



Optimization of effluent treatment from healthcare waste incineration by electrocoagulation with iron electrodes

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ABSTRACT

This study evaluated the efficiency of electrocoagulation in removing chemical oxygen demand (COD), turbidity, and apparent color from the incineration effluent generated in a gas cleaning system (GCS). Modeling and optimization of the variables electric current (I), hydraulic retention time (HRT), and electrode distance (ED) were also performed in a batch reactor using iron electrodes. A 2³ rotatable central composite design CCRD was used, with a total of 19 trials, with electric currents ranging from 1A and 5A, a retention time of the effluent in the reactor from 15 to 40 minutes, and electrode distance of 1 and 3 centimeters. An algorithm with the desirability function was created to optimize simultaneously the parameters studied. The treatment of GCS by electrocoagulation was satisfactory in removing turbidity, apparent color, and COD, with maximum removal efficiencies above 70% for all parameters, using HRT of 27.5 minutes, ED of 2 centimeters, and electric current of 1 A. The statistical analysis showed a good fit of the model, with a coefficient of determination of $R^2 > 0.9$. The optimum operating condition was observed at 1A electric current, 27 minutes HRT, and 2 centimeters of electrode distance, with removals of 82.07, 86.86, and 70.82% of COD, turbidity, and apparent color, respectively. The simulated trials showed that lower electrolysis times can be used without impairing the treatment efficiency. Therefore, electrocoagulation may be a potential tool in the treatment of GCS.

Keywords: coagulation, electrolysis, gas scrubber, incineration.

Otimização do tratamento de efluentes da incineração de resíduos de saúde por eletrocoagulação com eletrodos de ferro

RESUMO

Este estudo teve como objetivo avaliar a eficiência da eletrocoagulação na remoção da demanda química de oxigênio (DQO), turbidez e cor aparente do efluente de incineração gerado em um sistema de limpeza de gases (SLG). A modelagem e otimização das variáveis corrente elétrica (I), tempo de retenção hidráulica (TRH) e distância do eletrodo (DE) também foram realizadas em um reator em batelada utilizando eletrodos de ferro. Foi utilizado um



delineamento composto central rotacional (DCCR) na 2^3 com um total de 19 ensaios, com correntes elétricas variando de 1A e 5A, tempo de retenção do efluente no reator de 15 a 40 minutos e distância dos eletrodos de 1 e 3 centímetros. Um algoritmo com função de conveniência foi criado para otimizar simultaneamente os parâmetros estudados. O tratamento da GCS por eletrocoagulação foi satisfatório na remoção de turbidez, cor aparente e DQO, com eficiência máxima de remoção acima de 70% para todos os parâmetros, utilizando TRH de 27,5 minutos, ED de 2 centímetros e corrente elétrica de 1 A. A análise estatística apresentou um bom ajuste do modelo com coeficiente de determinação de $R^2 > 0,9$. A condição ótima de operação foi observada em corrente elétrica de 1A, 27 minutos de TRH e 2 centímetros de distância do eletrodo, com remoções de 82,07, 86,86 e 70,82% de DQO, turbidez e cor aparente, respectivamente. Os ensaios simulados mostraram que tempos de eletrólise mais baixos podem ser utilizados, sem prejudicar a eficiência do tratamento. Portanto, a eletrocoagulação pode ser uma ferramenta potencial no tratamento de SLG.

Palavras-chave: coagulação, eletrólise, incineração, purificador de gás.

1. INTRODUCTION

Incineration is the most used method to treat healthcare waste (HCW), as it provides a significant reduction in the volume of waste and inactivation of pathogenic microorganisms. However, the emissions from HCW incineration often contain dioxins, polycyclic aromatic hydrocarbons, numerous volatile organic compounds, heavy metals, sulfur dioxide, nitrogen dioxide, particulate matter, and others, thus requiring atmospheric emission control (Thind *et al.*, 2021).

Emission control applications include the use of scrubber systems, in which pollutants are removed from the gas stream through the contact between the gas stream and a solvent, water, alkaline, or acidic solutions (Mendes, 2016). The effluents generated in the gas scrubbers of incineration plants (GCS) have a high polluting potential, with low pH and high concentration of dissolved and suspended solids. The fly ash contains contaminants like polychlorinated biphenyls, and trace metals such as, arsenic, beryllium, cadmium, chromium, lead, mercury and nickel, which can subsequently condense to form metallic particles, acid gasses, some salts, and chlorinated organic compounds (Silva and Lange, 2008; Bhargava, 2016; Thind *et al.*, 2021).

In the state of Bahia, there is no specific legislation for effluent discharge, thus CONAMA Resolution 430/2011 is adopted, which has established that the pH of effluents for discharge must be between 5 and 6, among other parameters. Therefore, the removal of pollutants from the effluents generated in the incinerator scrubbers is essential to mitigate possible impacts to the environment from their release into water resources.

Electrocoagulation (EC) is considered a promising technique for the treatment of effluents generated during washing and separation of pollutants from HCW incineration. Studies have shown its efficiency in the removal of contaminants, such as textile dyes (Papadopoulos *et al.* 2019), organic matter (Modenes *et al.*, 2017; Piovesan, 2017), and nitrogen and phosphorus (Orssatto, 2017).

As reported by Moussa *et al.* (2017), EC is an emerging technology for wastewater treatment since it combines the benefits of coagulation, flocculation, flotation, and electrochemistry.

The EC process consists of the deposition of coagulants in situ, by electrolytic oxidation reactions of conducting and semiconducting metals, triggered by the application of electric current (Chen, 2004; Vepsäläinen and Sillanpää, 2020), and iron stands out as the most used metal for this purpose (Modenes *et al.*, 2017; Papadopoulos *et al.*, 2019). In EC (Figure 1), ions from electrolytic oxidation of anode and cathode (iron) destabilize and neutralize the repulsive

forces that keep the particles suspended in water, favoring aggregation of the destabilized phase, contributing to easy physical removal of pollutants (Chen, 2004; Moussa *et al.*, 2017). The main function of the sacrificial anode is the generation of polymeric hydroxides while the cathode allows a reduction of metal ions (Chen, 2004). The formation of bubbles during the process can also assist in the flotation of pollutants to the aqueous surface (Hakizimana *et al.*, 2017). At acidic pH, iron is predominantly found as Fe^{3+} ions that can react with anions present in the effluent (SO_4^{2-} , F^- , Cl^- , organic matter, among others) (Vepsäläinen and Sillanpää, 2020). It can occur in bivalent or trivalent form depending on the oxidation state, and when subjected to hydrolysis, different metal species can coexist in solution (Moussa *et al.*, 2017). According to Chen (2004), the dissolution of coagulants by the application of electric current with iron electrodes is three times “higher when compared to aluminum electrodes.

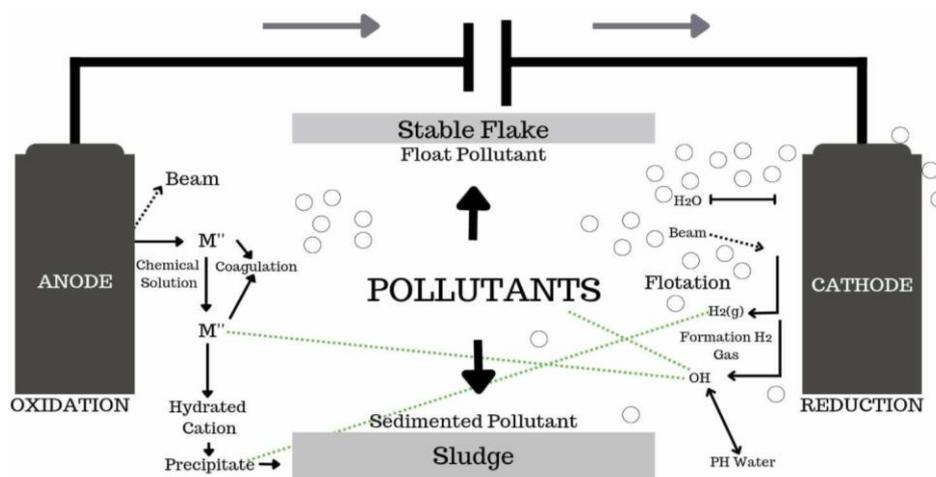


Figure 1. Schematic diagram of the electrocoagulation treatment.

The efficiency of electrocoagulation in the effluent treatment depends on several factors, including the concentration of pollutants and the operating conditions in the reactor, such as electrode area, hydraulic retention time, voltage, current density, types of electrodes, and the amount and spacing between them (Hakizimana *et al.*, 2017; Vepsäläinen and Sillanpää, 2020). Therefore, optimizing the factors involved in the process allows for problem-solving through the use of specific techniques under simulated conditions, enabling flexibility and application (Algeri *et al.*, 2017).

The importance of the waste treatment sector is noteworthy, given the increase in particle generation, mainly due to the COVID-19 pandemic, which has increased the demand for the health services around the world (OWS, 2020). There are no guidelines about the treatment of effluents from HCW incineration in fixed combustion chambers using water spray scrubbing systems.

Given the above, this study investigated the efficiency of electrocoagulation in removing COD, turbidity, and apparent color from HCW by modeling and optimization of the parameters, using a batch reactor and iron electrodes.

2. MATERIAL AND METHODS

2.1. Gas scrubber effluent from HCW incineration

The effluent from a gas scrubbing cleaning system was used in the study, from a dual-chamber incinerator, with a capacity of 50kg h^{-1} , located in a company that treats HCW from Groups A, B, and E. The company has an installed capacity of 385 tons of waste per year and generates $72\text{ m}^3\text{day}^{-1}$ of effluent. The system works in a closed circuit, with the reuse of the effluent generated, which is stored in three 5,000 L tanks.

The effluent was collected at the scrubber outlet after waste incineration (Groups A, B, and

E), and characterized for COD (closed reflux Method-5220 C), turbidity (nephelometric Method-2130 B), apparent color (Method 2120), pH (electrometric Method-4500 H+ B), iron concentration (flame atomic absorption spectrometry -FAAS), and electrical conductivity (Method 2510 A) (APHA *et al.*, 1995).

2.2. Experimental unit

The EC treatment consisted of a batch reactor composed of a rectangular glass box with capacity of 2 L. For each trial, 1.5 L of effluent was used, and the pH was corrected to 2.8 with sodium hydroxide (NaOH) solution. The system was kept under constant stirring (100 rpm).

The iron electrode was made from carbon steel plates A36. Electrodes consisting of four iron plates (7.0 cm width, 13.0 cm height, 1 mm thickness, 56 cm²) were arranged in parallel, and placed on a nylon technyl bar.

2.3. Experimental design and statistical analysis

A rotatable central composite design (RCCD) was used, with three independent variables (I, HRT, and ED). A 2³ complete factorial design was performed, including 6 axial points and 5 repetitions in the central point, totaling 19 trials, according to Table 1.

Table 1. Number of trials and combination of the actual (I, HRT, and ED) and coded independent variables.

N.	Trials	Coded levels			Actual Levels –Independent Variables		
		x1	x2	x3	I (A)	HRT (min)	ED (cm)
1	1	-1	-1	-1	1.809	20	1.5*
2	2	-1	-1	1	1.809	20	2.5*
3	3	-1	1	1	1.809	35	2.5*
4	4	1	-1	-1	4.19	20	1.5*
5	5	1	1	-1	4.19	35	1.5*
6	6	1	1	1	4.19	35	2.5*
7	7	-1	1	-1	1.809	35	1.5*
8	8	1	-1	1	4.19	20	2.5*
9	9	-1.68	0	0	1	27.5	2
10	10	1.68	0	0	5	27.5	2
11	11	0	-1.68	0	3	15	2
12	12	0	1.68	0	3	40	2
13	13	0	0	-1.68	3	27.5	1
14	14	0	0	1.68	3	27.5	3
15	15	0	0	0	3	27.5	2
16	15	0	0	0	3	27.5	2
17	15	0	0	0	3	27.5	2
18	15	0	0	0	3	27.5	2
19	15	0	0	0	3	27.5	2

* Values rounded from 1.4 cm to 1.5 cm and from 2.6 cm to 2.5 cm due to difficulties in setting up the design.

The electric currents varied from 1A (5.9 mA cm⁻²) to 5A (29.7mA cm⁻²), the retention time of the effluent in the reactor varied from 15 to 40 minutes, and the distance between the electrodes ranged from 1 to 3 centimeters. The operation conditions were determined in preliminary tests.

The results of COD, turbidity, and apparent color removal were used to determine the effluent purification efficiency.

From the RCCD data, statistical models were generated using the software Statistical Analysis System (SAS) 9.4 to evaluate the relationship between the response variables (removal

efficiency of COD, turbidity, and color) and the other independent variables. Analysis of variance (ANOVA) was performed to validate the models at a 95% confidence interval for F Test, and p-values indicated the significance of the models at a 95% confidence interval for the t-Test.

Response surface graphs were used to determine the optimal operating parameters, and the adjustment of the parameters of the quadratic models was done by regression analysis. Equation 1 shows the general model used.

$$\text{Removal of COD, or turbidity, or color} = \hat{\beta}_0 + \hat{\beta}_1 I + \hat{\beta}_2 \text{HRT} + \hat{\beta}_3 \text{DE} + \hat{\beta}_{12} I \cdot \text{HRT} + \hat{\beta}_{13} I \cdot \text{ED} + \hat{\beta}_{23} \text{HRT} \cdot \text{ED} + \hat{\beta}_{11} I^2 + \hat{\beta}_{22} \text{HRT}^2 + \hat{\beta}_{33} \text{ED}^2 \quad (1)$$

Where $\hat{\beta}_0, \hat{\beta}_1, \hat{\beta}_2, \hat{\beta}_3, \hat{\beta}_{12}, \hat{\beta}_{13}, \hat{\beta}_{23}, \hat{\beta}_{11}, \hat{\beta}_{22}, \hat{\beta}_{33}$ are the parameters estimated for the model; the coefficient $\hat{\beta}_0$ is the constant term; (I) is the electrical current; (HRT) is the hydraulic retention time; and (ED) is the electrode distance.

The desirability function approach defined by Derringer and Suich (1980) was used to simultaneously optimize the parameters, which provides direct search optimization algorithms in Scilab for the variables studied.

3 RESULTS AND DISCUSSION

3.1. Characterization of the effluent

A high iron concentration (259.97 mg L⁻¹) and high electrical conductivity (1396.65 ms cm⁻¹), was observed in the effluent, which contributes to an effective electrocoagulation, since it leads to a decrease in the ohmic resistance of the effluent (Tahreen *et al.*, 2020).

The effluent exhibited 319 mg O₂ L⁻¹, 360 NTU, and 170 mg Pt L⁻¹ for COD, turbidity, and apparent color, respectively, indicating a large amount of suspended and dissolved solids, giving it a dark appearance. There are few studies in the literature on the physicochemical properties of the effluent generated in scrubber systems after incineration of health service waste, without pH neutralization. Silva and Santos (2019) characterized the effluents from the gas cleaning system after incineration of hazardous solid waste, in a moving bed type plant, subjected to acidic or alkaline washing, through six monitoring trials. The authors reported turbidity up to 97 NTU, color changes from 10 to 208 mg Pt L⁻¹, total suspended solids between 65 and 3080 mg L⁻¹, and neutral pH of the effluent, due to the capture of the acid gases by the sodium hydroxide during the alkaline washing. They also reported that the COD of the ash washing water varied between 370 and 3004 mgO₂ L⁻¹, which indicates a relevant amount of organic matter present in that type of effluent.

The effluent of this study presented a high acidity, and pH 1.85. This result may be due to the formation of oxides during the combustion of solid waste, which forms strong acids when interacting with the water used in the atmosphere control system (Sampaio, 2014).

3.2. Pollutant Removal Efficiency

The different efficiencies of turbidity, COD, and apparent color removal after 19 trials are presented in Figure 2. All parameters reached more than 70% removal in at least one trial.

The turbidity removal efficiency varied between 71.39% and 89.77% indicating a great removal of suspended solids for all assays. The higher turbidity removal was observed for Trial 9, with 27.5 minutes of HRT, electrode distance of 2 cm, and 1^a of electric current or 5.9 mA.cm⁻² of current density. In turn, the lower efficiency was observed for Trial 6, with 35 minutes of HRT, 2.5 cm electrode distance, and 4.19 A or 29.7 mA.cm⁻² of current density.

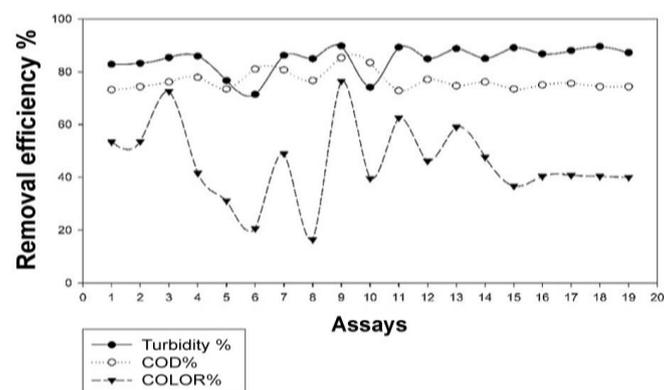


Figure 2. Removal efficiency for turbidity, COD, and apparent color.

For COD, removal efficiency ranged from 72.78% to 85.32%, proving the effectiveness of the treatment in the removal of organic compounds. A higher COD removal efficiency was observed for Trial 9, while Trial 11 presented the lower removal efficiency.

Concerning the apparent color, as also observed for turbidity and COD, Trial 9 exhibited the higher removal efficiency, with 76.47%, while Trial 8 presented the lower removal efficiency, with 16.47%. These results show that electrocoagulation is efficient in the removal of dissolved solids as a function of the processing conditions. The low color removal (47.06%) may be due to a large amount of Fe present in the samples resulting from anodic dissolution (Moussa *et al.* 2017), since pH values below 5 during electrocoagulation lead to a predominance of trivalent Fe and Fe (OH)₃ (Vepsäläinen and Sillanpää, 2020). Furthermore, the wastewater under study was subjected to no filtration procedure. After 24 h, iron decantation and a significant improvement of the effluent clarity were observed.

The use of EC for pollutant removal using iron electrodes in this study was also performed by other authors. Módenes *et al.* (2017) reported removal efficiencies of 92, 99.63, and 99.16% for COD, color, and turbidity, respectively, in the treatment of poultry slaughterhouse effluent, while Piovesan (2017) studied landfill leachate treatment and found 86.58, 70.09, and 55.69% removal of turbidity, color, and COD, respectively, using 2 cm of ED.

3.3. Modeling of the GCS electrocoagulation

The adjusted models that describe the behavior of the response variables at a 95% confidence interval ($p < 0.05$) are presented in Equations 2, 3, and 4.

$$\text{Turbidity Removal} = 57.1832 + 15.63602x_1 + 0.75156x_2 + 3.88284x_3 - 0.34162x_1 \cdot x_2 - 1.95622x_1 \cdot x_3 - 0.9157x_1^2 \quad (2)$$

$$\text{COD removal} = 80.99837 - 12.15065x_1 + 0.54821x_2 + 0.0546x_3 - 0.1625x_1 \cdot x_2 + 2.70046x_1^2 \quad (3)$$

$$\text{Apparent color removal} = 155.22376 - 5.57645x_1 - 2.65721x_2 - 57.56173x_3 - 13.51197x_1 \cdot x_3 + 1.43413x_2 \cdot x_3 + 3.24454x_1^2 + 13.59540x_3^2 \quad (4)$$

The model validation was performed by analysis of variance (ANOVA). The results (Table 2) showed reasonable regressions for turbidity, COD, and color, once they presented satisfactory fit to the model, with determination coefficients (R^2) higher than 90%, and adjusted R^2 higher than 84%, with a confidence interval of 95% ($p < 0.05$) in the F test, indicating no significant difference between the experimental and predicted values. The discrepancies may

be due to the presence of noisy data.

Table 2. ANOVA for turbidity, COD, and apparent color removal

Parameter	DF	SS	R ²	Adjusted -R ²	RMSE	CV	F tab.	F value	p-value
Turbidity	7	445.47060	0.9432	0.9034	1.63824	1.93973	3.135	23.71	<.0001
COD	6	173.48380	0.9020	0.8433	1.37263	1.80409	3.217	15.35	0.0002
Color	8	3786.966	0.9763	0.9526	3.38837	7.51635	3.438	41.23	<.0001

The response surface of the model (Figure 3) showed wide optimal ranges of electric current, hydraulic retention time, and electrode distance for high turbidity removals. For the electric current, the range with the best removal varied from 1 A (5.9 mA cm⁻²) to 3.2 A (19.04 mA cm⁻²), which was limited by increasing ED and HRT. The variables ED and HRT showed high turbidity removal for all conditions tested, with a small improvement for the conditions of 2.5 to 3 cm and 35 to 40 min, respectively.

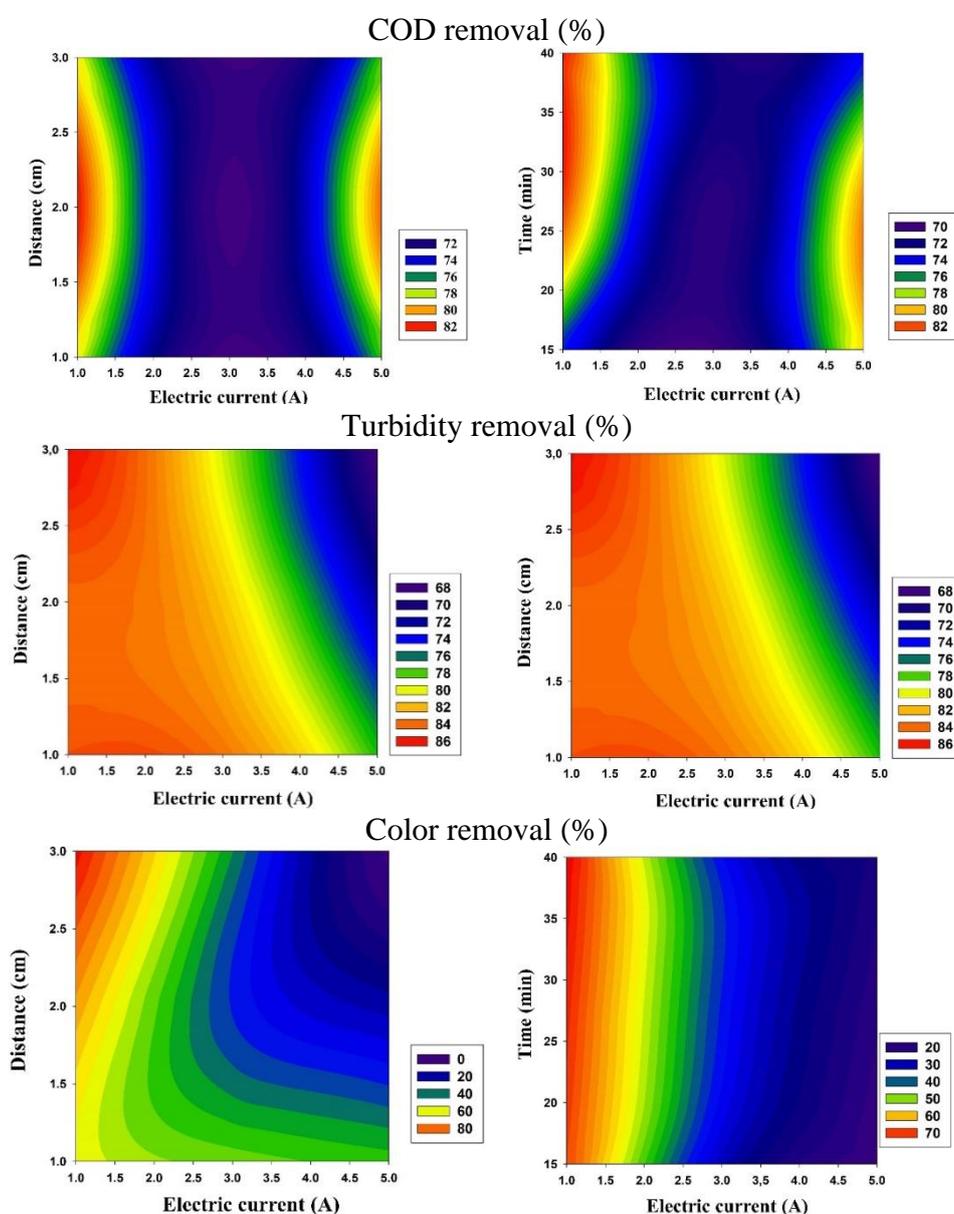


Figure 3. Response surfaces for percent removal of COD, turbidity, and apparent color as a function of electric current, electrode distance, and hydraulic retention time.

According to the response surface in Figure 3, the optimum COD removal was represented by a minimum point in the center and two optimal points for each independent variable, which was near 1A (5.9 mA cm^{-2}) and 5 A (29.7 mA cm^{-2}). For a lower current value, which is a desirable condition due to the lower operation costs, the optimal ED ranged from 1.3 -2.5 cm and HRT from 24 to 40 min.

The response surfaces for color removal efficiency showed a tendency towards a lower optimal range for the electric current, from 1 to 1.5 A, for all HRT conditions, as observed for COD, and an optimal range for ED from 1.7 - 3.0 cm.

The tendency of increasing the turbidity and color removals in Figure 3 is due to the higher efficiency of the electrocoagulation from the beginning of the process, with the application of a lower electric current. This behavior indicates that higher concentrations of iron hydroxide species are not necessary to clarify the effluent, once the crude effluent has high Fe concentration, which when submitted to hydrolysis in the system contributes to the formation of coagulants, which is a pH-dependent event (Moussa *et al.*, 2017; Vepsäläinen and Sillanpää, 2020).

However, the COD removal (Figure 3) demanded a longer hydraulic retention time, which was also observed by Combatt *et al.* (2017), who studied the elimination of COD from slaughterhouse effluent using an iron electrode and similar pH and current density conditions, and reported a removal efficiency of 88.8% for an HRT of 60 minutes.

Moreno-Casillas *et al.* (2007) investigated the electrocoagulation for COD removal and concluded that the removal efficiency and its variability depends on several factors, including the formation of agglomerates, which generally occurs at pH above 7.5 for Fe electrodes, the reactivity of organic compounds with Fe oxides, the solubility of the compounds formed, the final pH (especially for acidic compounds), and the pH increment with a consequent increase in the acidity. Probably, in this study, higher COD removals were not observed due to the low pH of GCS, which was lower than 6 for all trials.

3.4. Global optimization of the GCS electrocoagulation

The global optimization of GCS by electrocoagulation is necessary since wide and different regions of maximum efficiency were found for the three response variables. The Scilab algorithm using the desirability function allowed determining the global desirability $D=1$ (Figure 4) of the system, which was maximized when all responses reached the maximum removal. The optimal point of the modeled system was reached with an electric current of 1A (5.9 mA cm^{-2}), 27 minutes of HRT, and 2cm of ED, for simultaneous removal of 82.07, 86.86, and 70.82% of COD, turbidity, and apparent color, respectively.

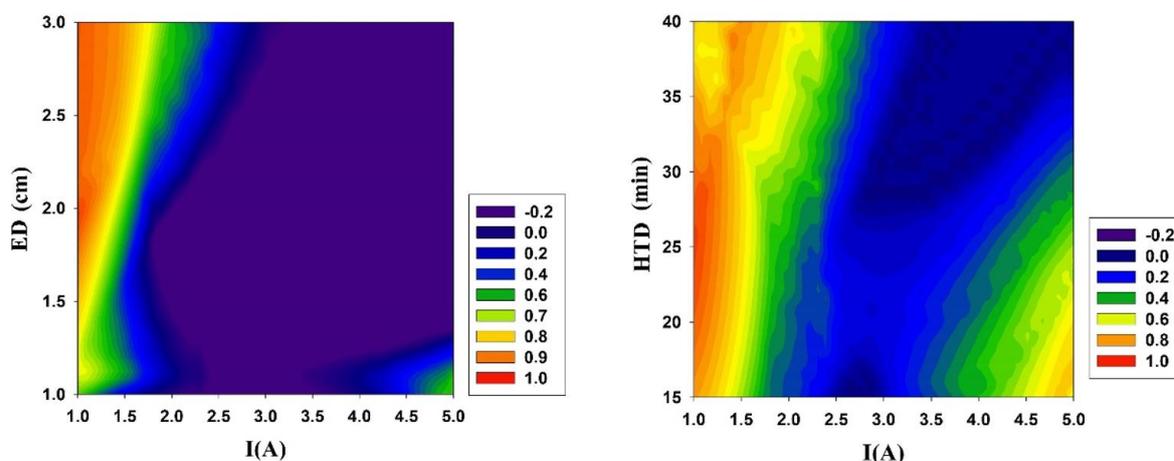


Figure 4. Response surface plots for overall desirability of GCS electrocoagulation using iron electrodes, for the variables ED (cm), I(A), and HRT (min).

The response surface plots for the global desirability function (Figure 4) confirmed the optimal point, showing that the factors ED and HRT can vary in a wide range without affecting the global desirability, i.e., a good removal efficiency can be reached unlike the electric current (I) that exhibited a narrow optimal range, causing a sharp drop in the global desirability even with small variations, which impairs the effluent treatment.

The optimum point observed for the current of 1A ($5.9 \text{ mA}\cdot\text{cm}^{-2}$) evidenced that this variable is a key operational parameter for the performance of the EC treatment of GCS. The antagonistic effect of this parameter for pollutant removal is due to the high iron concentration in the effluent. In contrast, some authors reported higher treatment efficiency with increasing the electric current, once it directly affects the dissociation of metal ions through anode oxidation and hydrogen formation at the cathode (Papadopoulos *et al.*, 2019; Vepsäläinen and Sillanpää, 2020). Therefore, increasing the EC in the treatment of effluents with high iron concentration may not improve treatment efficiency.

In contrast, Módenes *et al.* (2017) reported high removal efficiency of pollutants in wastewater by electrolytic processes at low current or current density, with maximum removals of 92.00, 99.63, and 99.16% for COD, color, and turbidity, respectively, with an electrolysis time of 50 min and current intensity of 2 A.

An electrode distance of 2 cm is effective to prevent the passivating effect of the iron electrode and the increase in resistivity. As shown in Figure 4, at a lower current, ED lower than 1.5 cm led to a reduction in desirability index. According to Feng *et al.* (2021), lower distances can hinder the rapid movement of the bubbles and agglomerates generated in the plate electrolysis, and accelerate plate passivation, increasing resistivity, leading to reduced current efficiency and ion production rate.

Despite the wide optimal range of ED from 1.5 cm (Figure 4), the increase in the spacing between the electrodes may not be an effective approach, once the passivating effect of the anode is potentiated with the increase of the distance between electrodes, leads to the formation of metal hydroxide film on the electrode surface, inhibits the oxidation of the anode, and decreases the amount of coagulant *in situ*, which leads to a lower efficiency in the removal of pollutants (Ghosh *et al.*, 2005). In addition, the decrease in ohmic potential leads to maximum pollutant removal efficiency. This decrease in potential is proportional to the distance between electrodes, so its reduction is required to decrease energy costs (Chen, 2004) and electrolysis time, and increase the efficiency of the process, because there is an increase in mass transfer during the CE due to turbulence generated by the large formation of gas bubbles in this condition (Martínez-Villafañe *et al.*, 2009).

The electrolysis time can also directly affect the coagulant production *in situ*. According to Chen (2004), the COD removal in electrocoagulation occurs by oxidation of organic matter at the anode at higher detention times, allowing all electrochemical reactions to occur. The increase in the concentration of ions and their hydroxide agglomerates is proportional to the electrolysis time.

4. CONCLUSION

The crude effluent from GCS presented high concentrations of COD, iron, as well as very acidic pH and high turbidity and apparent color, therefore requiring treatment before release into water bodies.

The high removal of turbidity, COD, and apparent color of this study showed that electrocoagulation was effective for the treatment of gas scrubber effluents.

Statistical analysis showed that mathematical models can be used to simulate the effects of the parameters electric current intensity, hydraulic retention time, and electrode distance on turbidity, COD, and color removal. All models presented a good fit to the data, with a coefficient of determination $R^2 > 0.9$ at a 95% confidence interval.

The optimum point was reached with an electrical current of 1A, a hydraulic retention time of 27 minutes, and electrode distance of 2 centimeters, with removals of 82.07, 86.86, and 70.82% of COD, turbidity, and apparent color, respectively. The simulated conditions showed that lower electrolysis times can be used without impairing the treatment efficiency.

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