

# PASSIVATING EFFECT OF WASTEWATER ON IRON ELECTROCOAGULATION TREATMENT EFFICIENCY

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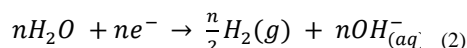
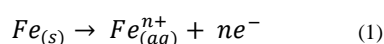
## Abstract

Iron electrocoagulation (Fe-EC) is a cost-effective water/wastewater treatment method that has demonstrated effectiveness in the remediation of many forms of polluted water. This technique utilizes a Fe sacrificial anode to produce coagulant in-situ, hence offering several advantages over conventional iron-based chemical coagulation procedures. However, the occurrence of solid precipitation on the electrodes during operation results in a passivating effect that has been reported to reduce treatment effectiveness and increase energy consumption. The objective of this investigation was to evaluate the passivation effect of oily wastewater (OW) and chicken slaughterhouse wastewater (CSW) on Fe anode, specifically in relation to the Faradaic efficiency (FE), using initial pH (4, 7, and 9) and applied current intensity (0.1, 0.2, and 0.4 A) as variables. The study also sought to assess the potential passivating impact of the wastewaters on the efficacy of Fe-EC in the removal of chemical oxygen demand (COD) from the wastewaters. The results revealed that the mean passivating effect was 13.4%, 7.67%, and 22.7% for OW and 22.3%, 17.13%, and 12.3% for CSW at 0.1 A, 0.2 A, and 0.4 A, respectively. FE ranged from 0.77 to 0.92 and 0.78 to 0.88 for OW and CSW, respectively. The initial pH of the wastewater was observed to significantly influence the FE at the given applied currents. While the mean COD removal from both wastewaters was greater than 83%, there was a lack of correlation between the FE and COD removal. This finding suggests that the mass of the coagulant is one of several potential COD removal factors. Further research includes electroanalytical studies of Fe corrosion in various wastewaters.

**Keywords:** Electrocoagulation, chemical oxygen demand, Faradaic efficiency, passivation

## 1. Introduction

Iron electrocoagulation (Fe-EC) is an electrochemical technique for removing contaminants from water and wastewater using Fe anode. The technique involves generating coagulants in-situ from the sacrificial Fe anode by passing an electric current through a pair(s) of connected anode-cathode electrodes immersed in wastewater. The reactions that occur at the anode and cathode are expressed in equations 1 and 2, respectively:



where  $n$  is the number of electrons involved in charge transfer.

The ensuing Fe coagulants can eliminate pollutants through one or more of the following mechanisms: adsorption, complexation, enmeshment, precipitation, and co-precipitation [1-3]. Fe-EC has been proven effective at reducing the concentration of a variety of pollutants found in wastewater generated from the food processing industry [4-6], the textile and tanning industry [7-9], the petroleum refinery industry [10-12], the pulp and paper industry [13-15], and the pharmaceutical industry [16-18], among others. The EC technique is characterized by simple equipment, easy operation, no or minimal chemical addition, and a decreased amount of produced sludge [1, 19]. These attributes make EC an attractive alternative to conventional methods of treating water and wastewater.

As an electrochemical method, the efficiency

of EC is closely linked to the reactions occurring at the surface of the electrodes and the solution chemistry [20]. If the reactions at the anode's surface result in the formation of a metal oxide film that reduces the discharge of ions from the anode, the quantity of coagulants is reduced, and consequently, pollutant removal is diminished [21-23]. The formation of a charge-insulating film on the anode is called passivation. Passivation reduces the effective surface area of the electrodes and leads to increased energy consumption in the EC cell [24-26]. Visibly, the occurrence of passivation is by way of solid deposits on the surface of the anode. Quantitatively, the effect of passivation can be assessed by determining the Faradaic efficiency (FE). This is done by measuring experimentally the mass ( $m$ ) of iron dissolved in an EC system and comparing this mass to the theoretical mass ( $m_{Th}$ ) obtained by Faraday's law (Eq. 3). The resulting value is the FE (Eq. 4), also called current efficiency.

$$m_{Th} = \frac{ItM_w}{nF} \quad (3)$$

$$FE = \frac{m}{(m_{Th})} = \frac{nFm}{ItM_w} \quad (4)$$

where  $m$  and  $m_{Th}$  are measured in grams (g),  $I$  is the current (A),  $t$  is the time of operation (s),  $M_w$  is the molecular weight of the anode material, and  $F$  is Faraday's constant (96,485 C/mol). Apart from pollutant reduction, FE is also used to evaluate the performance of EC [23]. Ideally, an FE of 1 indicates no passivation, and values close to unity are preferred in the EC treatment of wastewater.

Several studies, including those by [22, 27-30] have sought to identify the iron speciation generated during Fe-EC. While this knowledge is important to advance the Fe-EC technology, [31] advise that researchers in the field of EC should not solely focus on the chemical properties of the coagulants. Rather, they should also consider the influence of the solution on the operational aspects of the EC system. Therefore, this present study investigated the passivating influence of real wastewaters on the EC process in an iron-based system, specifically in terms of reducing chemical oxygen demand (COD). The study aimed to quantify the passivating effect on Faradaic efficiency and COD reduction when oily wastewater (OW) and chicken slaughterhouse wastewater (CSW) are in direct contact with the iron anode. Also considered are the effects of different levels of applied current and initial pH on the passivating effect of the wastewaters. This research contributes to the existing scientific literature on the passivation of Fe in corrosive wastewater environments, its correlation with the amount of Fe produced, and the ensuing removal of COD from industrial effluents.

## 2. Materials and Methods

### 2.1 Materials

The wastewater samples used in this study were obtained from a vegetable oil processing facility in Kingston, Jamaica, and a chicken slaughterhouse in Saint Catherine, Jamaica. Samples were collected in sterilized bottles and transported at a temperature of  $30 \pm 2^\circ\text{C}$ . The wastewaters were analyzed for COD, pH, total dissolved solids (TDS), total suspended solids (TSS), phosphate, nitrate, sulphate and Fe content in accordance with Standard Methods [32].

All chemicals used in the study were of reagent grade. Sodium chloride (NaCl) was used as the supporting electrolyte, and sodium hydroxide (NaOH) and hydrochloric acid (HCl) were used to adjust the pH of the wastewater. Double-distilled water was used in the making of solutions and the rinsing of glassware and tools.

### 2.2 Experimental setup and procedure

A bench-scale EC reactor made from acrylic sheet was used in the experiments. The reactor had a 1-L working volume and was operated in batch mode. The electrodes were mild steel rods with effective surface areas of  $13.36\text{ cm}^2$  and were positioned 3 cm apart and 4 cm above the base of the reactor. Direct current (DC) was supplied by a Sencore PS402 Triple Output (0 - 30 V, 3 A) power supply. The reactor was placed on a magnetic stirrer (Corning PC - 4100) used to agitate the contents of the reactor. A schematic diagram of the EC setup is shown in Fig. 1.

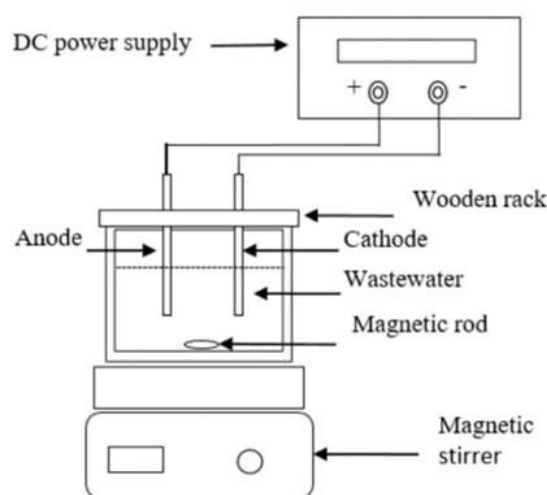


Figure 1. Schematic diagram of the EC setup

Prior to EC treatment, the wastewater was removed from storage and allowed to reach ambient temperature. The content of the reactor was either a 1-L mixture of 100 mL of 1:10 diluted wastewater, 2.93 g of NaCl, and deionized water, or 1 L of a 2.93 g/L NaCl solution. The factors varied were initial pH (4, 7, and 9) and applied current (0.1, 0.2, and 0.4 A). The initial pH of the solution was adjusted to the desired value using either 0.1 M HCl or 0.1 M NaOH. The electrodes were submerged in the solution and connected to the power supply. The power was switched on, and electrolysis was carried out under galvanostatic currents for 8 minutes and 38 seconds to generate a theoretical Fe(II) mass of 15 mg, 30 mg, and 60 mg, corresponding to currents of 0.1, 0.2, and 0.4 A, respectively. Simultaneously, the reactor's contents were agitated with the magnetic rod and stirrer at 300 rpm. At the end of electrolysis time, the electrodes were quickly removed from the reactor, the power supply switched off, and two 2-mL samples were withdrawn from the reactor for Fe content analysis. Following this, the content of the reactor was allowed to undergo flocculation at a mixing speed of 60 rpm for an additional 12 minutes.

At the end of treatment, a 10 mL sample was taken, filtered, and the filtrate analyzed for COD. The experiments were carried out at a temperature of  $30 \pm 2^\circ\text{C}$  and were replicated three times, resulting in a total of 27 runs. Prior to each run, the electrodes were soaked in 1 M HCl for 10 minutes, rinsed with distilled water, scrubbed with steel wool, re-rinsed with distilled water, and wiped clean with a paper towel to remove all residue.

### 2.3 Analytical procedures

Total Fe concentration was determined by flame atomic absorption spectrophotometry (F-AAS, Agilent 240FS, SMEWW 3111). The determined Fe values were adjusted by subtracting the initial Fe content of OW and CSW. The COD concentration was determined using HACH reagents and a HACH DR 6000 UV-VIS

spectrophotometer at a detection wavelength of 620 nm. Solution pH was measured using a HACH HQ440d multi-parameter meter. The removal efficiency of COD was calculated using the following equation:

$$COD\ removal\ (\%) = \frac{COD_i - COD_t}{COD_i} \times 100\% \quad (5)$$

where  $COD_i$  and  $COD_t$  are the initial COD concentration and the COD concentration at time  $t$ , respectively.

### 3. Results and Discussion

#### 3.1 Wastewater characteristics

Table 1 presents the characteristics of the wastewater samples. The initial mean COD concentrations of OW and CSW were comparable, at 3400 mg/L and 3500 mg/L, respectively, allowing for a comparison of the efficacy of Fe-EC in removing COD from the two effluents.

Table 1  
Characteristics of OW and CSW

Parameter	Value (mean)	
	OW	CSW
COD (mg/L)	3400	3500
pH	8.71	6.55
Phosphate (mg/L)	60	50
Nitrate (mg/L)	120	67.5
Sulphate (mg/L)	nd*	100
TSS (mg/L)	1000	2000
TDS (mg/L)	970	1330
Fe (mg/L)	0.63	4.73

\* nd – not detected

#### 3.2 Total Fe content analysis

Table 2 provides a comparison of the quantity of total Fe dissolved from the anode in the NaCl solution and the wastewaters. The results indicate that the measured quantities of Fe were less than the theoretical masses if  $n$  is assumed to be 2. The measured masses also vary according to the type of solution and the applied current. The passivating effect, which refers to the decrease in Fe dissolution, was observed to be 9%, 17.5%, and 11.4% for NaCl at current values of 0.1, 0.2, and 0.4 A, respectively. Similarly, for OW, the passivating effect was found to be 13.4%, 7.67%, and 22.7%, while for CSW, it was measured to be 22.3%, 17.13%, and 12.3% at the same current values. Fig. 2 shows how the results in Table 2 compare with the theoretical masses of Fe(II) and

Fe(III). The findings suggest that the masses tend to exhibit a closer proximity to the Fe(II) line compared to the Fe(III) line, therefore providing support for the claim made by [33-36] that Fe(II) is the species released from the anode.

Table 2  
Comparison of Total Fe Content

I (A)	No. of runs	$m_{Th}$ Fe(II) (mg/L)	Experimental mass (mg/L)					
			0.05M NaCl		OW		CSW	
			M	S.D.	M	S.D.	M	S.D.
0.1	9	15	13.64	1.22	12.99	0.10	11.66	1.44
0.2	9	30	24.74	1.93	27.70	6.67	24.86	1.16
0.4	9	60	53.19	2.01	46.37	3.18	52.65	2.15

M – mean; S.D. – standard deviation

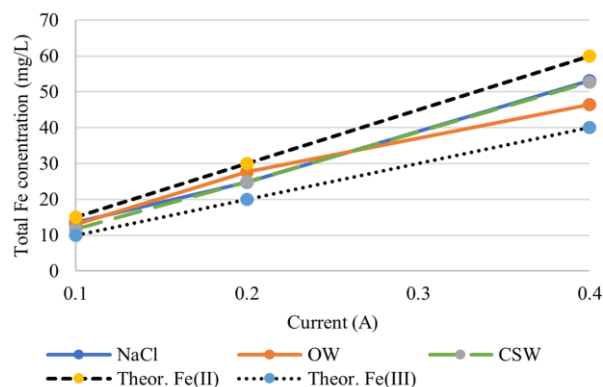


Figure 2. Fe content analysis comparison

However, at the higher current of 0.4 A, the mass of OW is in closer proximity to the Fe(III) line. This occurrence can be attributed to the increased passivation of the anode, as seen in Fig. 3(c). Among the three solutions, it was observed that the mass of Fe liberated in the NaCl solution was generally higher. This finding is similar to that of [34] who reported a greater Fe release from the anode in the NaCl solution than in pretreated wastewater. The presence of chloride ions facilitates the process of anode dissolution by pitting corrosion. Conversely, the presence of oxyanions, such as nitrates, phosphates, or sulfates, in wastewater has a comparatively less favorable impact on anode dissolution [22, 24, 38, 39].

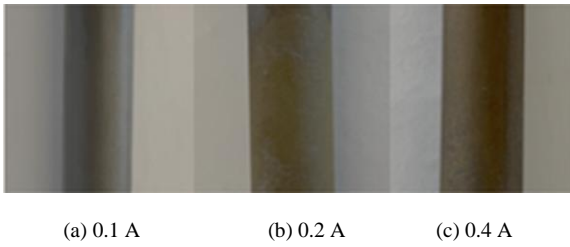


Figure 3. Surface layer buildup on the Fe anode in OW at initial pH 9

### 3.3 Effect of applied current on Faradaic efficiency

Fig 4 displays the mean FE obtained for the solutions as a function of applied current, compared to the ideal FE of 1. The values of mean FE ranged from 0.82 to 0.91 for NaCl solution, 0.77 to 0.92 for OW, and 0.78 to 0.87 for CSW. The FE values are consistent with the findings presented in Table 2 and Fig. 2. FE values have been found to be relatively high for Fe-EC and independent of applied current [35, 38], particularly in the presence of Cl<sup>-</sup> ions, which promote pitting corrosion [27, 37, 39]. In the presence of higher currents, FE can be reduced [27, 40] because these conditions may facilitate oxygen evolution [27, 38]. Fig. 4 further highlights an abnormality that was detected at 0.2 A. At this current value, the lowest FE was seen for NaCl, whereas the maximum FE was observed for OW. Furthermore, there was a noticeable alteration in the slope of the FE plot for CSW at 0.2 A. The observed behaviour at 0.2 A for each plot may be attributed to a complex interaction between solution chemistry and electrode processes, which requires further investigation.

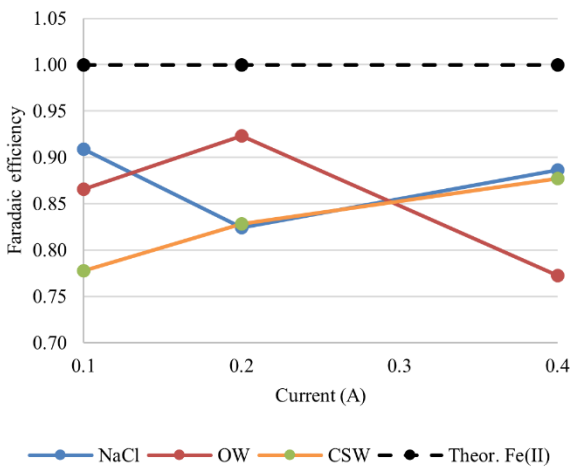


Figure 4. Mean Faradaic efficiency for Fe anode in NaCl, OW and CSW as a function of current

### 3.4 Effect of initial pH on Faradaic efficiency

The effect of initial pH on FE was investigated at currents of 0.1, 0.2, and 0.4 A, and the results are displayed in Fig. 5.

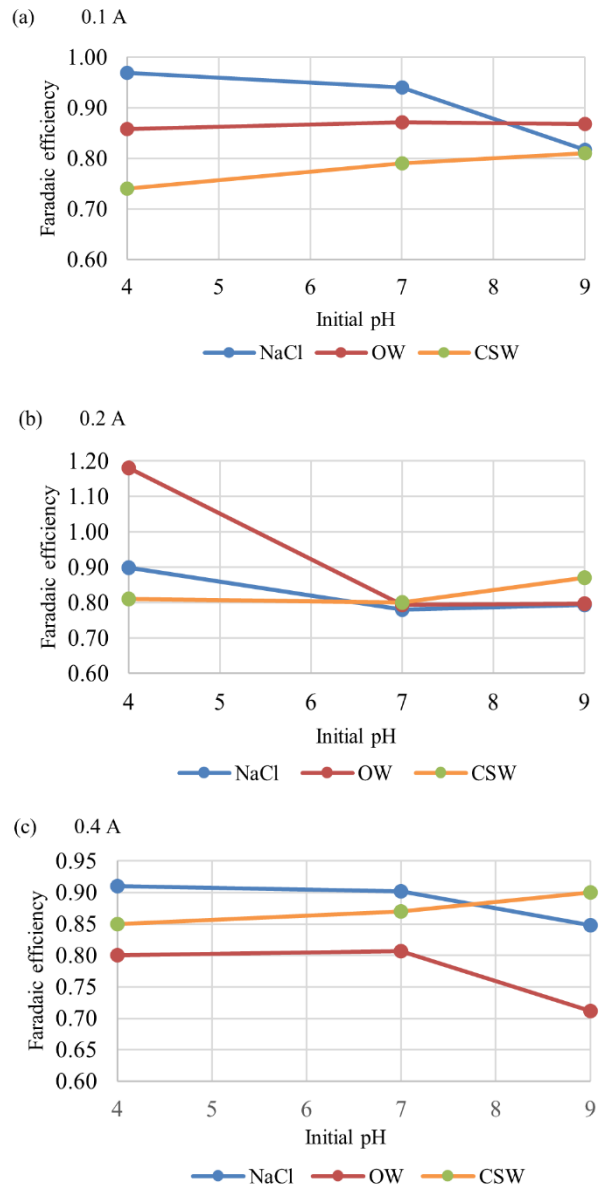


Figure 5. Effect of initial pH on Faradaic efficiency at: (a) 0.1 A, (b) 0.2 A, and (c) 0.4 A

As seen in Fig. 5, initial pH has a significant effect on FE. At acidic and alkaline initial pH values, FE is high and low, respectively [1, 36, 37]. Except for CSW, where there was a minor increase in FE as the initial pH increased at all current levels, the FE for NaCl and OW remained generally stable or exhibited little change when the initial pH rose from 4 to 7 for currents of 0.1 and 0.4 A. At 0.1 A, it was found that increasing the initial pH from 7 to 9 resulted in a noticeable decrease in the FE for NaCl, whereas no apparent change was observed for OW. For both solutions, a similar increase in initial pH at 0.4 A resulted in a decline in FE. In contrast, at 0.2 A, a decline in FE was observed when the initial pH increased

from 4 to 7. No further decrease was evident beyond an initial pH of 7.

The types of surface films generated on the anode due to pH-mediated reactions among Fe coagulants, dissolved oxygen, and the constituents of the solution may explain the various FE outcomes as a function of initial pH. The pH-influenced Fe(II/III) speciation can lead to a variety of oxyhydroxides on the anode that have differing passivation properties [21]. A porous surface film is less passivating than a nonporous one due to its ability to still transfer charge [23]. The decrease in FE at elevated pH levels, as found in the presence of NaCl and OW, might be attributed to the development of passivating films consisting of Fe(III)oxyhydroxides. These films hinder the flow of electrons across the anode and the release of iron from the anode [37].

Also at 0.2 A, a FE value of 1.18 was observed at initial pH 4 for OW, which indicates a non-electrochemical contribution to the Fe released from the anode [36, 39].

### 3.5 COD removal from wastewater

Relatively high COD removals of greater than 85% were achieved for both OW and CSW. In Fig. 6, which displays the mean COD removal efficiencies at the applied currents and the associated mean FE, there is a positive correlation between FE and COD removal from the two wastewaters between the currents 0.1 and 0.2 A. The increase in FE with increasing current indicated that a larger quantity of Fe coagulant was available to facilitate the removal of COD, as a higher coagulant concentration traps more pollutants. This finding is consistent with those of [41] and [42]. At 0.4 A, which is anticipated to yield the highest quantity of dissolved Fe, the removal of COD does not align with the governing FE. A greater quantity of COD removal from OW was observed in comparison to what was indicated by the FE. Conversely, a lesser quantity of COD was removed from CSW, despite the FE exhibiting the highest value at 0.4A. Previous research conducted by [43 - 45], has demonstrated that the relationship between current intensity and pollution removal is not linear. These and other studies have revealed that there exists an optimal current value beyond which the effectiveness of pollution removal diminishes, indicating that an increase in current does not necessarily result in higher levels of pollution removal. The findings of the study highlight two main points: (a) a minimum quantity of Fe may be effective in reducing COD, and (b) there are other factors that influence COD removal beyond the amount of coagulant utilized.

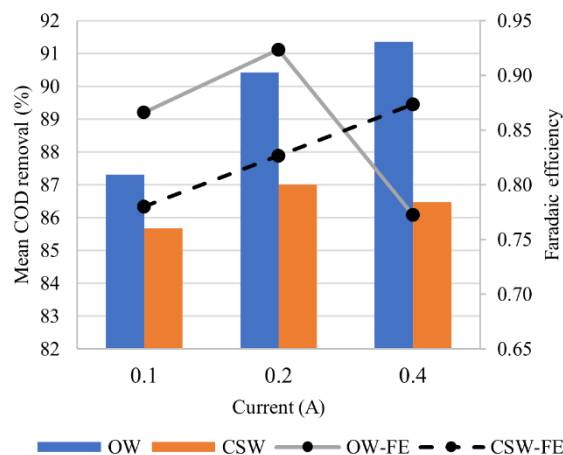


Figure 6. Effect of current on COD removal from OW and CSW with Faradaic efficiency

Fig. 7 shows the effect of pH on COD removal from the two wastewaters. According to the literature, the pH governs the speciation of the Fe coagulant.

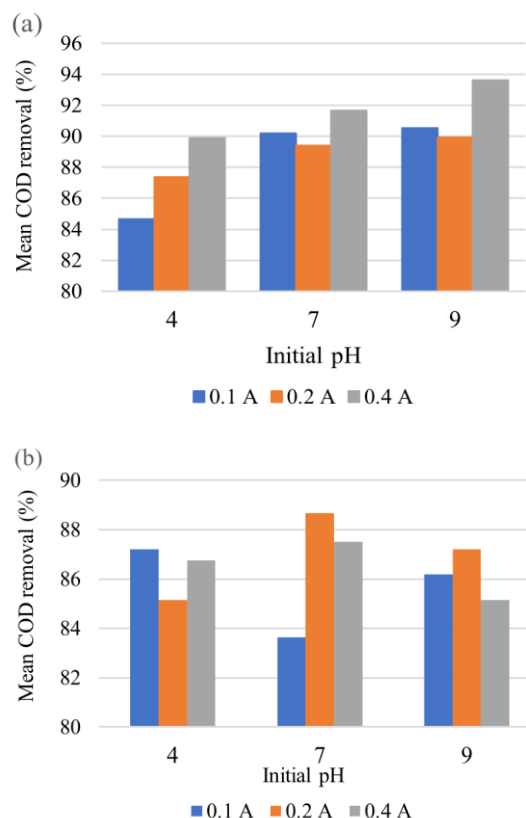


Figure 7. Effect of initial pH on COD removal (a) OW, and (b) CSW

Examining Fig. 7(a) reveals that COD removal increases as the initial pH increases for OW. The insoluble  $\text{Fe}(\text{OH})_2$  and  $\text{Fe}(\text{OH})_3$  are the

coagulating species that are purported to be present at acidic and alkaline pH values, respectively [42, 46]. At a neutral pH, a mixture of the two hydroxides can occur. The  $\text{Fe}(\text{OH})_3$  species, according to [47], is said to have high affinity for suspended and colloidal particles; hence, this species presence may be responsible for the relatively high removals observed at initial pH 7 and 9. Previous studies conducted by [41] and [48] and have also documented comparable outcomes in terms of high COD removal from OW, but primarily at an initial pH of 9. In the case of CSW, initial pH 7 resulted in the highest mean removal, followed by initial pH 4. High COD removals greater than 85% from CSW were also reported by [5], [49] and [50] at near neutral pH values. This suggests that  $\text{Fe}(\text{OH})_2$  may be the favored coagulant for effectively reducing COD levels in this wastewater sample.

In terms of the passivating effect of OW and CSW, there is no clear correlation between COD removal and FE. Examination of the FEs presented in the plots displayed in Fig. 5 in conjunction with the corresponding COD removals illustrated in Fig. 7(a) and 7(b) revealed a mismatch between the FEs and COD removal. High FEs ( $>0.80$ ) do not necessarily correspond to high COD removal rates, and conversely, low FEs ( $<0.80$ ) do not necessarily result in low COD removal rates. These findings indicate that significant COD reductions were achieved across the applied initial pH and current combinations for both wastewaters. Hence, it can be inferred that factors other than the kind and quantity of the coagulant may also play a role in the removal of COD from wastewater.

Based on similar COD content, the results clearly show that higher COD removals were obtained from OW than from CSW under the same conditions of initial pH and applied current. Hence, the effectiveness of Fe-EC treatment is influenced by the types of constituents in the wastewater that contribute to COD.

#### 4. Conclusion

In this study, the passivating effect of real and untreated wastewater (OW and CSW) on Fe-EC was investigated. The findings revealed that there were differences among the masses of Fe generated in the wastewaters and a 0.05M NaCl solution at similar conditions of initial pH (4, 7, and 9) and applied current (0.1, 0.2, and 0.4 A). The reduction in the mass of Fe generated from the anode in the wastewater samples was generally greater than the mass generated in the NaCl solution, therefore indicating that the wastewaters had a greater passivating effect. While the resulting FEs ( $> 0.70$ ) were relatively high for both wastewaters, the initial pH was found to have a great influence on the mass of Fe

dissolved from the anode at the applied currents. High COD removal efficiencies greater than 80% were observed for both wastewaters across all combinations of initial pH and applied current. A lack of meaningful correlation between FE and COD removal indicated that other factors, apart from the mass of the coagulant, may contribute to pollutant removal by Fe-EC. This finding reveals the potential error that exists when quantitative models for COD removal are based on the theoretical mass of Fe generated at the anode. Potential areas of investigation for future research include the examination of the relationship between the chemical composition of wastewaters and the passivation of the anode, as well as the utilization of electroanalytical techniques to better understand the underlying electrode processes that contribute to the corrosion behavior of iron in varying types of wastewaters.

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