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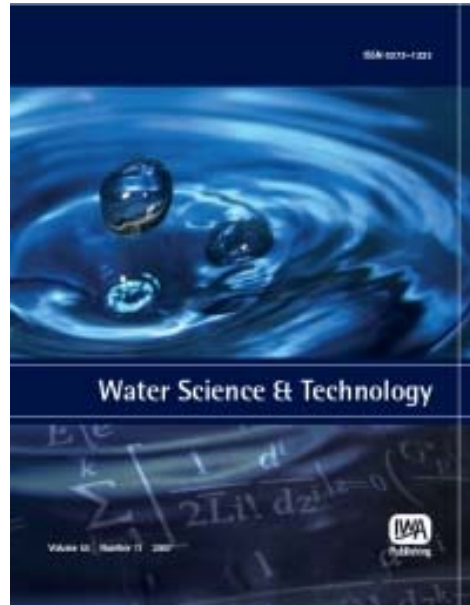


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## Treatment of automotive industry oily wastewater by electrocoagulation: statistical optimization of the operational parameters

Edison GilPavas, Kevin Molina-Tirado and Miguel Ángel Gómez-García

### ABSTRACT

An electrocoagulation process was used for the treatment of oily wastewater generated from an automotive industry in Medellín (Colombia). An electrochemical cell consisting of four parallel electrodes (Fe and Al) in bipolar configuration was implemented. A multifactorial experimental design was used for evaluating the influence of several parameters including: type and arrangement of electrodes, pH, and current density. Oil and grease removal was defined as the response variable for the statistical analysis. Additionally, the BOD<sub>5</sub>, COD, and TOC were monitored during the treatment process. According to the results, at the optimum parameter values (current density = 4.3 mA/cm<sup>2</sup>, distance between electrodes = 1.5 cm, Fe as anode, and pH = 12) it was possible to reach a c.a. 95% oils removal, COD and mineralization of 87.4% and 70.6%, respectively. A final biodegradability (BOD<sub>5</sub>/COD) of 0.54 was reached.

**Key words** | Al electrode, electrocoagulation, Fe electrode, oily wastewater, optimization

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### INTRODUCTION

Oily wastewater is widely resulting in metal industry processing, since these fluids provide the combination of cooling and lubrication required by numerous metalworking operations. Among many, they include metallurgic industry, oil and gas exploration and processing, mechanic industry (e.g., auto-parts), and the automobile industry. Oily wastewater is comprised of many compounds, amongst them the more important are oil, grease, and surfactants. Oil can appear in water in three different conditions: as an immiscible substance, as an unstable emulsion, or as a secondary oily emulsion. In the first two cases, oil separation is quite simple and involves the use of mechanical or physicochemical means. Due to the presence of surfactants and co-surfactants, secondary emulsions are quite stable, with the presence of micrometric emulsified oily droplets which makes difficult their separation by conventional methods (Zouboulis & Avranas 2000). The treatment of oily wastewater has been addressed by different

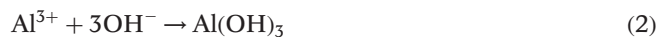
techniques, but the most commonly used are membrane processes (microfiltration and ultrafiltration) (Scott *et al.* 2001; Moulai-Mostefa *et al.* 2005), chemical destabilization (conventional coagulation) (Rios *et al.* 1998), flocculation (Pinotti *et al.* 2001), electroflotation (Burns *et al.* 1999; Chen *et al.* 2002; Moulai-Mostefa & Tir 2004), air flotation (Al Shamrani *et al.* 2002), and electrochemical destabilization (electrocoagulation, EC) (Lai & Lin 2004; Xu & Zhu 2004; Yang 2007). EC has been established firmly as an industrial wastewater treatment since it does not require extra chemical (additives) for destabilizing the oily emulsion. Moreover, it involves simple equipment which is easily operable; it has very low cost investment, and a minimum sludge generation (Rajeshwar & Ibanez 1997; Yousuf *et al.* 2001). The EC process uses direct current (DC) for releasing an active coagulant (cation) from the anode (the “sacrificed” anode), which in turn reacts with the hydroxide ions generated on the cathode, destabilizing suspended,

emulsified, or dissolved contaminants in the aqueous medium (Zhu *et al.* 2007). Finally, colloidal matters agglomerate to be eliminated by flotation or settling. The EC process involves three main steps: surface reaction on the electrodes, coagulant formation by electrolytic oxidation in the aqueous phase, and the adsorption of colloidal particles in the coagulant. Therefore, electrodes selection (kind and arrangement) is a main concern for applying this technology. Typically, Al or Fe is used for EC due to their lower price and accessibility (Chen 2000; Koyba *et al.* 2003; Yousuf *et al.* 2004; Kobya *et al.* 2006). Some of the chemical reactions taking place at the anode are given as follows.

For the aluminium anode:



at alkaline conditions:



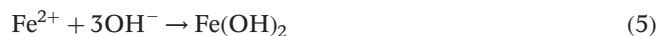
at acidic conditions:



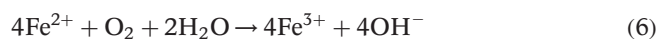
For the iron anode:



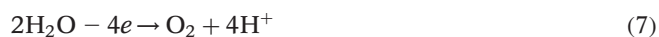
at alkaline conditions:



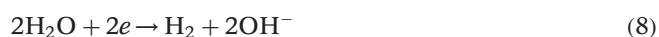
at acidic conditions:



In addition, there is oxygen evolution reaction as:



And, the reaction at the cathode is:



If iron or aluminium electrodes are used, the generated  $\text{Fe}^{3+}$  (aq) or  $\text{Al}^{3+}$  (aq) ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. For example: by

hydrolysis,  $\text{Al}^{3+}$  ions may generate  $\text{Al}(\text{H}_2\text{O})_6^{3+}$ ,  $\text{Al}(\text{H}_2\text{O})_5\text{OH}^{2+}$ , and  $\text{Al}(\text{H}_2\text{O})_4(\text{OH})^{2+}$ ; these hydrolysis products may form, over a wide pH range, many monomeric and polymeric species such as:  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}(\text{OH})_2^+$ ,  $\text{Al}_2(\text{OH})_2^{4+}$ ,  $\text{Al}(\text{OH})^{4-}$ ,  $\text{Al}_6(\text{OH})_{15}^{3+}$ ,  $\text{Al}_7(\text{OH})_{17}^{4+}$ ,  $\text{Al}_8(\text{OH})_{20}^{4+}$ ,  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ ,  $\text{Al}_{13}(\text{OH})_{34}^{5+}$  (Rebhun & Lurie 1993). Similarly, ferric ions generated by electrochemical oxidation of iron electrode may form monomeric ions,  $\text{Fe}(\text{OH})_3$ , and, depending on the pH of the aqueous medium, polymeric hydroxy complexes namely:  $\text{Fe}(\text{H}_2\text{O})_6^{3+}$ ,  $\text{Fe}(\text{H}_2\text{O})_5(\text{OH})^{2+}$ ,  $\text{Fe}(\text{H}_2\text{O})_4(\text{OH})_2^+$ ,  $\text{Fe}_2(\text{H}_2\text{O})_8(\text{OH})_2^{4+}$ , and  $\text{Fe}_2(\text{H}_2\text{O})_6(\text{OH})_4^{4+}$  (Pykhiteev *et al.* 1999). These hydroxides/polyhydroxides/polyhydroxymetallic compounds have strong affinity for dispersed particles as well as counter ions to cause coagulation. The gases evolved at the electrodes may have an effect on the flotation of the coagulated materials (Chen 2004; Yousuf *et al.* 2004).

The optimization of an EC process implies to establish the operational condition for oil release from the water-oil emulsion. This problem could be defined as the emulsion destabilization: destruction of the interfacial layer overcoming the double-layer electric repulsion effects, allowing larger oil drop formation by coalescence. Due to the quantity of variables and interactions involved in such a process, its analysis requires a lot of efforts and is time consuming. The experimental design technique is an efficient method to establish the relative importance between many variables and their interactions (Pykhiteev *et al.* 1999). Among those analytical methods, the response surface method (RSM) was chosen and implemented as proposed by many other authors in the literature Nordin *et al.* (2004), Montgomery (2005), Murugesan *et al.* (2007), Wang *et al.* (2007) and Tir & Moulai-Mostefa (2008).

This study uses an EC process for treatment of oily wastewater generated from an automotive industry placed in Medellín (Colombia). Both, Fe or Al anodes were tested as “sacrificed” anode. The wastewaters are characterized by Biological Oxygen Demand ( $\text{BOD}_5$ ), Chemical Oxygen Demand (COD), Total Organic Carbon (TOC), and oil and grease removal from the emulsion. Four factors were included to be analyzed in the EC process: pH, type and configuration of electrodes, and current density. The RSM was used in order to define the optimal operational values of chosen factors.

**Table 1** | Oily wastewater characterization

Parameter	Conductivity (mS/cm)	Al, mg/l	Fe, mg/l	Grease (mg/l)	COD, mg/l	BOD <sub>5</sub> , mg/l	TOC, mg/l	pH
Value	15	1.97	0.03	800	5,582	919	800	12

## MATERIALS AND METHODS

### Sample

Samples were taken directly from their source: an automotive industry located in Medellin (Colombia). That factory produces c.a. 12 m<sup>3</sup>/day of an oily wastewater which characteristics are presented in Table 1.

From data presented in Table 1, a BOD<sub>5</sub>/COD ratio equals to 0.1646 indicates that the oily wastewater is not biodegradable.

### Electrocoagulation cell

The removal of oily wastewater by electrochemically generated coagulants was carried out in a 2.7 liters batch electrocoagulation glass cell, which consists of four vertical plate electrodes (2 made of Fe and 2 made of Al), having equally effective surface areas of 65 cm<sup>2</sup> and a gap between 1 cm and 3 cm which was optimized. Electrodes were connected in a bipolar arrangement. Batches of 2 litres of the wastewater were run into the EC cell at 450 rpm. The completely submerged electrodes were connected to the positive and negative terminals of the DC power supply unit (BK Precision, Protek). Experiments were carried out by varying cell voltage (0–30 V; 0–5 A). All tests were performed during 180 minutes. The solution pH was adjusted with H<sub>2</sub>SO<sub>4</sub> (Merck).

### Analytical methods

Standard methods were used for the quantitative analysis of grease and oil content, COD, BOD<sub>5</sub>, and TOC, as in APHA, AWWA, WPCF (2005). The Soxhlet method (5520D) was used to characterize the wastewater grease and oil content. The COD analyses were performed following the closed reflux method, with colorimetric measurements (5310D). Total organic carbon (TOC) was analysed with a Shimadzu analyser (model TOC-5,000A), and the BOD<sub>5</sub> measurements were performed following the respirometric method (5210B).

The treatment process efficiency was defined from the grease and oil removal percentage, COD, and TOC, each calculated from the following expression:

$$\%DF = \frac{(C_0 - C)}{C_0} \times 100 \quad (9)$$

where  $C_0$  and  $C$  correspond to the initial and final concentrations, respectively.

### Experimental design & statistical analysis

The response surface method (RSM) was chosen and implemented to define the optimal operational conditions of the EC process. A multifactorial experimental design was defined in order to evaluate the influence of several parameters including: type and separation of electrodes, pH, and current density. Three or two different levels (values) were chosen for each parameter. Two

**Table 2** | Chosen variables levels for each group of experiments: (A) First, (B) Second

Variable	X code		
	–1	1	0
	Level 1	Level 2	Level 3
A			
pH	4	8	12
Electrode	Fe	Al	–
B			
Electrodes gap (cm)	1	2	3
Current density (mA)	1.4	2.85	4.3

**Table 3** | Experimental results for the first group of variables

Test	pH	Anode	%GR	%GR, duplicate
1	12	Fe	98.87	98.87
2	12	Al	79.81	80.21
3	8	Fe	74.22	75.17
4	8	Al	47.48	46.91
5	4	Fe	17.18	18.99
6	4	Al	10.38	8.13

**Table 4** | ANOVA for grease and oil removal. Results for the first group of variables

Factor	Square sum	Freedom degrees	Square average	f ratio	p value
A: pH	11,238.8	1	11,238.8	600.72	0.0000
B: type of electrode	987.36	1	987.36	52.78	0.0003
AA	219.555	1	219.555	11.74	0.014
AB	528.906	1	528.906	2.83	0.1437
Blocks	0.001875	1	0.001875	0	0.9923
Total error	112.255	6	187.088		

$R^2 = 99.11\%$

different experiments were planned: in a first group of experiments, the effect of pH and type of electrode were analyzed; in a second group of experiments, the distance between electrodes and the current density were studied. Variable levels and codifications are presented in Table 2.

Variables were coded according to Equation (10) (Rebhun & Lurie 1993)

$$X_i = \frac{(x_i - x_{pc})}{\Delta x_i} \quad (10)$$

where  $X_i$  is the code level,  $x_i$  is the uncoded value,  $x_{pc}$  corresponds to uncoded value at the central point, and  $\Delta x_i$  is the change value between levels (Montgomery 2005).

Oil and grease removal (%GR) was defined as the response variable for the statistical analysis, which was based on the analysis of variance (ANOVA), Pareto diagram, response surface plot, and the correlation coefficients. Experimental data were analyzed by using Statgraphics 5.1 (1999–2004). For the response surface method, the experimental results were adjusted to a second order multivariable polynomial as presented in Equation (11):

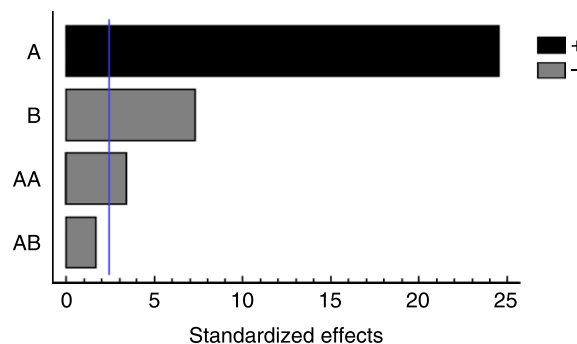
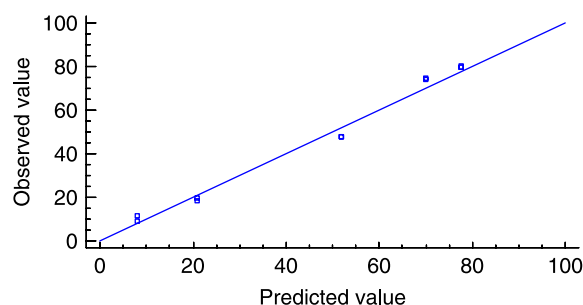
$$Y_i = \beta_0 + \sum_1^2 \beta_i X_i + \sum_1^2 \beta_{ii} X_i^2 + \sum_1^2 \beta_{ij} X_{ij} \quad (11)$$

where  $Y_i$  was the response variable;  $\beta_0$ ,  $\beta_i$ ,  $\beta_{ii}$ , and  $\beta_{ij}$  were the regression coefficients which corresponds to the intercept, lineal, quadratic, and interactions, respectively; and  $X_i$  and  $X_j$  are the independent variables. The quality of this model and its prediction capacity are related to the variation coefficient,  $R^2$ . This experimental design was selected in order to optimize the number of experiments. Thus, at first, the two variables of this first group of

experiments were optimized for %GR; then, those optimal results were kept fixed for the second group as proposed by Montgomery (2005).

## RESULTS AND DISCUSSION

Experimental results for the first group of variables are presented in Table 3, for the conditions presented in Table 2(A).

**Figure 1** | Pareto diagram for the EC process for the first group of factors: A = pH and B = Type of electrode.**Figure 2** | Comparison of observed and predicted values for the %GR for the first group of factors.

**Table 5** | Experimental (Exp.) and adjusted results for the second group of variables

Test	Electrode gap, (cm)	Current density (mA/cm <sup>2</sup> )	%RG		% RG, duplicate	
			Exp.	Adjusted	Exp.	Adjusted
1	1	1.4	91.05	90.81	90.54	90.84
2	1	2.85	95.0	95.34	95.2	95.37
3	1	4.3	99.1	98.73	98.95	98.75
4	2	1.4	94.1	94.02	94.3	94.05
5	2	2.85	97.5	97.09	97.3	97.12
6	2	4.3	98.64	99.02	98.5	99.05
7	3	1.4	93.4	95.14	96.64	95.17
8	3	2.85	97.86	96.76	95.59	96.78
9	3	4.3	97.5	97.23	97.34	97.25

The higher pH value, the higher %RG was obtained (Table 3). The best results were obtained using Fe as the “sacrificed” anode. The ANOVA, Table 4, allows analyzing the significance and interaction between variables.

From Table 4 it is possible to conclude from all four factors (A, B, AA, AB), three are statistically significant for the %GR; and that it is pH the most significant parameter. Since  $R^2 = 99.11\%$ , it can also be concluded that proposed model adjust properly the variability of %GR. The Pareto diagram, Figure 1, allows assigning the priority order or significance of the analyzed factors for the EC process. They correspond to all those values that surpass the inner vertical line.

From Figure 1, it can be seen that pH (A), type of electrode (B), and pH-pH (AA) affect significantly the EC process. The only factor that has not influence on the process is the interaction pH-type of electrode (AB). These results confirm the ANOVA ones. In fact, it has been

proposed in the literature that pH is one of the main factors influencing the EC process (Do & Chen 1994). During the EC experiments, the emulsion pH was monitored. It was found that its value increases. This fact has been explained by different authors considering the characteristics of the process itself: hydrogen formation on the electrode (Vik *et al.* 1984), CO<sub>2</sub> liberation (Rebhun & Lurie 1993), chemical dissolution of the metal and formation of species such as Fe<sup>3+</sup>, Fe(OH)<sup>2+</sup>, and Fe(OH)<sup>4+</sup> (Bensadok *et al.* 2008; Yilmaz *et al.* 2008). So, the best results for %GR were obtained at alkaline pH. It suggests that the presence of hydroxides species facilitates the formation of agglomerates which are finally removed from the treated solution (Heidmann & Calmano 2008; Song *et al.* 2008). At the studied conditions, the %GR is properly adjusted by a second order regression model as presented in Figure 2 which compares the observed and predicted values.

In Figure 2, data adjust to a straight line with a correlation factor  $R^2$  equals to 0.991. From the results obtained for the first group of experiments, the optimal values of the considered factors are: pH = 12 and the anode should be made of Fe. These two variables will be kept fixed for the second group of experiments.

The second group of experiments includes as factors: the current density and the gap between electrodes. They were analyzed including three values or levels defined in Table 2(B). Both, experimental and adjusted results (to a second order regression model, see Equation (11)) are presented in Table 5.

From Table 5 one can see that, in all cases, the %GR goes beyond 80%. Even, at experimental conditions corresponding to test number 3, the %GR reaches

**Table 6** | ANOVA for grease and oil removal. Results for the second group of variables

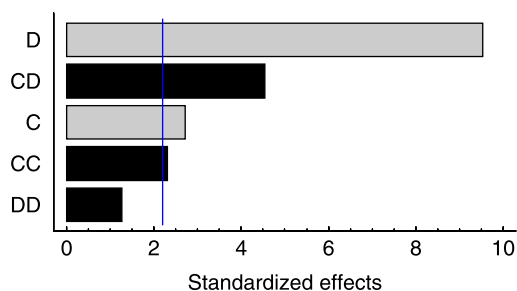
Factor	Square sum	Freedom degrees	Square average	f ratio	p value
D: gap between electrodes	6.00668	1	6.00668	7.26	0.0209
C: current density	75.0	1	75.0	90.62	0.0000
DD	4.34722	1	4.34722	5.25	0.0426
CD	16.9944	1	16.9944	20.53	0.0009
CC	1.2996	1	1.2996	1.57	0.2362
Blocks	0.00245	1	0.00245	0.0	0.9576
Total error	9.10425	11	0.827659		
$R^2 = 91.93\%$ ; $R^2$ adjusted = 89.59%					



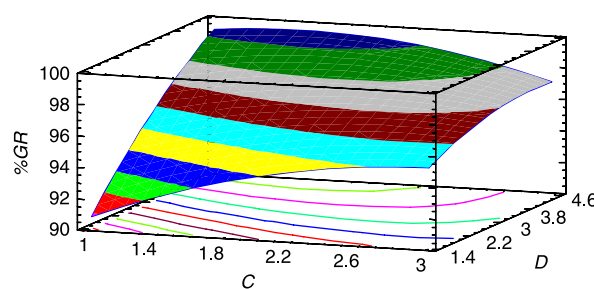
the c.a. 99%. The ANOVA proves that analyzed variables are significant ( $p$  value below 0.05) and influence the EC process.

The values obtained for the statistical parameters presented in Table 6 suggest that the factors A (gap between electrodes), B (current density), and their interactions (AA and AB) are significant for the EC process. Additionally, the Pareto diagram (Figure 3) reveals the %GR is directly influenced by the factors A and B but in a negative way by their interactions.

The maximum %GR is reached when the gap between the electrodes is c.a. 1.5 cm (as obtained from Statgraphics). It is interesting to notice from Table 5 that further increases in the gap between electrodes implies a decrease in the %GR, which could be attributed to the lower electrostatic: the movement of the ions decreases, reducing the coagulation and flocculation capacity of the EC cell, as also was observed by Song *et al.* (2008). The effect of the current density on the %GR is quite straightforward (Table 5). The higher current density, the higher %GR is observed. This could be related to the fact that high current density is responsible of high Fe ions concentration in solution. Those ions are known to in charge of contaminants trapping, which lead to the increase in the efficiency of the EC process. This was also reported in the literature by other authors Feng *et al.* (2003), Canizares *et al.* (2007), Heidmann & Calmano (2008), Song *et al.* (2008) and Yilmaz *et al.* (2008). Figure 4 summarizes the effect of the current density and the gap between electrodes in a surface response plot. The quadratic regression model allows obtaining the Equation (12), as a function of factors C and D (see Table 6), which represents the surface as a function



**Figure 3** | Pareto diagram for the EC process for the second group of factors: D: gap between electrodes and C: current density.



**Figure 4** | Surface response plot for the %GR as a function of the two selected factors: the gap between electrodes (C) and current density (D).

of both factors: the current density and the gap between electrodes and their interactions.

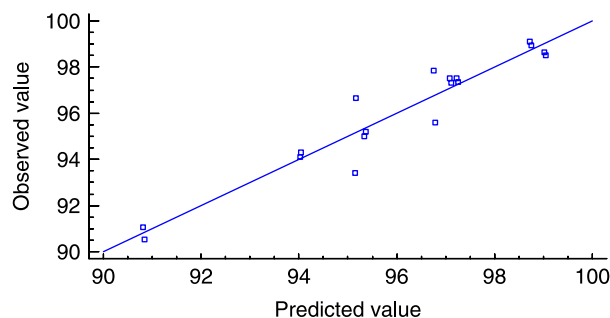
$$\%GR = 78.673 + 7.7422C + 5.2798D - 1.0425C^2 - 1.0052C \cdot D - 0.2711D^2 \quad (12)$$

### Effect of H<sub>2</sub>O<sub>2</sub> dose

The %GR is properly adjusted by the second order regression model ( $R^2 = 0.9193$ ). This can be compared from Figure 5 which includes both the observed and predicted values.

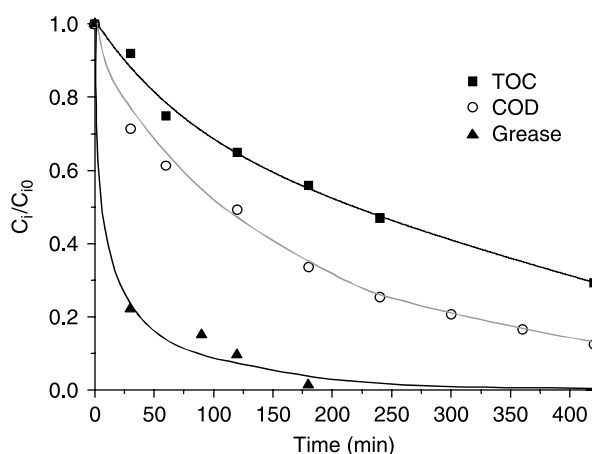
### Kinetics studies

At optimal parameter values (pH 12, “sacrificed” anode: Fe, gap between electrodes: 1.5 cm, and current density: 4.3 mA/cm<sup>2</sup>) a kinetic study was developed. The evolution of the Chemical Oxygen Demand (COD), Total Organic Carbon (TOC), and oil and grease removal (grease), was monitored as a function of time. Experiments were performed during 7 hours. Figure 6 presents the TOC, COD, and greases evolution as a function of time.



**Figure 5** | Comparison of observed and predicted values for the %GR for the second group of factors.

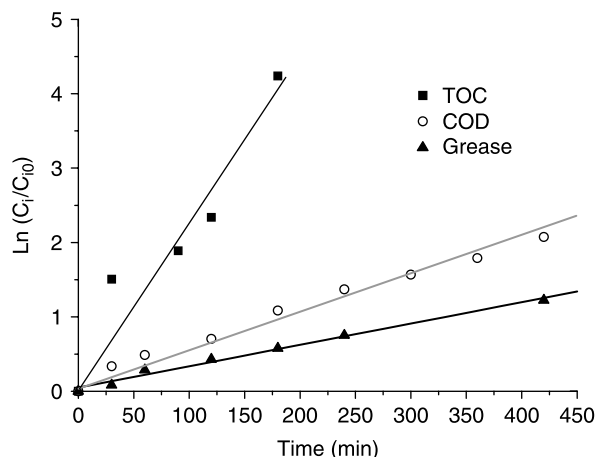




**Figure 6** | Plot of TOC, COD, and grease adimensional concentration as a function of time during the EC process.

It is possible to observe that during the first 30 minutes of electrolysis, the %GR decreases rapidly down in c.a 78% (Figure 6). It could be related to the fact that when the current is applied to the emulsion by the electrodes, the anode dissolution generates metallic ions (in this case  $\text{Fe}^{2+}$ ). This coagulant reduces the superficial charge of oil drops, breaking the emulsion. Finally, by coalescence, larger oil drops are formed and trapped by  $\text{Fe}(\text{OH})_2$  dispersed in the solution. This creates large flocs which in turn are easily joined by the gas bubbles participating in the EC process. At the end of the treatment process, a proper separation of the oil in water emulsion is made; and the organic phase is removed by simple decantation or flotation.

Different models were analyzed as proposed by Fogler (2006). Data adjusts properly to a pseudo-first order model



**Figure 7** | Linearizing plot of TOC, COD, and grease data.

with kinetic constant values of 0.0031, 0.0052, and  $0.0226 \text{ min}^{-1}$  for TOC, COD, and greases, respectively.

Figure 7 presents the obtained results.

The kinetic results notably agree with the regression model. It is interesting to notice that, after 7 hours of electrolysis, the COD and TOC decreases to 87.45 and 70.6%, respectively, and the %GR reaches the 98.56%. A final biodegradability ( $\text{BOD}_5/\text{COD}$ ) of 0.54 was reached.

## CONCLUSIONS

At the studied conditions, it was proved that the electrocoagulation process is useful for the treatment oily wastewater resulting from an automotive industry placed in Medellin (Colombia). After a statistical analysis, the optimal values of the chosen parameters were: current density:  $4.3 \text{ mA/cm}^2$ , pH: 12, Fe as the “sacrificed” electrode, gap between electrodes: 1.5 cm. For an electrolysis period of 7 hours, the COD and TOC decreases to 87.45 and 70.6%, respectively, and the %GR reaches the 98.56%. This allows fulfilling the minimal Colombian regulations for oil and greases present in wastewaters. The electrocoagulation process highly recuperates the wastewater. It can be estimated its reprocess in c.a. 90%, which can decrease the consumption of this important natural supply. The scale up of this study is now in progress.

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