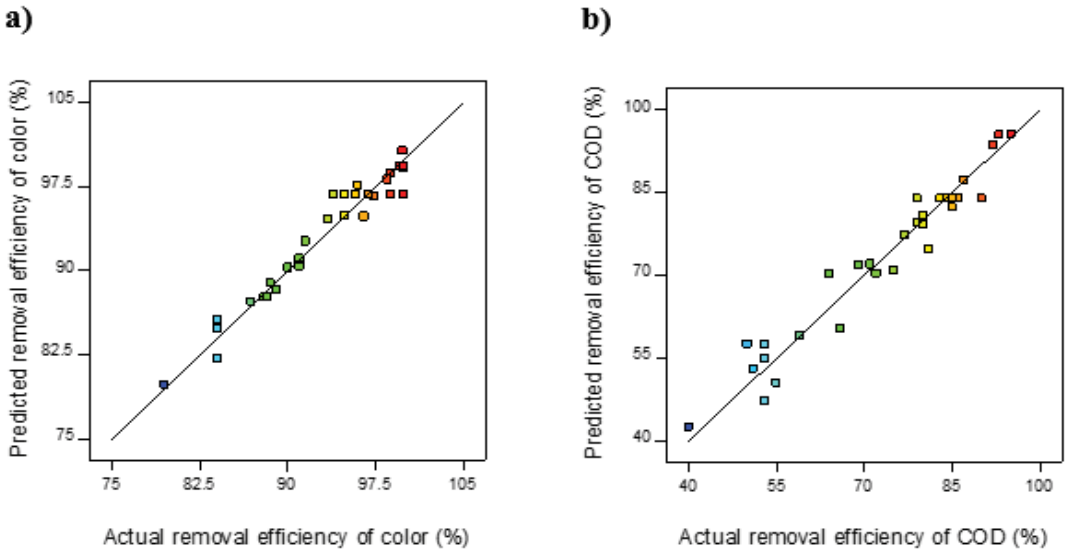

Fig. 2. Actual vs. predicted plots in both cases for (a) color removal efficiency and (b) COD removal efficiency.

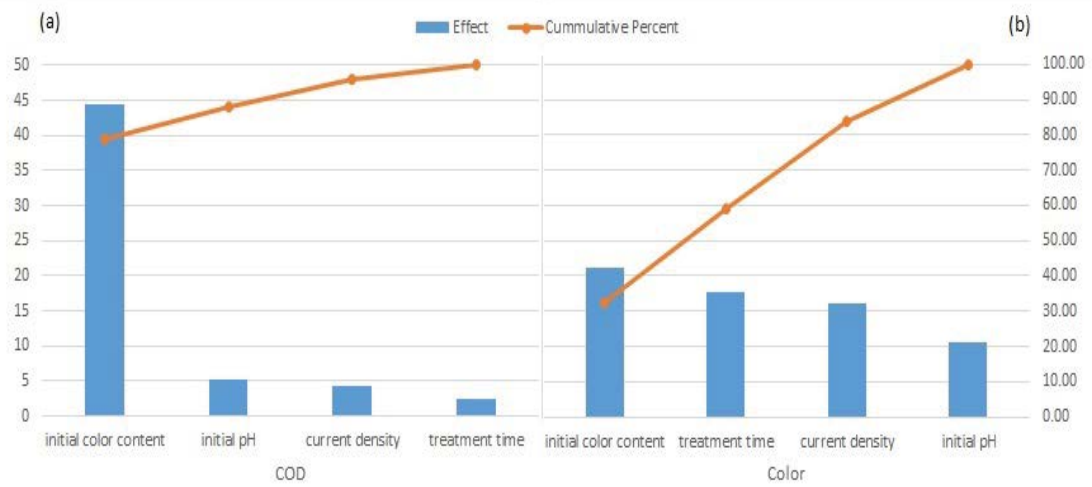

Fig. 3. Pareto effects analysis for (a) COD and (b) color removal efficiencies.

The effect of initial pollutant content on removal efficiency is a function of the type of pollutant [60]. Figs. 4a and b show the simultaneous efficacy of initial color content and initial pH for color and COD removal efficiencies. As can be seen, with an initial color content of 1,843.44 ADMI, treatment time of 40 min and current density of 14.10 mA/cm^2 in addition to an increase in pH up to 7–8, removal efficiencies are enhanced to 97.5%. The rate of increase in the corresponding removal of COD is negligible. This may be due to the low production of Al^{3+} at acidic pH. When the initial color content is increased at a high level of initial pH, removal efficiencies of both pollutants are decreased. At higher initial pollutant contents, the amount of aluminum hydroxide flocs produced is insufficient to coagulate sizeable quantities of color molecules [61]. This result is in line with Bazrafshan and Mahvi's [62], research who found at optimum operating conditions of pH 7, voltage 50 V, reaction time 60 min, initial concentration 50 mg/L, and conductivity 3,000 μS/cm, the removal efficiency of basic red 18 dye using aluminum electrodes was equal to 97.7%. In another study, Nandi and Patel [63] achieved 99% brilliant green dye removal at a neutral pH range of 7–8 by electrocoagulation process.

The solution pH is a key parameter in the removal of pollutants, significantly altering the physicochemical properties of the electrocoagulation process [64]. In the current work, the assays were conducted between 3.56 and 10.44 to evaluate the simultaneous efficacy of initial pH and treatment time for color and COD removal efficiencies (Figs. 5a and b). At the highest and lowest limits of treatment time, initial color content of 7,000 ADMI and current density of 14.10 mA/cm^2, color removal increased when the initial pH increased. This can be attributed to the reaction between Al^{3+} and OH^- in the solution [65]. At a pH of 7 and a treatment time of 40 min (at constant value current density of 14.10 mA/cm^2 and initial color content of 7,000 ADMI), the highest removal efficiencies of color

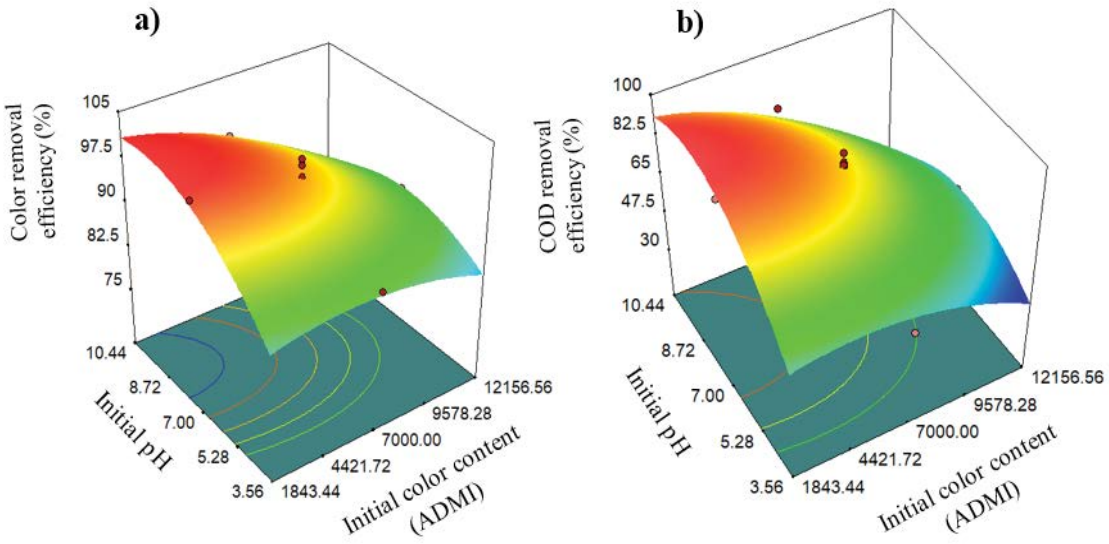

Fig. 4. 3D response surface plots for: (a) color and (b) COD removal efficiencies as a function of initial color content and initial pH (current density = 14.10 mA/cm^2 and treatment time = 40 min).

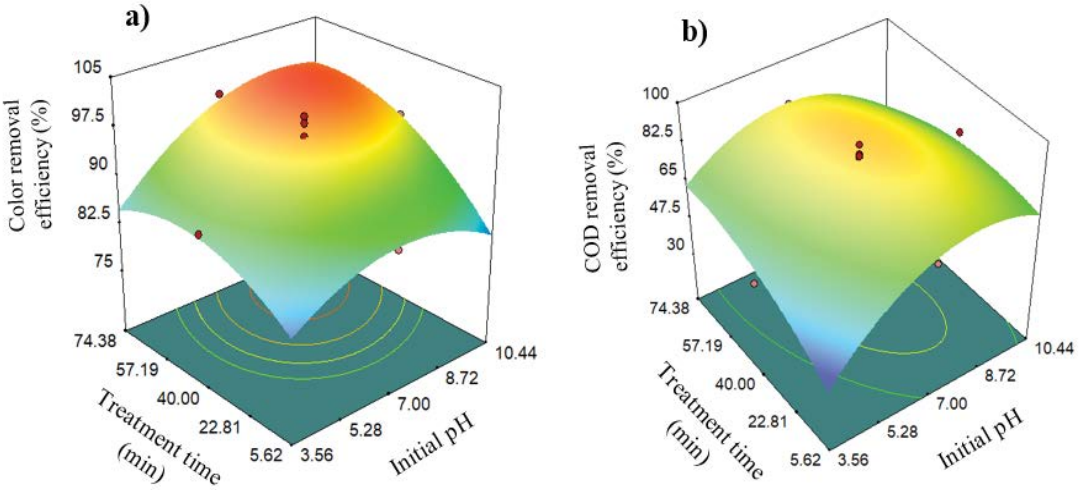

Fig. 5. 3D response surface plots for: (a) color and (b) COD removal efficiencies as a function of initial pH and treatment time (current density = 14.10 mA/cm^2 and initial color content = 7,000 ADMI).

and COD were obtained, 99.85% and 90%, respectively. As previously noted by other authors, at a neutral pH level, the maximum removal is observed [66,67]. This outcome is in contrast with the data reported by Aygun et al. [57] and Bayramoglu et al. [68], who observed that at an optimum pH of 5, the removal efficiencies of color and COD were increased. The reason for this contradiction can be attributed to the aluminum ions that appear in different phases and forms, depending on the pH and the chemical features of the solution. This finds that in the pH range of 5–6, aluminum may be in the forms of $Al(OH)^{2+}$, while, in the pH neutral range (6–8), aluminum changes to the form of $Al(OH)_3$, which means more pollutants removal and stability [69,70].

As we know from the literature [32,71], treatment time and current density are critical characteristics regarding the efficiency of pollutants removal from wastewater by ECF. According to Figs. 6a and b where the simultaneous efficacy of treatment time and current density on the removal efficiencies of color and COD from the PPWW are shown, at low initial treatment time with increasing current density, color removal efficiency are increased. COD removal efficiency is slightly increased, thereafter decreasing rapidly. When treatment time is increased, conditions are different. With the increase in current density (over 6 mA/cm^2) at longer treatment time (over 40 min), COD removal dramatically increased (64%–90%). Under these same conditions, color removal efficiency reached its highest rate (99.95%). When treatment time and current density increase, there are also higher production rates of metal ions and their hydroxide flocs as well as gas bubbles. Consequently, the concentration of pollutants decreases

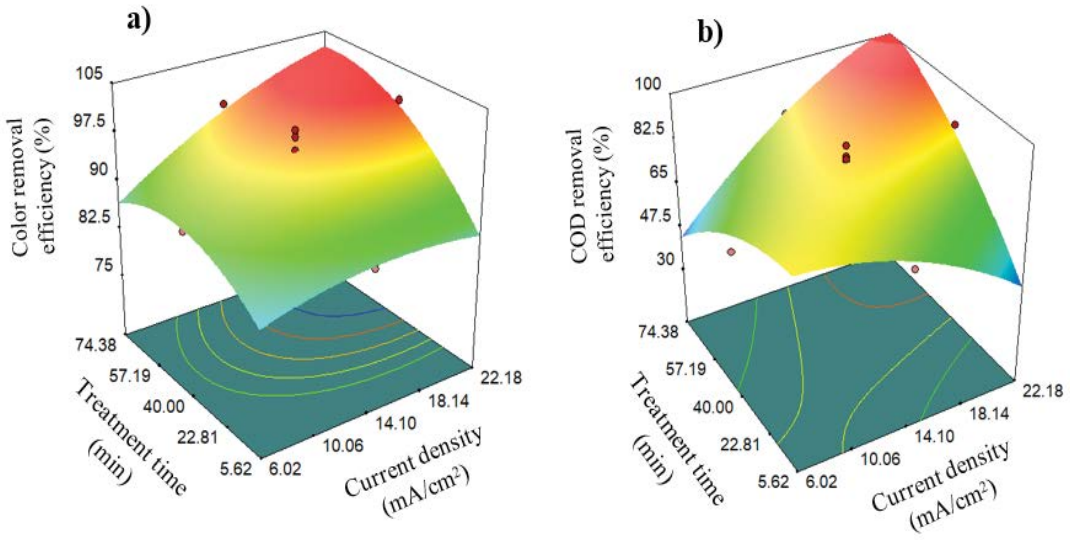

Fig. 6. 3D response surface plots for: (a) color and (b) COD removal efficiencies as a function of current density and treatment time (initial pH = 7 and initial color content = 7,000 ADMI).

faster [20,72–74]. The optimal treatment time and current density needed for efficient removal can be considered as 14.10 mA/cm^2 and 40 min, respectively.

3.4. Optimization step and cost estimation

After data analysis, optimization, which is one of the main objectives of RSM, was performed to achieve the optimum values of variables for maximum removal efficiency from the model. In the optimization step, the desired goal for color and COD removal efficiencies was selected as a maximum value of 100.68% and 95.99%, respectively, and the variables of initial color content, initial pH, current density, and treatment time were selected to be within range. Based on the results obtained by the model, the optimal operating conditions are as follows: an initial color content of 5,576.38 ADMI, initial pH of 7.29, a current density of 18.49 mA/cm^2 and treatment time 59.76 min. According to Fig. 7, the overlay plot for the optimal region, (the yellow portion, gave the allowable values of the two variables by maximizing color and COD removal efficiencies. As can be seen in Fig. 7, when the process was conducted at a current density of 18.6 mA/cm^2 and treatment time of 59.68 min, initial pH and initial color content should be 8.04 and 4821.65 ADMI, respectively, to maximize color and COD removals. Further assays under optimal conditions showed that color (97.8%) and COD (92.1%) removal efficiencies were close to the predicted values, which confirms the accuracy and validity of the optimization under optimal conditions.

Optimization has been studied using RSM and CCD for various wastewater treatments. For instance, Abbasi et al. [75] investigated a novel continuous EC reactor by RSM and CCD for the removal of 90.1% color, 89.4% COD, 82% turbidity and 73.3% alkalinity from a real licorice processing wastewater system under optimal conditions of electrolysis time (81.8 min), current density (350 A/m^2), NaCl

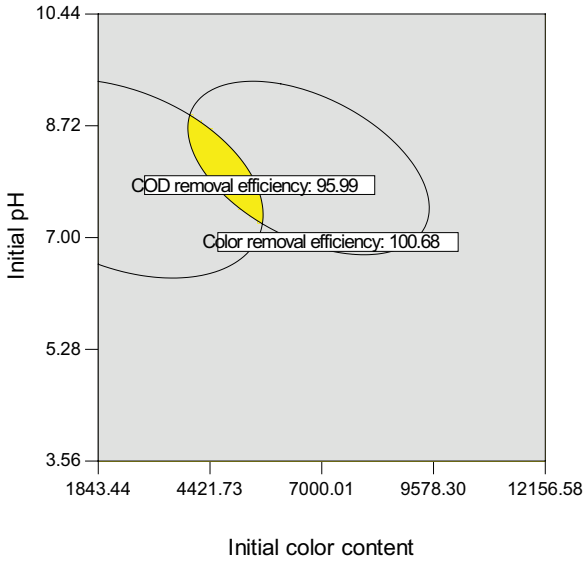

Fig. 7. The overlay plot for optimal region at current density = 18.6 mA/cm^2 and treatment time = 59.68 min.

concentration (300 mg/L) and mixing intensity (45 rpm). The study of Adamovic et al. [27] reported ECF efficiency for the removal of ~90% copper, ~90% turbidity and ~50% organic substances from printing developer waste using RSM. Chawaloesphosiya et al. [28] suggested combining ECF and physical processes for the removal of 96.7% linear alkylbenzene sulfonate and 87.65% phenol from automotive service wastewater using RSM-CCD to optimize the process parameters of acceptable results. Furthermore, Sankar and Sivasubramanian [67] studied the removal of 89.344% Congo red dye from an aqueous solution using optimization EC parameters namely 65 mg/L of the initial

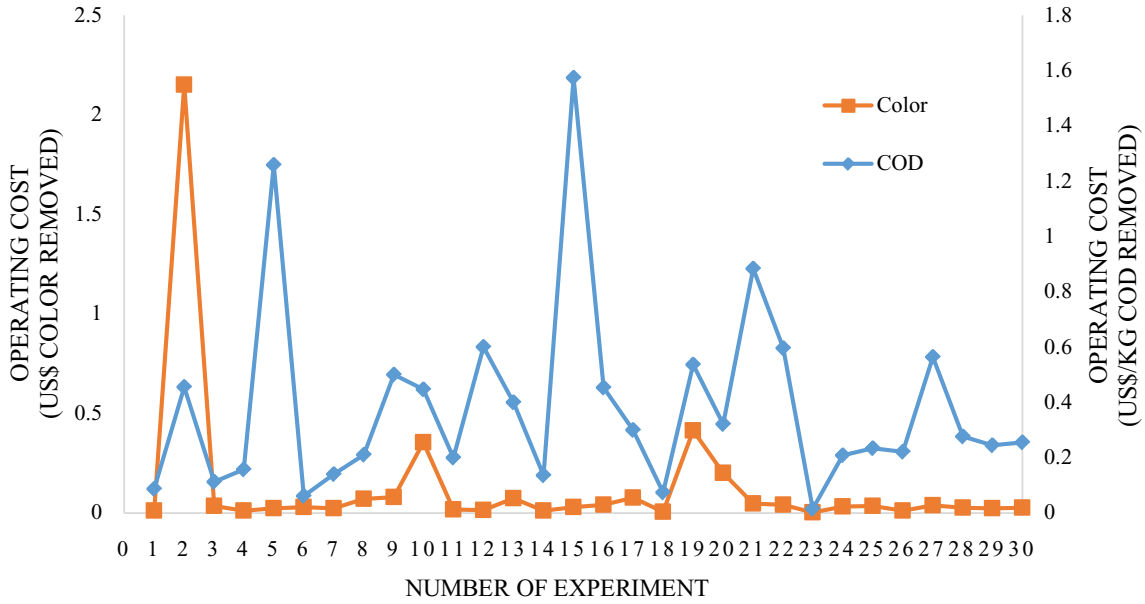

Fig. 8. The operating costs per color and COD were removed through 30 experiments designed by RSM.

concentration, 9.5 of pH, 8.75 V of voltage and reaction time of 25.08 min with the RSM technique. According to the literature, RSM is a convenient, reliable and valid technique used to optimize the key parameters for the ECF process and is significantly superior to classic statistical methods.

The operating costs (OPCs) of the process were described in all experiments designed by RSM in Fig. 8, showing that COD removal OPCs are higher than color removal OPCs. The average OPC of all experiments for color and COD removal is 0.1 US$/(kg color removed) and 0.3 US$/(kg COD removed), respectively. OPCs under the optimum conditions mentioned above, were 0.07 US$/(kg color removed) and 0.4 US$/(kg COD removed). These results differ from the results of studies [76–78] suggesting that the use of ECF as a process for real printing and packaging wastewater treatment by RSM optimization, could be a feasible technology due to high COD and color removal efficiency and economical viability in terms of operating costs.

A series of recent studies conducted on real wastewater treatment by electrochemical and other processes are presented in Table 5. In contrast to the current study, Niazmand et al. [20] reported that the efficiency of pollutant removal from olive debittering wastewater, using EC followed by filtration under optimized conditions of pH 4, electrolysis time 30 min, current density 7.5 mA/cm^2 and at an electrode distance 1 cm, was 78.51%, 90.44% and 97.92% for COD, total polyphenolic compounds and turbidity, respectively. Yavuz and Ögütveren [79] who applied Fe-EC in the treatment of industrial estate wastewater, reported removal efficiency of 92% for COD at a current density of 30 mA/cm^2, supporting electrolyte of 3 mM Na_2SO_4, pH of 6, and H_2O_2 concentration of 1,500 mg/L. A further study by Gönder et al. [80] treated carwash wastewater by EC, reporting removal efficiencies as 84% COD, 99.3% anionic surfactant, and 82% oil–grease. Operating costs were 9.67 $/m^3 at optimum conditions of pH 4, current density 30 A/m^2 and stirring speed 250 rpm. Mahesh et al. [81] obtained 80% COD and 90% color removal efficiencies using EC for real carwash wastewater treatment. In other research, ~97% color, ~98% turbidity, ~64% COD, ~65% total Kjeldahl nitrogen removals and the cost of treatment as 1.08 US$ m^{-3} were obtained using a combination of electrocoagulation and tannin-based coagulant for the slaughterhouse and packing wastewater treatment by Orssatto et al. [82]. Asselin et al. [83] employed ECF to remove organic compounds from slaughterhouse wastewater and found that 86%, 99%, 50% and 82% respective removal efficiencies for BOD, oil-grease, soluble COD, total COD, at an operating cost of 0.71 USD $/m^3, this including energy and electrode consumption, chemicals and sludge disposal. A previous study achieved 60% and 98% for the removals of COD and turbidity, respectively, and 0.48–5.42 US$/m^3 for operating cost [84] with potato chips manufacturing wastewater. Aygun et al. [57] treating reactive dyebath wastewater by EC, reported 85.8% color and 76.9% COD removal with 1.84 €/m^3 operating costs for Al electrode under pH 5.01, electrolysis time 64 min and current density 28.5 A/m^2. In comparison with other studies, the current results demonstrated that substantial removal of COD and color with low operating costs were also achievable through ECF-sedimentation under optimum conditions. The diverging results between the operating costs of this study and those of other studies may be due to differences in the configuration of the ECF reactor, the purity of the electrodes used, and perhaps the composition of the real wastewater. Because of higher electrical conductivity, the voltage required by the DC power supply will be lower, facilitating lower operating costs.

It is therefore concluded that this process is preferable to other, similar treatment processes. To go further in our understanding of the influence of variables, other operating parameters including type of electrode and distance between electrodes against pollutants treatment such as heavy metals, phenols, organic and inorganic substances and sludge amount in PPWW should be investigated, using ECF-sedimentation and combination electrodes of iron, aluminum and steel.

Table 5
Comparison of different study results

Real wastewater	Pollutant	Technique	Removal (%)	Operating cost	References
Olive processing wastewater	COD	EC-filtration	78	0.12 US$/m^3	Niazmand et al. [20]
	Total polyphenolic compounds		90.44		
	Turbidity		97.92		
Industrial estate wastewater	COD	EC	91.67	3.41 €/m^3	Yavuz and Ögütveren [79]
Carwash wastewater	COD	EC	83	9.67 US$/m^3	Emamjomeh et al. [31]
	Oil-grease		99.3		
	Methylene blue active substances		82		
Pulp and paper mill wastewater	COD	EC	80	–	Mahesh et al. [81]
	Color		90		
Slaughterhouse and packing plant wastewater	Color	Combined EC/ organic coagulation	<97	1.08 US$/m^3	Orssatto et al. [82]
	Turbidity		<98		
	COD		<64		
	Total Kjeldahl nitrogen		<65		
Slaughterhouse wastewater	BOD	EC	86	0.71 US$/m^3	Asselin et al. [83]
	Oil-grease		99		
	COD_S		50		
	COD_T		82		
Potato chips manufacturing wastewater	COD	EC	60	0.48–5.42 US$/m^3	Kobya et al. [84]
	Turbidity		98		
Dyebath wastewater	COD	EC	76.9	1.84 €/m^3	Aygun et al. [57]
	Color		85.8		
Printing and packaging wastewater	COD	ECF-sedimentation	92.1	0.0001 US$/ kg COD removed	Present study
	Color		97.8	0.00007 US$/ kg color removed	

4. Conclusions

Color and COD as target pollutants were removed from printing and packaging wastewater using electrocoagulation/flotation-sedimentation. The present study established the simultaneous efficacy of critical parameters by RSM and electrodes consumption, as well as energy consumption as the value of operating costs. A four-factor (initial color content, initial pH, current density and treatment time), five-level, two series, of thirty assays of response surface optimization, were employed for color and COD removal efficiencies as target responses. The simultaneous efficacy of current density and treatment time demonstrates the most important effect. An increase in current density and treatment time over 6 mA/cm^2 and 40 min, respectively, leads to a dramatic increase in the percentage of color (<90%) and COD (<80%) removal. The optimum conditions which achieve greater pollutant removals were defined as an initial color content of 5,576.38 ADMI, initial pH of 7.29, a current density of 18.49 mA/cm^2 and a treatment time of 59.76 min. Concerning the results, to achieve 97.8% color and 92.1% COD removal from PPWW under optimized conditions, the operating costs were established as 0.07 US$ per kg color removed and 0.4 US$ per kg COD removed. Our results demonstrated that the removal of color and COD, at reasonable operating costs, was improved in comparison with other studies. This was attributed to electrode consumption rather than energy consumption. In conclusion, the ECF-sedimentation process, in collaboration with RSM optimization, is an efficient and cost-effective tool for the removal of a wide range of pollutants from industrial wastewater.

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References

[1] M. Esmaeili Bidhendi, Z. Asadi, A. Bozorgian, A. Shahhoseini, M.A. Gabris, S. Shahabuddin, R. Khanam, R. Saidur, New magnetic Co_3O_4/Fe_3O_4 doped polyaniline nanocomposite for the effective and rapid removal of nitrate ions from ground

water samples, Environ. Prog. Sustainable Energy, 39 (2020) 13306–13316.
[2] S.D. Islam, Electrocoagulation (EC) technology for wastewater treatment and pollutants removal, Sustainable Water Resour. Manage., 5 (2019) 359–380.
[3] N.T. Thuy, Application of electro-coagulation for treatment of wastewater from package printing process, Vietnam J. Sci. Technol., 55 (2017) 192–197.
[4] C.H. Tung, S.Y. Shen, J.H. Chang, Y.M. Hsu, Y.C. Lai, Treatment of real printing wastewater with an electrocatalytic process, Sep. Purif. Technol., 117 (2013) 131–136.
[5] L. Khannous, A. Elleuch, I. Fendri, N. Jebahi, H. Khlaf, N. Gharsallah, Treatment of printing wastewater by a combined process of coagulation and biosorption for a possible reuse in agriculture, Desal. Water Treat., 57 (2016) 5723–5729.
[6] D. Lichao, C. Yunnen, F. Jingbiao, An overview of the treatment of print ink wastewaters, J. Environ. Chem. Ecotoxicol., 3 (2011) 272–276.
[7] K.P. Papadopoulos, R. Argyriou, C.N. Economou, N. Charalampous, S. Dailianis, T.I. Tatoulis, A.G. Tekerlekopoulou, D.V. Vayenas, Treatment of printing ink wastewater using electrocoagulation, J. Environ. Manage., 237 (2019) 442–448.
[8] G. Borbély, E. Nagy, Removal of zinc and nickel ions by complexation–membrane filtration process from industrial wastewater, Desalination, 240 (2009) 218–226.
[9] S.V. Mousavi, A. Bozorgian, N. Mokhtari, M.A. Gabris, H.R. Nodeh, W.A.W. Ibrahim, A novel cyanopropylsilane-functionalized titanium oxide magnetic nanoparticle for the adsorption of nickel and lead ions from industrial wastewater: equilibrium, kinetic and thermodynamic studies, Microchem. J., 145 (2019) 914–920.
[10] E. GilPavas, I. Dobrosz-Gómez, M.Á. Gómez-García, Coagulation–flocculation sequential with Fenton or photo-Fenton processes as an alternative for the industrial textile wastewater treatment, J. Environ. Manage., 191 (2017) 189–197.
[11] D. Xu, Y. Li, L.F. Yin, Y.Y. Ji, J.F. Niu, Y.X. Yu, Electrochemical removal of nitrate in industrial wastewater, Front. Environ. Sci. Eng., 12 (2018) 9–23.
[12] A. Pourabadeh, B. Nasrollahzadeh, R. Razavi, A. Bozorgian, M. Najafi, Oxidation of FO and N_2 molecules on the surfaces of metal-adopted boron nitride nanostructures as efficient catalysts, J. Struct. Chem., 59 (2018) 1484–1491.
[13] Y. Zhang, H.Y. Shi, Y. Qian, Biological treatment of printing ink wastewater, Water Sci. Technol., 47 (2003) 271–276.
[14] A. Meteš, N. Koprivanac, A. Glasnovic, Flocculation as a treatment method for printing ink wastewater, Water Environ. Res., 72 (2000) 680–688.
[15] X. Chen, H.L. Sun, Dyeing and printing wastewater treatment using fly-ash coated with chitosan, Chin. J. Oceanol. Limnol., 27 (2009) 875–881.
[16] X. Zheng, J.X. Liu, Dyeing and printing wastewater treatment using a membrane bioreactor with a gravity drain, Desalination, 190 (2006) 277–286.
[17] X.J. Ma, H.L. Xia, Treatment of water-based printing ink wastewater by Fenton process combined with coagulation, J. Hazard. Mater., 162 (2009) 386–390.
[18] K.S. Hashim, R. AlKhaddar, A. Shaw, P. Kot, D. Al-Jumeily, R. Alwash, M.H. Aljefery, In: R. AlKhaddar, R. Singh, S. Dutta, M. Kumari, Advances in Water Resources Engineering and Management, Springer, Singapore 2020, pp. 219–235.
[19] K.S. Hashim, A. Shaw, R. Al Khaddar, M.O. Pedrola, D. Phipps, Defluoridation of drinking water using a new flow column-electrocoagulation reactor (FCER)-experimental, statistical, and economic approach, J. Environ. Manage., 197 (2017) 80–88.
[20] R. Niazmand, M. Jahani, S. Kalantarian, Treatment of olive processing wastewater by electrocoagulation: an effectiveness and economic assessment, J. Environ. Manage., 248 (2019) 109262–109270.
[21] A. García-García, V. Martínez-Miranda, I.G. Martínez-Cienfuegos, P.T. Almazán-Sánchez, M. Castañeda-Juárez, I. Linares-Hernández, Industrial wastewater treatment by electrocoagulation–electrooxidation processes powered by solar cells, Fuel, 149 (2015) 46–54.
[22] V. Kuokkanen, T. Kuokkanen, Recent applications of electrocoagulation in treatment of water and wastewater—a review, Curr. Opin. Green Sustainable Chem., 3 (2013) 89–121.
[23] M.M. Emamjomeh, Electrocoagulation Technology as a Process for Defluoridation in Water Treatment, University of Wollongong Thesis Collection, Australia, 2006.
[24] J.N. Hakizimana, B. Gourich, M. Chafi, Y. Stiriba, C. Vial, P. Drogui, J. Naja, Electrocoagulation process in water treatment: a review of electrocoagulation modeling approaches, Desalination, 404 (2017) 1–21.
[25] P.K. Holt, G.W. Barton, C.A. Mitchell, The future for electrocoagulation as a localised water treatment technology, Chemosphere, 59 (2005) 355–367.
[26] M. Elazzouzi, K. Haboubi, M. Elyoubi, Enhancement of electrocoagulation-flotation process for urban wastewater treatment using Al and Fe electrodes: techno-economic study, Mater. Today: Proc., 13 (2019) 549–555.
[27] S. Adamovic, M. Prica, B. Dalmacija, S. Rapajic, D. Novakovic, Z. Pavlovic, S. Maletic, Feasibility of electrocoagulation/flotation treatment of waste offset printing developer based on the response surface analysis, Arabian J. Chem., 9 (2016) 152–162.
[28] N. Chawaloesphosiya, J. Mongkolnauwarat, C. Prommajun, K. Wongwailikhit, P. Painmanakul, Treatment of cutting-oily wastewater by electrocoagulation-flotation (ECF) process: modeling approach, Environ. Eng. Res., 20 (2015) 392–396.
[29] F. Ghanbari, M. Moradi, A. Eslami, M.M. Emamjomeh, Electrocoagulation/flotation of textile wastewater with simultaneous application of aluminum and iron as anode, Environ. Process., 1 (2014) 447–457.
[30] S.M. Safwat, A. Hamed, E. Rozaik, Electrocoagulation/electroflotation of real printing wastewater using copper electrodes: a comparative study with aluminum electrodes, Sep. Sci. Technol., 54 (2019) 183–194.
[31] M.M. Emamjomeh, H.A. Jamali, Z. Naghdali, M. Mousazadeh, Carwash wastewater treatment by the application of an environmentally friendly hybrid system: an experimental design approach, Desal. Water Treat., 160 (2019) 171–177.
[32] A. Dimoglo, P. Sevim-Elibol, Ö. Dinç, K. Gökmen, H. Erdoğan, Electrocoagulation/electroflotation as a combined process for the laundry wastewater purification and reuse, J. Water Process Eng., 31 (2019) 100877.
[33] N.V. Narayanan, M. Ganesan, Use of adsorption using granular activated carbon (GAC) for the enhancement of removal of chromium from synthetic wastewater by electrocoagulation, J. Hazard. Mater., 161 (2009) 575–580.
[34] G.H. Chen, X.M. Chen, P.L. Yue, Electrocoagulation and electroflotation of restaurant wastewater, J. Environ. Manage., 126 (2000) 858–863.
[35] R. Katal, H. Pahlavanzadeh, Influence of different combinations of aluminum and iron electrode on electrocoagulation efficiency: application to the treatment of paper mill wastewater, Desalination, 265 (2011) 199–205.
[36] A.H. Essadki, B. Gourich, C. Vial, H. Delmas, M. Bennajah, Defluoridation of drinking water by electrocoagulation/electroflotation in a stirred tank reactor with a comparative performance to an external-loop airlift reactor, J. Hazard. Mater., 168 (2009) 1325–1333.
[37] J. Heffron, Removal of Trace Heavy Metals from Drinking Water by Electrocoagulation, Thesis, Marquette University, Milwaukee, Wisconsin, 2015.
[38] B. Abdulhadi, P. Kot, K. Hashim, A. Shaw, R. Al Khaddar, Influence of current density and electrodes spacing on reactive red 120 dye removal from dye water using electrocoagulation/electroflotation (EC/EF) process, IOP Conf. Ser.: Mater. Sci. Eng., 584 (2019) 012035.
[39] S. Chaturvedi, P.N. Dave, Removal of iron for safe drinking water, Desalination, 303 (2012) 1–11.
[40] M.M. Emamjomeh, M. Sivakumar, Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes, J. Environ. Manage., 90 (2009) 1663–1679.
[41] G. Chen, Electrochemical technologies in wastewater treatment, Sep. Purif. Technol., 38 (2004) 11–41.

[42] M.M. Emamjomeh, M. Sivakumar, A.S. Varyani, Analysis and the understanding of fluoride removal mechanisms by an electrocoagulation/flotation (ECF) process, Desalination, 275 (2011) 102–106.
[43] Z. Naghdali, S. Sahebi, M. Mousazadeh, H.A. Jamali, Optimization of the forward osmosis process using aquaporin membranes in chromium removal, Chem. Eng. Technol., 43 (2020) 298–306.
[44] E. Nazlabadi, M.A. Moghaddam, In: F. Kurisu, A. Ramanathan, A. Kazmi, M. Kumar, Trends in Asian Water Environmental Science and Technology, Springer, Cham, 2017, pp. 49–60.
[45] S. Zodi, O. Potier, F. Lapicque, J.P. Leclerc, Treatment of the industrial wastewaters by electrocoagulation: optimization of coupled electrochemical and sedimentation processes, Desalination, 261 (2010) 186–190.
[46] O. Chavalparit, M. Ongwandee, Optimizing electrocoagulation process for the treatment of biodiesel wastewater using response surface methodology, J. Environ. Sci., 21 (2009) 1491–146.
[47] X.S. Qin, B. Yang, F.R. Gao, G.H. Chen, Treatment of restaurant wastewater by pilot-scale electrocoagulation-electroflotation: optimization of operating conditions, J. Environ. Manage., 139 (2013) 1004–1016.
[48] M. Prica, S. Adamovic, B. Dalmacija, L. Rajic, J. Trickovic, S. Rapajic, M. Becelic-Tomin, The electrocoagulation/flotation study: the removal of heavy metals from the waste fountain solution, Process Saf. Environ. Prot., 94 (2015) 262–273.
[49] F. Ozyonar, B. Karagozoglu, Operating cost analysis and treatment of domestic wastewater by electrocoagulation using aluminum electrodes, Pol. J. Environ. Stud., 20 (2011) 173.
[50] S. Koyuncu, S. Arıman, Domestic wastewater treatment by real scale electrocoagulation process, Water Sci. Technol., 81 (2020) 656–667.
[51] A. Shokri, Application of electrocoagulation process for the removal of Acid orange 5 in synthetic wastewater, Iran. J. Chem. Chem. Eng., 38 (2018) 113–119.
[52] B.Y. Tak, B.S. Tak, Y.J. Kim, Y.J. Park, Y.-h. Yoon, G.-h. Min, Optimization of color and COD removal from livestock wastewater by electrocoagulation process: application of Box–Behnken design (BBD), J. Ind. Eng. Chem., 28 (2015) 307–315.
[53] APHA, AWWA, WPCF, WEF, Standard Methods for the Examination of Water and Wastewater, 2nd ed., American Public Health Association, American Water Works Association, Water Pollution Control Federation, Water Environment Federation, New York, 1915.
[54] M.M. Emamjomeh, M. Mousazadeh, N. Mokhtari, H.A. Jamali, M. Makkiabadi, Z. Naghdali, K.S. Hashim, R. Ghanbari, Simultaneous removal of phenol and linear alkylbenzene sulfonate from automotive service station wastewater: optimization of coupled electrochemical and physical processes, Sep. Sci. Technol., 54 (2019) 1–11.
[55] M. Mousazadeh, M.M. Emamjomeh, Evaluation of the Performance of Adsorption, Electrochemical (ECF), and Physical Hybrid Processes in Carwash Wastewater Treatment, Thesis, Qazvin University, Iran, 2019.
[56] M.E. Olya, A. Pirkarami, Electrocoagulation for the removal of phenol and aldehyde contaminants from resin effluent, Water Sci. Technol., 68 (2013) 1940–1949.
[57] A. Aygun, B. Nas, M.F. Sevimli, Treatment of reactive dyebath wastewater by electrocoagulation process: optimization and cost-estimation, Korean J. Chem. Eng., 36 (2019) 1441–1449.
[58] T. Karichappan, S. Venkatachalam, P.M. Jeganathan, Optimization of electrocoagulation process to treat grey wastewater in batch mode using response surface methodology, J. Environ. Health Sci. Eng., 12 (2014) 29.
[59] K. Praveen, K. Radha, N. Balasubramanian, Electrochemical treatment of plating effluent: kinetics and statistical modeling, Arch. Environ. Sci., 5 (2011) 17–24.
[60] N. Fayad, The Application of Electrocoagulation Process for Wastewater Treatment and for the Separation and Purification of Biological Media, Université Clermont Auvergne, France 2017.
[61] A. Dalvand, M. Gholami, A. Joneidi, N.M. Mahmoodi, Dye removal, energy consumption and operating cost of electrocoagulation of textile wastewater as a clean process, Clean–Soil Air Water, 39 (2011) 665–672.
[62] E. Bazrafshan, A.H. Mahvi, Textile wastewater treatment by electrocoagulation process using aluminum electrodes, Iran. J. Health Sci., 2 (2014) 16–29.
[63] B.K. Nandi, S. Patel, Effects of operational parameters on the removal of brilliant green dye from aqueous solutions by electrocoagulation, Arabian J. Chem., 10 (2017) S2961–S2968.
[64] Z. Beiramzadeh, M. Baqersad, M. Aghababaei, Application of the response surface methodology (RSM) in heavy metal removal from real power plant wastewater using electrocoagulation, Eur. J. Environ. Civ. Eng., (2019) 1–19, https://doi.org/10.1080/19648189.2019.1640139.
[65] E. Karamati-Niaragh, M.R.A. Moghaddam, M.M. Emamjomeh, E. Nazlabadi, Evaluation of direct and alternating current on nitrate removal using a continuous electrocoagulation process: economical and environmental approaches through RSM, J. Environ. Manage., 230 (2019) 245–254.
[66] S. Elabbas, N. Ouazzani, L. Mandi, F. Berrekhis, M. Perdicakis, S. Pontvianne, M.N. Pons, F. Lapicque, J.P. Leclerc, Treatment of highly concentrated tannery wastewater using electrocoagulation: influence of the quality of aluminum used for the electrode, J. Hazard. Mater., 319 (2016) 69–77.
[67] M.R. Sankar, V. Sivasubramanian, Application of statistical design to optimize the electrocoagulation of synthetic Congo red dye solution and predicting the mechanism, Int. J. Environ. Sci. Technol., 17 (2020) 1373–1386.
[68] M. Bayramoglu, M. Eyvaz, M. Kobya, Treatment of the textile wastewater by electrocoagulation: economical evaluation, Chem. Eng. J., 128 (2007) 155–161.
[69] D.T. Moussa, M.H. El-Naas, M. Nasser, M.J. Al-Marri, A comprehensive review of electrocoagulation for water treatment: potentials and challenges, J. Environ. Manage., 186 (2017) 24–41.
[70] O. Sahu, Suitability of aluminum material on sugar industry wastewater with chemical and electrochemical treatment processes, Int. J. Ind. Chem., 10 (2019) 335–347.
[71] J. Ge, J. Qu, P. Lei, H. Liu, New bipolar electrocoagulation–electroflotation process for the treatment of laundry wastewater, Sep. Purif. Technol., 36 (2004) 33–39.
[72] G. Hasani, A. Maleki, H. Daraei, R. Ghanbari, M. Safari, G. McKay, K. Yetilmezsoy, F. Ilhan, N. Marzban, A comparative optimization and performance analysis of four different electrocoagulation-flotation processes for humic acid removal from aqueous solutions, Process Saf. Environ. Prot., 121 (2019) 103–117.
[73] F. Janpoor, A. Torabian, V. Khatibikamal, Treatment of laundry waste-water by electrocoagulation, J. Chem. Technol. Biotechnol., 86 (2011) 1113–1120.
[74] M. Solak, M. Kılıç, Y. Hüseyin, A. Şencan, Removal of suspended solids and turbidity from marble processing wastewaters by electrocoagulation: comparison of electrode materials and electrode connection systems, J. Hazard. Mater., 172 (2009) 345–352.
[75] S. Abbasi, M. Mirghorayshi, S. Zinadini, A. Zinatizadeh, A novel single continuous electrocoagulation process for treatment of licorice processing wastewater: optimization of operating factors using RSM, Process Saf. Environ. Prot., 134 (2020) 323–332.
[76] H.C.L. Geraldino, J.I. Simionato, T.K.F. de Souza Freitas, J.C. Garcia, O. de Carvalho Júnior, C.J. Correr, Efficiency and operating cost of electrocoagulation system applied to the treatment of dairy industry wastewater, Acta Sci. Technol., 37 (2015) 401–408.
[77] S. Moosavirad, Treatment and operation cost analysis of greywater by electrocoagulation and comparison with coagulation process in mining areas, Sep. Sci. Technol., 52 (2017) 1742–1750.
[78] F. Deniz, C. Akarsu, Operating cost and treatment of boron from aqueous solutions by electrocoagulation in low concentration, Global Challenges, 2 (2018) 1800011.
[79] Y. Yavuz, Ü. Ögütveren, Treatment of industrial estate wastewater by the application of electrocoagulation process using iron electrodes, J. Environ. Manage., 207 (2018) 151–158.

[80] Z. Gönder, G. Balcıoğlu, Y. Kaya, I. Vergili, Treatment of carwash wastewater by electrocoagulation using Ti electrode: optimization of the operating parameters, Int. J. Environ. Sci. Technol., 16 (2019) 8041–8052.
[81] S. Mahesh, B. Prasad, I. Mall, I. Mishra, Electrochemical degradation of pulp and paper mill wastewater, Part 1. COD and color removal, Ind. Eng. Chem. Res., 45 (2006) 2830–2839.
[82] F. Orssatto, M.H.F. Tavares, F.M. da Silva, E. Eyng, L. Fleck, L.M. Frare, Optimization of the treatment of wastewater from a slaughterhouse and packing plant by the combination of electrocoagulation and tannin-based coagulant, Desal. Water Treat., 102 (2018) 82–92.
[83] M. Asselin, P. Drogui, H. Benmoussa, J.F. Blais, Effectiveness of electrocoagulation process in removing organic compounds from slaughterhouse wastewater using monopolar and bipolar electrolytic cells, Chemosphere, 72 (2008) 1727–1733.
[84] M. Kobya, H. Hiz, E. Senturk, C. Aydiner, E. Demirbas, Treatment of potato chips manufacturing wastewater by electrocoagulation, Desalination, 190 (2006) 201–211.