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Rachmad Ardhianto

rachmad@itpln.ac.id

Institut Teknologi PLN

Valencia Safir Maharani

Institut Teknologi PLN

Nadia Paramita

Institut Teknologi PLN

Bening Laksa Intan

Soegijapranata Catholic University, Central Java

Eqiu Alkahfi Tarif

Institut Teknologi PLN

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Integrated electrocoagulation and clarifier air flotation (EC-CAF) as a pretreatment method for wastewater treatment in the used oil refinery industry: a review of full-scale applications

Rachmad Ardhianto[1], Valencia Safir Maharani[1], Nadia Paramita[1], Bening Laksa Intan[2], Eqiu Alkahfi Tarif[1]

[1]Department of Environmental Engineering, Faculty of Infrastructure and Regional Technology, Institut Teknologi PLN, Jakarta, Indonesia 11750, Indonesia

[2]Study program of Environmental Infrastructure Engineering, Faculty of Environmental Science and Technology, Soegijapranata Catholic University, Semarang 50234, Central Java, Indonesia

*Corresponding Author: rachmad@itpln.ac.id

Abstract

This study assessed a full-scale electrocoagulation–chemical air flotation (EC–CAF) system for treating wastewater from used oil refining processes, which are characterised by high non-biodegradable pollutant concentrations. The system showed excellent removal of total suspended solids (TSS), with efficiencies ranging from 86.8% to 90.7% (averaging 88.6%). This performance resulted from the effective formation and flotation of iron hydroxide flocs. However, the removal of dissolved pollutants was limited, with average efficiencies of 12.5%, 7.6%, and 13.3% for chemical oxygen demand (COD), ammonia nitrogen (NH_3-N), and 13.3% for sulphate (SO_4^{2-}). These low rates are due to recalcitrant organic compounds and dissolved ionic species that were not effectively removed through coagulation–flotation mechanisms. Increasing the electrical input (2.06–3.60 kWh/m³) and current density enhanced floc formation and TSS removal but minimally affected dissolved contaminants. The Faraday efficiency ranged from 53% to 66%, indicating nonideal electrochemical conditions resulting from side reactions and electrode passivation. Sludge characterisation revealed amorphous iron-based flocs with a high adsorption capacity for PM. Although the EC–CAF system is effective as a pretreatment, it requires integration with advanced oxidation or biological processes to achieve comprehensive removal of dissolved pollutants in nonbiodegradable wastewater.

1. Introduction

The used oil refining sector is a high-risk industry characterised by generating wastewater from refining residuals. Wastewater generated from refining used oil generally has intricate properties, comprising a blend of organic and inorganic chemicals. The primary constituents of this effluent are dissolved oil and aromatic chemicals, including benzene, toluene, and xylene (BTX) [1],[2]. Moreover, the concentrations of phenol, suspended particles, and sulphate ions (SO_4^{2-}) are elevated. The significant concentration of SO_4^{2-} may arise from chemical purification operations, including reactions with sodium hydroxide (NaOH) to eliminate hydrogen sulphide (H_2S), yielding a caustic sulphide solution abundant in sulfur compounds [3]. This category of wastewater also has elevated concentrations of heavy metals, ammonia, and total organic carbon [4], ammonia, and high TOC [[5]. This industrial effluent exhibits an exceedingly high chemical oxygen demand (COD) value ranging from 3500 to 5300 mg/L [[6].

The waste produced originates from oil sources and contains hazardous materials, presenting a considerable environmental concern; therefore, ensuring appropriate treatment and adherence to the current standards prior to disposal is essential. Consequently, numerous processing techniques have been employed and recorded in the scientific literature to manage this type of waste. While several current technologies demonstrate efficiency during the processing phase in terms of cost and energy usage, others have difficulties associated with elevated energy and maintenance expenses, rendering them less environmentally sustainable. Consequently, evaluating a method's energy efficiency, capacity to handle diverse pollutants, and volume of trash produced as a by-product is essential for the advancement and application of any treatment system [7]. The management of this type of waste is typically categorised into two principal phases: a pretreatment phase aimed at diminishing pollutant concentrations, including oil, grease, and dissolved solids, and a phase in which pollutants are converted to a level suitable for safe discharge [8],[9]. Reported treatment strategies in the literature encompass biological processes [5][10][11]. Coagulation processes [12],[13], and advanced oxidation processes [[14],[15].

A comprehensive review of electrocoagulation across industrial applications reveals that development at the industrial level is minimal, with most research concentrated at the laboratory scale..Most published studies have focused on batch-scale laboratory experiments with synthetic wastewater conducted under controlled conditions, which fail to capture the complexities of real-world industrial operations in which influent characteristics fluctuate and operational periods extend over months or years. Long-term operational performance data are essential for sustainability and commercial viability assessment. Electrocoagulation is the most studied method at the laboratory scale; however, its

development at an industrial level remains minimal, particularly for oil refinery applications [16] One of the most critical research gaps is the lack of comprehensive data from full-scale electrocoagulation installations treating wastewater from used oil refineries..A recent comprehensive review of combined electrocoagulation processes emphasised the need for pilot-scale investigations under continuous flow conditions and highlighted that challenges, including limited pilot- and full-scale applications, persist [17].[17].This substantial gap between batch studies and industrial operations makes predicting lifecycle performance and developing realistic maintenance schedules difficult..

Energy consumption is a critical determinant of commercial viability; however, substantial variations exist across studies.In batch laboratory settings, a combined electrocoagulation-electrooxidation process for treating vegetable oil refinery wastewater required a specific energy consumption of 0.513 kWh/kg COD for the EC stage alone, which increased to 2.067 kWh/kg COD when combined with electrooxidation [18] Similarly, a combined photo-electrocoagulation system treating petroleum refinery wastewater achieved energy consumption of 28.44 kWh/kg COD [19] However, these laboratory-optimised values may not reflect actual industrial-scale demands operating under variable wastewater characteristics..

A significant research gap exists in the comprehensive economic analysis of full-scale electrocoagulation as a pretreatment step. Historical reviews of the operating costs of electrocoagulation reveal tremendous variation, ranging from 0.435 to 2.87 $/m³ in pilot-scale and full-scale applications of electrocoagulation in the textile and electroplating industries [20],[21], respectively, making it to establish reliable cost projections for industrial implementation. However, scholarly investigations into the use of electrocoagulation for treating wastewater resulting from used-oil purification are scarce. The cost–benefit analysis comparing electrocoagulation with conventional methods, such as dissolved air flotation and coagulation–flocculation, remains inadequately developed specifically for used-oil refinery wastewater. Although general contaminant removal has been extensively studied, the treatment of specific problematic pollutants in used-oil refinery wastewater via electrocoagulation remains inadequate. Waste lubricating oil regeneration wastewater contains sulphate (SO_4) 895 mg/L and COD 1226.8 mg/L [22]. Research specifically examining the application of electrocoagulation for these contaminants remains scarce, particularly concerning its industrial-scale implementation. The behaviour of electrocoagulation in treating complex multi contaminant mixtures, which are prevalent in industrial wastewater, significantly differs from that in single or binary contaminant systems typically investigated in laboratory settings. However, such studies remain fragmented.

Based on this explanation, this study employed EC and clarifier air flotation as alternatives to the traditional chemical coagulation method in a wastewater pretreatment system for the full-scale treatment of effluents from used oil refining operations. This study comprehensively reviews the concurrent implementation of EC and clarifier air flotation over a continuous three-month period in a full-scale setting. The efficacy of the system in removing total suspended solids (TSS), chemical oxygen demand (COD), ammonia, and sulphate (SO4) from wastewater produced during used oil purification was evaluated. Furthermore, critical operational parameters, such as current density, power consumption, electrode replacement, electrode wear, and COD/SO_4 ratio, were thoroughly examined to provide a direct assessment of the system's performance. This analysis aims to provide valuable insights into wastewater treatment and address previously identified research gaps.

2. Materials and methods used

2.1. Used oil refinery wastewater characteristicsc

This study examined wastewater effluents from the used-oil processing industry, which is characterised by elevated concentrations of organic contaminants, including chemical oxygen demand (COD), ammonia (NH_3-N), and sulphate. The wastewater was distinguished by a high COD content and low biochemical oxygen demand (BOD), indicating its nonbiodegradable nature, as evidenced by the COD/SO_4 ratio values presented in **Table 1**. These pollutants pose significant challenges in wastewater treatment, particularly during electrocoagulation, in which sulphate can impede floc formation. The wastewater produced from solvent extraction and centrifugation processes is rich in organic substances and inorganic ions, including sulphates. Before treatment, the wastewater was evaluated based on its physicochemical characteristics, including pH, COD, TSS, NH_3-N, Cl, and sulphate concentrations. The COD/SO_4 ratio was meticulously analysed to assess its impact on the efficacy of EC. Understanding the interaction between organic solubility and sulphate levels is crucial for enhancing electrocoagulation efficiency.

Table 1. Characteristics of the used oil wastewater

No	Parameters	Unit	Value
1	pH	-	6.93 ±1.33
2	Total dissolved solids (TDS)	mg/L	864.90 ±76.83
3	Total Suspended Solid	mg/L	380.52 ±112.82
4	Biochemical oxygen demand (BOD)	mg/L	674.16 ±333.46

No	Parameters	Unit	Value
5	Chemical Oxygen Demand	mg/L	3856.61 ±1628.18
6	Sulphate (SO_4)	mg/l	379.33 ±76.81
7	Ammonia (NH_3-N)	mg/l	114.10 ±26.90
8	Clorin (Cl^-)	mg/l	206.80 ±174.08
9	COD/SO_4	-	10.21 ±0.79
10	BOD/COD	-	0.174 ±0.2

105

106 2.2. Reactor configuration

107 This pretreatment process is implemented to complement the existing system, which utilizes a biological method
108 based on activated sludge, as illustrated in **Fig 1.** The system employed in this study comprised an electrocoagulation
109 reactor equipped with iron (Fe) electrodes serving as the coagulant source and a clarifier air flotation mechanism for
110 the separation process (**Fig. 2**). The anode electrodes were strategically positioned at specific intervals to optimise the
111 iron ion release efficiency during the electrocoagulation process. The anode comprised nine (9) units, each containing
112 99.97% iron, classified as ST 41 type, with a diameter of 40 mm and a height of 1000 mm. A direct current (DC) of
113 60 V and 1200 A from Haney Electrochemical Ltd., China, was applied to oxidise the anode electrons, resulting in
114 the formation of Fe^{2+} ions that react with pollutants in the wastewater. The cathode in the electrocoagulation system
115 comprised a multi-rod helical system (MRHS) with a nine (9) configuration provided by PT. Selaras Alam Varia
116 Energi, Bandung, Indonesia. The MRHS structure had a height of 1000 mm, and the gap between the MRHS and the
117 anode ranged from 5 to 8 mm. The system was configured in series between the anode and cathode, featuring a one-
118 on, one-off mechanism to facilitate ease of operation. An induced air flotation system was employed to facilitate the
119 separation of the formed flocs following electrocoagulation. This flotation process enhanced separation efficiency by
120 expediting the removal of flocs to the wastewater surface, resulting in cleaner water at the reactor base.

121 The EC-CAF process is initiated by entering wastewater into the mixing tank for homogenisation.
122 Subsequently, the wastewater is processed in the EC-CAF reactor, where the iron ions released from the electrodes
123 form flocs with pollutants in the water. Integration of electrocoagulation and induced air flotation enhances the
124 efficiency of pollutant removal from oil refinery wastewater. The contact time for electrocoagulation is 1 min, whereas
125 that for the clarification air flotation process is 30 min.

126

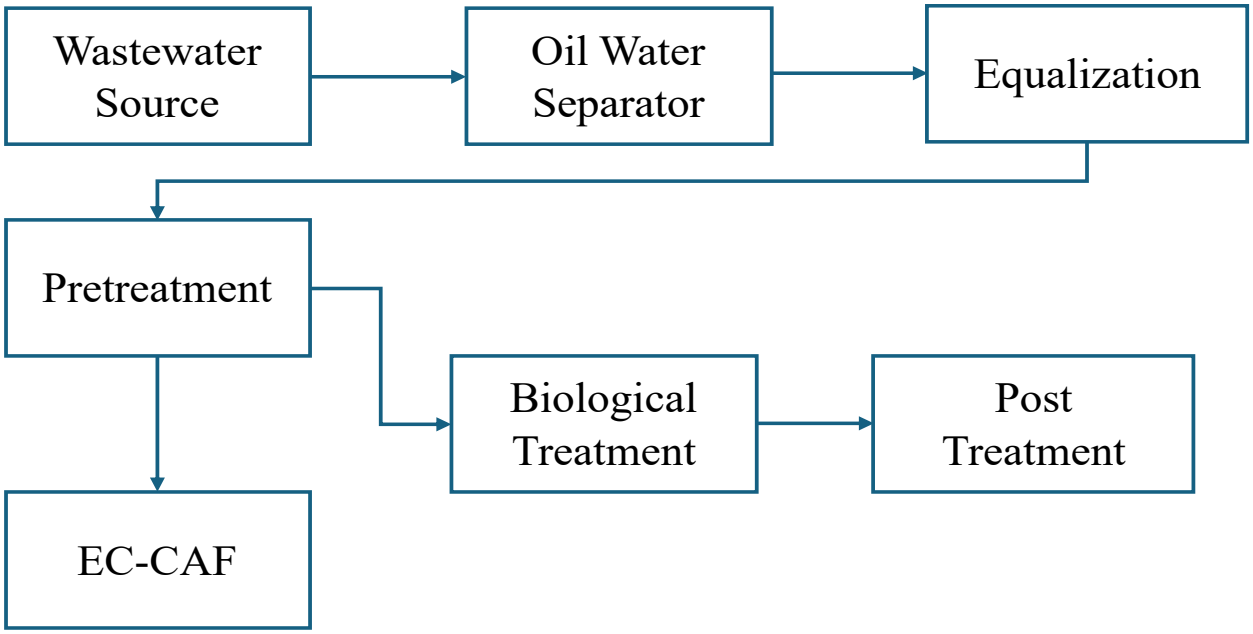


Fig. 1 Flow diagram of the full-scale application of the EC-CAF as pretreatment Integrated with Biological Treatment

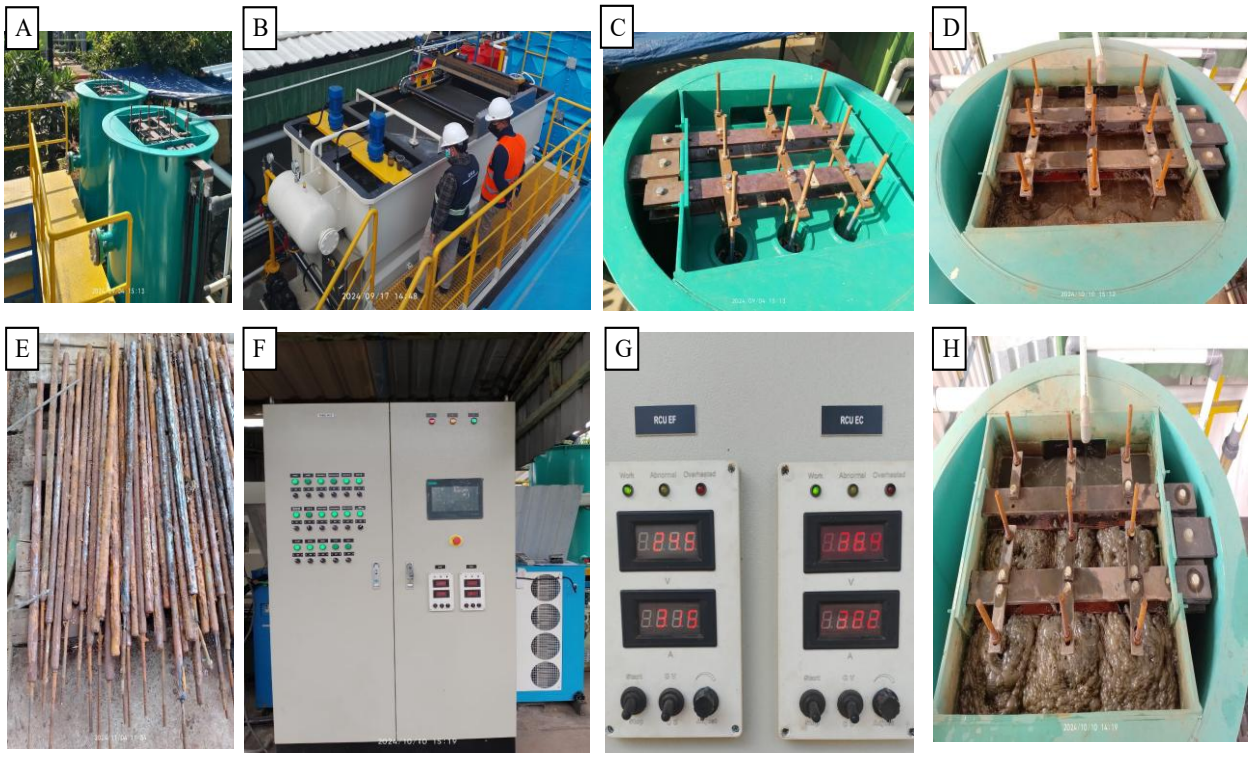


Fig. 1 Full-scale application of the EC-CAF reactor includes several components: **A**. Electrocoagulation reactor, **B**. Air flotation clarifier, **C**. Electrode configuration, **D**. Operating conditions, **E**. Anode post-replacement, **F**. Control panel system, **G**. DC power supply management, **H**. Sludge floating during the process.

2.3. Analytical data

In this study, descriptive analysis was conducted by comparing wastewater quality before and after EC treatment with induced air flotation (EC-CAF). The main analysed parameters included chemical oxygen demand (COD), total suspended solids (TSS), NH_3, sulphate, and pH, as well as the characterisation of sludge from electrocoagulation. The COD was measured using UV-Vis spectrophotometry with a closed reflux method, while the TSS was analysed gravimetrically through the process of filtering and drying the samples. NH3-N concentration was analysed using UV-Vis spectrophotometry, sulphate was determined using the turbidimetric method with a spectrophotometer, and pH was measured using a digital pH metre. In addition, the electrocoagulated sludge was characterised to understand the structure and composition of the flocs formed during the process. This analysis included scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDX) to observe the morphology and elemental distribution in the flocs. EDX analysis was conducted before and after the electrocoagulation process to evaluate the changes in elemental composition. SEM-EDX (JEOL) analysis was conducted at 10–20 kV, utilising magnifications of 500 ×, 1000 ×, 3000 ×, and 5000 ×. In addition to SEM-EDX, X-ray fluorescence (XRF) with a Merk RIGAKU was used to examine the sludge after electrocoagulation.

2.4. Data Analysis

The current density during continuous electrocoagulation may vary owing to the applied current and change in the anode surface area. The current density formula is given by **Eq. 1.**

$$J = \frac{I}{A} \quad (1)$$

where j is the current density (A/m^2), I is the applied current (A), and A is the electrode surface area (m^2). The removal efficiencies of TOC, COD, and TSS were calculated using **Eq. 2.**

$$R(\%) = \frac{C_o - C_1}{C_o} x\ 100\% \quad (2)$$

where R is the heavy metal removal efficiency, C_0 is the initial concentration of heavy metals in the feed solution (mg L − 1), and C_t is the concentration of COD, NH_3, SO_4, and TSS in the concentrated compartment (mg L − 1). Theoretically, the amount of dissolved metal ions can be determined using Faraday's law (Eq. (3) [23,24]. However, the actual anode dissolution per volume of treated wastewater can be calculated using **Eq. (3) as** follows:

$$m_{theoritical} = \frac{I\ t\ M}{z\ F\ V} \quad (3)$$

where $m_{theoretical}$ is the theoretical quantity of anode dissolution (g/L), I is the applied current (A), t is the electrocoagulation time (s), M is the molar mass of the anode (g/mol), z is the number of electrons involved in the reaction, F is Faraday's constant (96,485 As/mol), and V *is* the volume of wastewater treated (L). Faradaic efficiency (FE) is typically used to evaluate the performance of electrocoagulation [25],[26] as shown in Eq. 4**.**

$$FE\ (\%) = \frac{Actual\ Dissolution}{Theoritical\ Dissloultion}\ x\ 100\% \quad (4)$$

where m_{actual} is the actual quantity of anode dissolution (g/L), m_0 is the mass before the electrocoagulation process (g), m_t is the mass after the electrocoagulation process (g), *and* V is the volume of wastewater treated (L).

Energy consumption is the major cost component of the electrocoagulation process [[27]. This can be calculated using **Eq. (5).**

$$C = \frac{U\ I\ t}{V} \quad (5)$$

where C_{energy} is the energy consumption (kWh/m^3), U is the applied voltage (V), i is the applied current (A), t is the electrocoagulation time (h), and V is the wastewater volume (m^3).

3. Results and Discussion

3.1. Efficiency of COD and TS removalS

The integrated EC-CAF system significantly reduced suspended solids from used oil refining wastewater throughout the monitoring period. The total suspended solids (TSS) in the influent ranged from 318.17 to 440.45 mg/L, while the effluent concentrations decreased to approximately 40–45 mg/L. The removal efficiencies were 88.2%, 86.8%, and 90.7% in October, November, and December, respectively, with an average efficiency of 88.6% (Fig. 3). The CAF stage helps separate solids by attaching tiny bubbles to clumps, allowing them to float and be easily removed from the liquid. The consistent reduction of total suspended solids (TSS) over a three-month period demonstrates the efficacy of the electrocoagulation–chemical air flotation (EC-CAF) system, even amidst influent concentration variations. This finding aligns with the separation capabilities inherent in electrocoagulation technology, as well as the chemical coagulation process utilising dissolved air flotation, which achieves a TSS reduction of 68-77% at both laboratory and full-scale levels [28],[29].

Conversely, chemical oxygen demand (COD) removal was less effective. The COD levels in the influent varied between 3705.67 and 4063.55 mg/L, whereas the effluent concentrations decreased to 3223.55–3501.29 mg/L, resulting in removal efficiencies of 13.8%, 8.6%, and 15.0%, respectively, with an average reduction of 12.5%. These

findings differ from those obtained through the electrocoagulation process applied to petroleum wastewater, which achieved a substantial COD reduction of 99% with a retention time of 45.15 min at a current density of 30 mA/cm² or 300 A/m²[30]. Another study indicated that using a perforated scrap model led to yields exceeding 90% with a current density of 22 mA/cm² or 220 A/m² and a retention time of 39 min [31]. Other results demonstrated that EC treatment achieved a 52% efficiency in phenol removal with a retention time of 120 min [32].

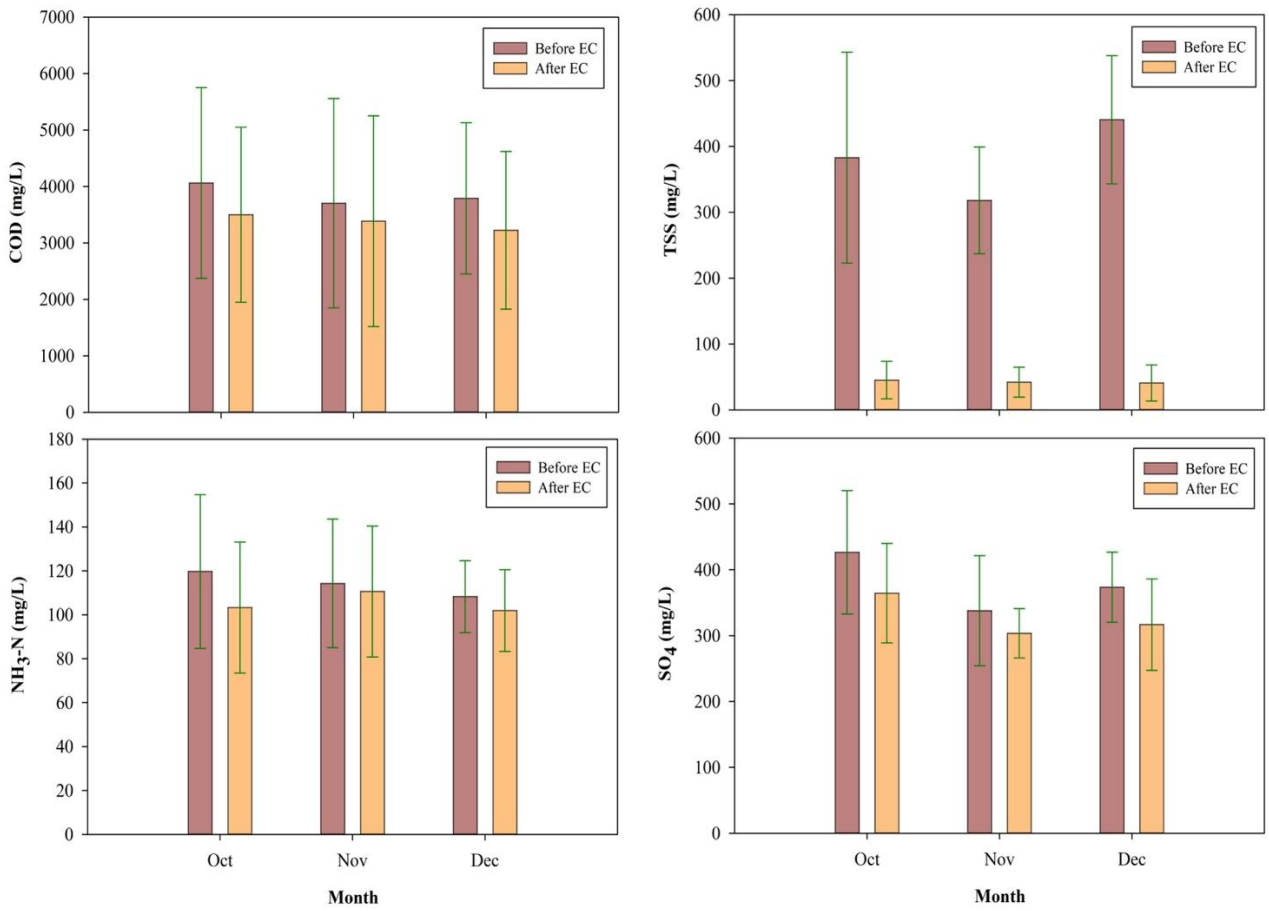


Fig. 3 Performance of full-scale application of EC-CAF for COD, NH_3-N, SO_4, and TSS

As shown in **Fig. 3,** modest COD removal can be attributed to dissolved organic compounds, such as hydrocarbons, aromatic compounds, and residual additives, in used oil refining wastewater, which are less effectively removed by coagulation and flotation processes. This scenario occurs because of the unique properties of wastewater, as the waste being processed is used oil that is redistilled into new fuel. Consequently, the wastewater produced differs from that associated with petroleum operations. Therefore, the treatment process must be adjusted to accommodate

non-biodegradable wastewater, for example, by combining it with the EC-activated sludge [33] process or with advanced oxidation processes using EAOP [34], [35].

3.2. Efficiency removal of NH_3-N and SO_4

Fig. 3 shows that the dissolved nitrogen and sulphate during the (EC-CAF) treatment showed a distinct trend compared with the particulate parameters, such as the total suspended solids (TSS). The influent ammonia nitrogen (NH_3-N) concentration ranged from 108.26 to 119.74 mg/L, while the effluent concentrations remained between 101.97 and 110.60 mg/L, corresponding to removal efficiencies of 3.2%–13.7%, with an average of 7.6%. This limited removal is due to ammonia existing predominantly as dissolved ammonium (NH_4^+) under near-neutral pH conditions (pH 6–8), as shown in **Fig. 4.** Unlike colloidal particles, ammonium ions are not readily destabilised by electrocoagulation. Electrochemical reactions at the sacrificial anode produce metal ions that form hydroxide flocs, which primarily adsorb suspended contaminants.

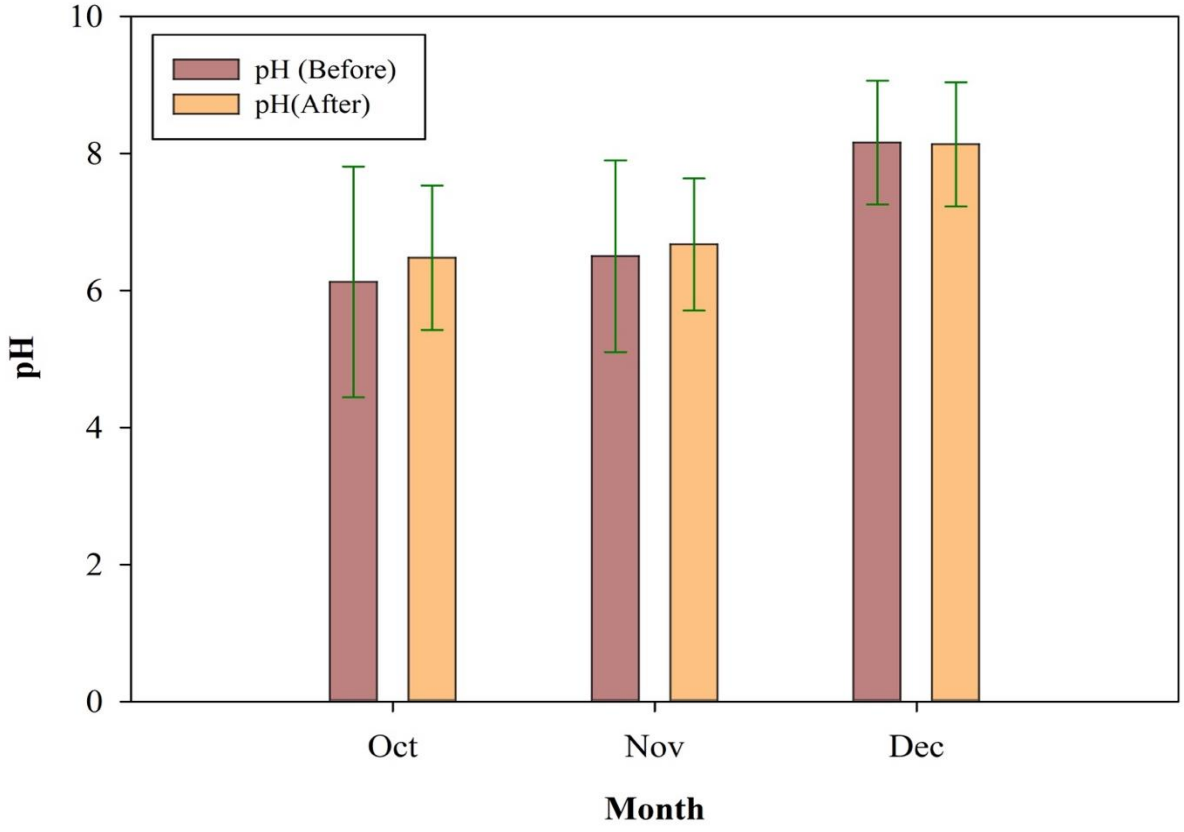


Fig. 4. pH during the operation of EC-CAF

The interaction between these flocs and dissolved ammonium is weak, thereby limiting the ammonia removal efficiency. The observed minor reductions may be attributed to the partial adsorption of ammonia onto the hydroxide surfaces during flotation. These findings clearly demonstrate that the electrocoagulation process will not achieve maximal impact on ammonia levels owing to the non-biodegradable nature of wastewater. Conversely, when addressing biodegradable wastewater, electrocoagulation can serve as a pretreatment to reduce ammonia or ammonium content, as reported by [36], [37]. Sulphate (SO_4^{2-}) exhibited similar behaviour, with influent concentrations of 338.00–426.45 mg/L decreasing to 303.47–364.52 mg/L in the effluent, achieving a removal efficiency of 10%–15%. sulphate remains predominantly dissolved under electrocoagulation conditions, with removal occurring through limited electrostatic interactions with metal hydroxide flocs. While ironelectrodese produce $Fe(OH)_3$ orprovidesa high surface area for adsorbing anionic species, sulphate exhibits weak affinity compared with other anions, such as phosphate. Insufficient precipitation reactions during the operation further limit the reduction of sulphate. These observations indicate that different types of wastewater yield varying outcomes when treated with the electrocoagulation process, implying that this technique is not suitable for nonbiodegradable wastewater, particularly regarding the NH_3-N and SO_4^{2-} parameters. However, previous studies have shown that electrocoagulation can effectively reduce SO_4^{2-} by 82.7 to 87% with a retention time of 135 min at a pH of 5.5 and an energy consumption of 3.617 kWh/m^3 [38],[39]. Additionally, the combined process using an inert BDD electrode along with electrocoagulation achieved optimal sulphate removal, underscoring the oxidative capability of the BDD electrode as documented in earlier research [40].

The relationship between ammonia nitrogen (NH_3-N), sulphate (SO_4^{2-}), and chemical oxygen demand (COD) in EC-CAF-treated wastewater provides insights into the pollutants present in used oil refining effluents. COD concentrations remained high after treatment, with removal efficiencies averaging approximately 12.5%, while NH_3-N and SO_4^{2-} showed limited reductions of approximately 7.6% and 13.3%, respectively. This indicates that a substantial fraction of COD originates from dissolved compounds rather than particulate organic matter. In addition, the COD/SO4 ratio factor exceeded the ratio indicating biodegradability. According to Archilha et al. [[41], an increase in the COD/SO4 ratio is associated with a decrease in organic content, resulting in a decline in process efficiency as the ratio rises from 1 to 3. The findings from the electrocoagulation application in this study reveal that the COD/SO4 ratio exceeds 8, as illustrated in **Fig. 5**, indicating that the wastewater from the used oil refining process contains non-biodegradable organics. Furthermore, the BOD/COD ratio of this wastewater, with a value of 0.2 (**Table 1**),

corroborates its classification as nonbiodegradable. Consequently, only a portion of the wastewater is treated in this process, and it is ineffective in significantly reducing COD, SO4, and ammonia content, even with the application of high energy. This observation aligns with [42], which differentiates COD based on its biodegradability, categorising it as completely reduced, partially reduced, or not reduced at all. In this study, COD was predominantly partially reduced; however, considering the COD/SO_4 ratio, it tended to remain unreduced.

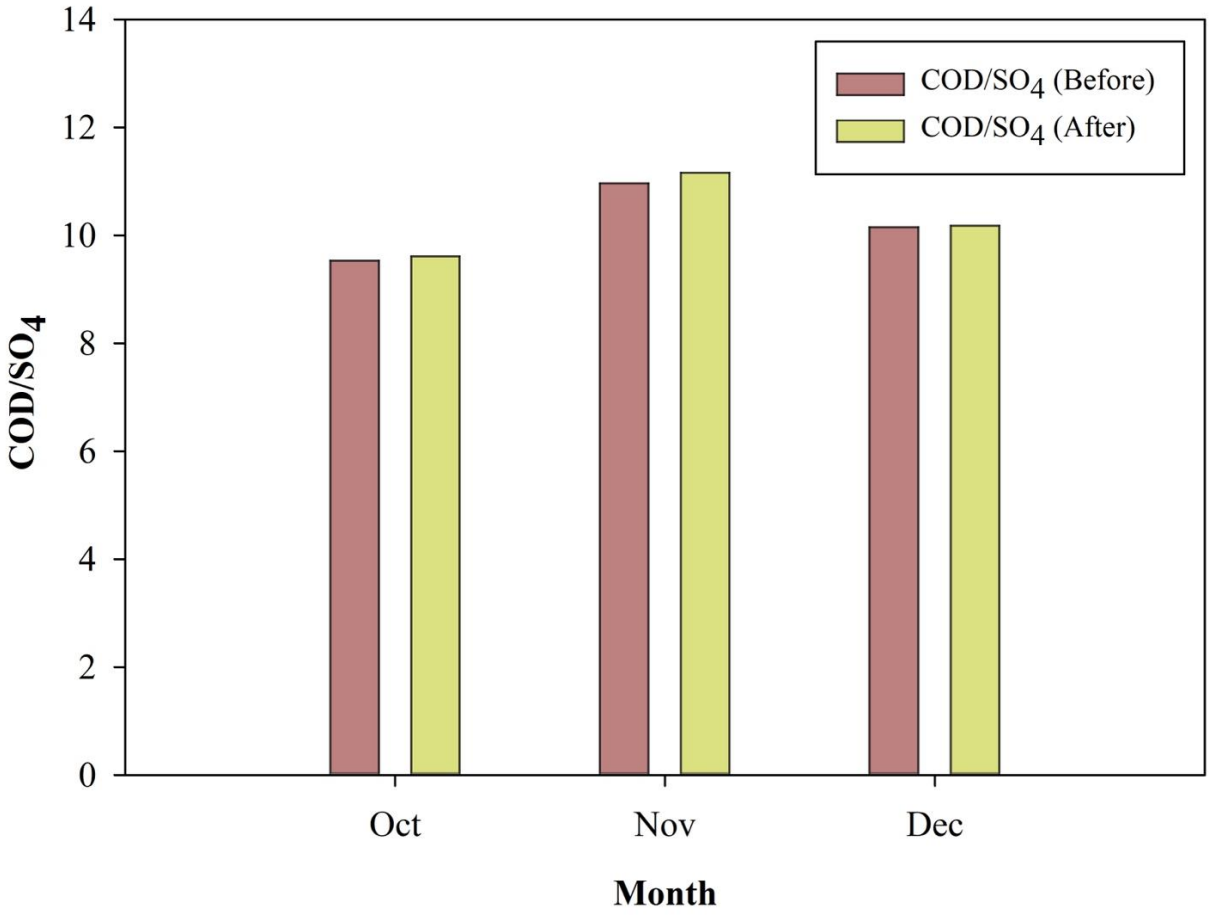


Fig. 5 COD/SO_4 ratio during pretreatment

3.3. Effect of power consumption and current densityy

Operational parameters, such as voltage, current, and specific energy consumption, are pivotal in assessing the electrocoagulation–CAF (EC-CAF) system for treating industrial wastewater from used oil refining. During operation, the voltage increased from 28.24 V in October to 47.97 V in December, whereas the current fluctuated between 415 and 496 A as shown in **Fig. 6**. This increase in electrical input corresponded to an increase in the specific energy consumption from 2.06 to 3.50 kWh/m³, as shown in **Fig. 7**. An enhanced electrical input facilitates electrode

dissolution, resulting in intense foaming within the reactor, as shown in Fig. 1 (H). These flocs act as coagulants, destabilising colloidal particles and promoting aggregation, which accounts for the high TSS removal efficiency (86%–90%). The increase in the TSS removal efficiency in December (90.21%) coincided with the highest voltage and current. However, the efficiency remained notably low for the chemical oxygen demand (COD), ammonia, and sulphate (SO4) parameters, despite the power input being consistent with previous studies [38], ranging from 3.6 kWh/m³ or higher, as shown in **Fig. 7.**

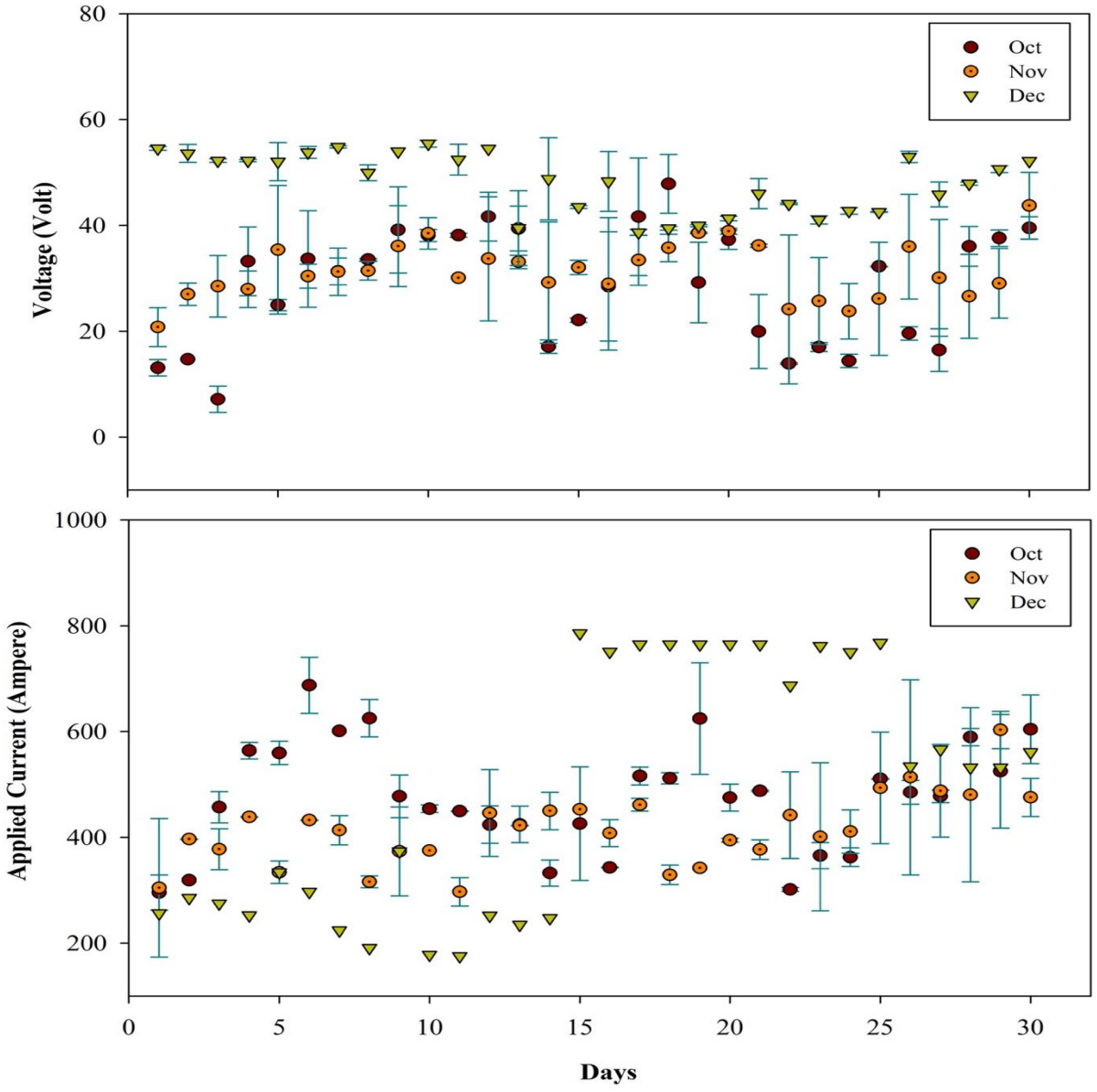


Fig. 6. Applied voltage and current during full-scale operation

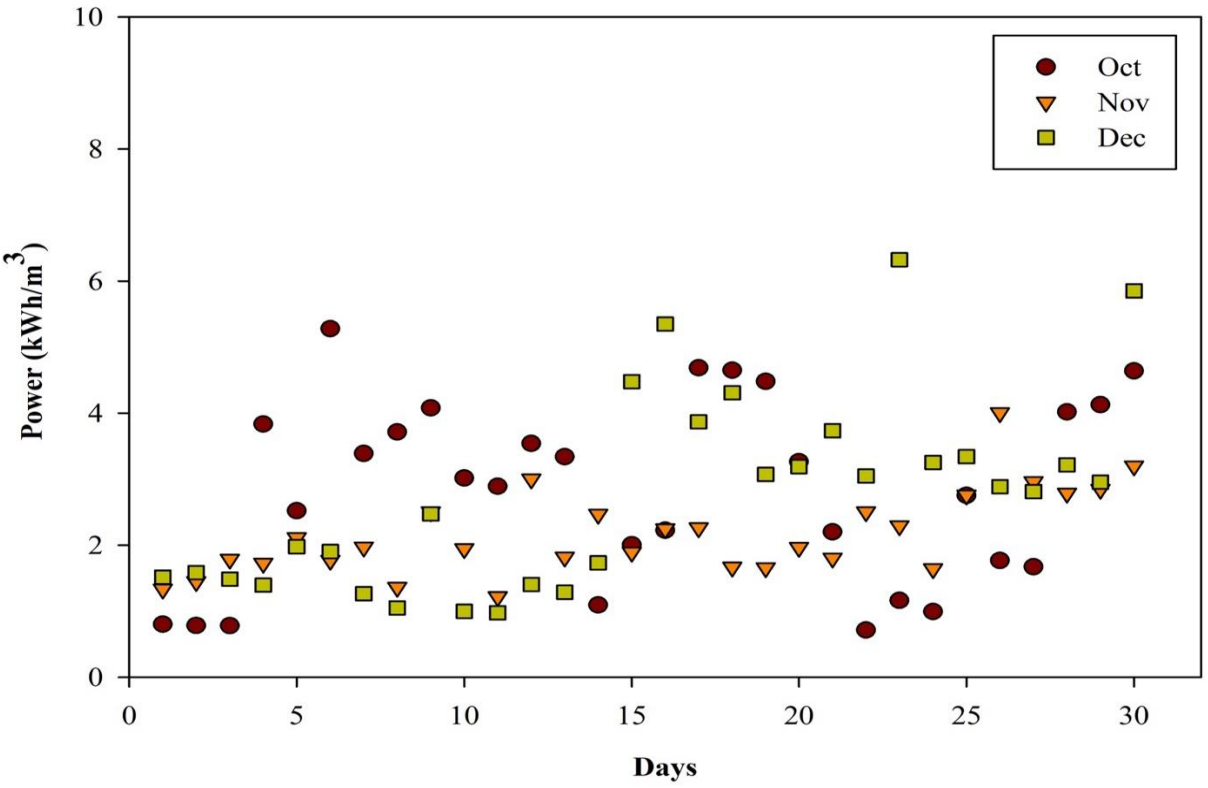


Fig. 7. Applied power during full-scale operation

Analysis of **Figs. 7** and **3** reveals a moderate correlation between electrical input and chemical oxygen demand (COD) removal. In December, an increase in voltage and energy consumption resulted in a COD removal efficiency of 16.59%, compared to 14.92% and 11.20% in October and November, respectively. This enhancement is attributed to the increased formation of hydroxide flocs and improved destabilisation of emulsified oil droplets and organic colloids. In accordance with Faraday's law, a higher current facilitates the rapid dissolution of the anode, thereby augmenting the production of coagulants and surface adsorption capacity. Additionally, the increased current intensifies hydrogen gas evolution at the cathode, producing microbubbles that aid in the flotation of oil-organic flocs. Nevertheless, overall COD removal remains constrained, as most COD in used oil refinery wastewater comprises dissolved hydrocarbons and stable organic compounds, which are less amenable to removal via electrocoagulation. The removal efficiency of dissolved species such as NH_3-N and SO_4^{2-} exhibits a weaker correlation with electrical parameters. Although there is a slight increase in efficiency at higher energy input, with SO_4 removal reaching 15.95% in December, the efficiency remains low. This underscores the limitations of the electrocoagulation process in removing dissolved ionic species. As illustrated in **Fig. 8,** increased current can enhance current density and produce

high metal hydroxide formation and electrochemical reactions. Ammonium and sulphate ions remain stable in solution and interact only weakly with the generated coagulants.

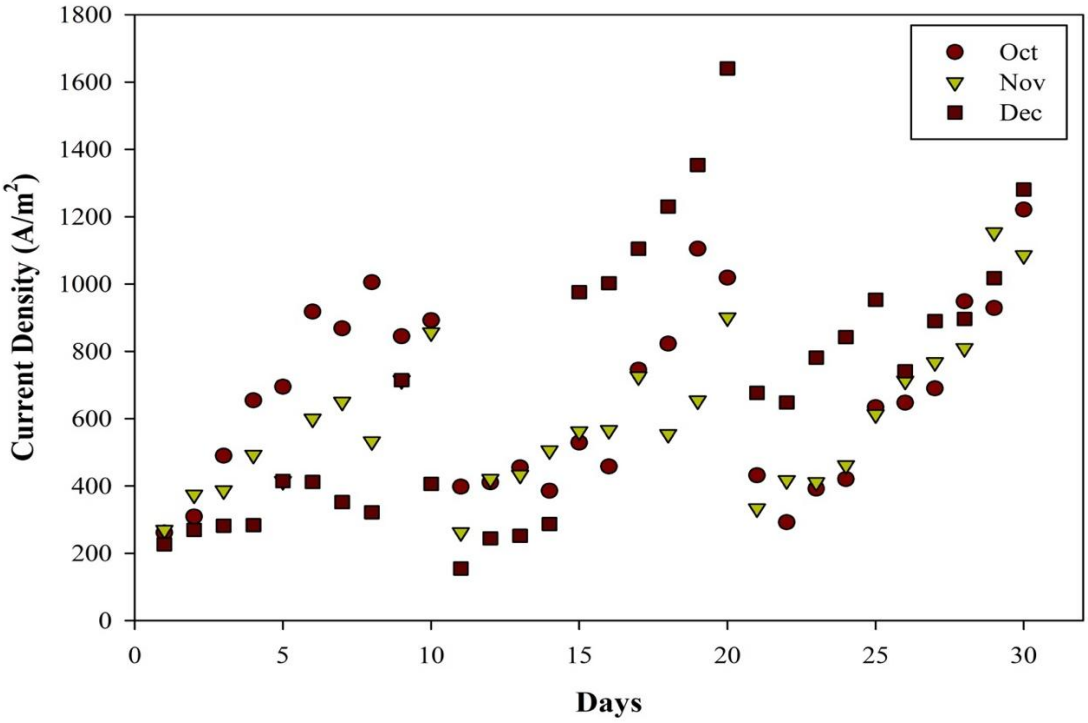


Fig. 8. Applied current density during full-scale operation

The results suggest that increasing the electrical input improves the coagulation-flotation process, affecting total suspended solids (TSS) and particulate COD, but has a limited effect on dissolved nitrogen and sulphate ions. Consequently, this study contradicts earlier research, which claimed that boosting power would enhance the reduction of organic parameters, as previously demonstrated by [43], [44], [45]. The brief contact time of only 1 min within the multi-rod helical system reactor is a significant factor contributing to this outcome. Furthermore, the chemical oxygen demand to sulphate (COD/SO_4) ratio indicates that the process was ineffective, despite the application of high current density values as illustrated in **Fig. 8**, which range from 200 to 1600 A/m^2. In previous studies [46], increasing the current density above 1000 A/m^2 resulted in very good efficiency for COD removal from domestic wastewater; however, this showed an inverse ratio with the biodegradability level compared to wastewater categorised as non-biodegradable.

3.4. Electrode consumption and Faradaic efficiency

The impact of increased power and current density led to electrode consumption during full-scale operation, where the anode diameter decreased from 40 mm to 16.3 ± 1.63 mm in the first 10 days, 16.16 ± 0.57 mm in the second 10 days, and 16.16 ± 1.154 mm in the third 10 days, with electrodes being used up to 75% of their original size as shown **Fig. 9**. This consumption only affected the TSS reduction, as shown in **Fig. 3**, whereas the COD, sulphate, and ammonia parameters were not effectively reduced for the wastewater from the used oil purification. The electrode consumption lasted up to 10 days, as indicated by a 75% weight reduction; continuing beyond this would increase the power due to a higher current density. Replacing the iron anode after 10 days provides energy usage stability. In October, the system showed a Faradaic efficiency of 53.35%, where the actual consumption (50.63 ± 1.72 kg/10 days) was half of the theoretical consumption (95.59 ± 5.22 kg/10 days). This difference indicates that electrode passivation from the formation of the oxide layer requires electrode cleaning every 12 h. In November, the performance improved with an FE of 66.13 %, with the theoretical consumption (83.36 ± 9.61 kg/10 days) being closer to the actual consumption (55.13 ± 0.00 kg/10 days). In December, the FE dropped to 55.50%, with variations in the theoretical consumption (97.96 ± 39.6 kg/10 days), as shown in **Table 2**.

Table 2. Faradaic efficiency during the full-scale operation of pretreatment of used oil refineries

Month	Consumption Anode Theory (Kg/10 Day)	Actual consumption anode (Kg/10 Day)	Faradaic efficiency (%)
October	95.59 ± 5.22	50.63 ± 1.72	53.35
November	83.36 ± 9.61	55.13 ± 0.00	66.13
December	97.96 ± 39.6	54.37 ± 1.29	55.50

As shown in **Table 2** and **Fig. 9**, these fluctuations indicate instability in the operational parameters, such as voltage and current density, as well as possible changes in the wastewater characteristics, as shown in **Figs. 6** and **7**. The presence of complex organic compounds, suspended solids, and ions, such as chloride and sulphate, in the used oil purification waste can disrupt the electron transfer process and increase side reactions, thereby reducing the anode dissolution efficiency. Overall, the Faradaic efficiency values (ranging from 53% to 66%) indicate that under ideal electrochemical conditions, electrocoagulation has not occurred. The difference between the theoretical and actual consumption indicates that part of the supplied electrical energy is dissipated through side reactions rather than being fully converted into coagulant formation.

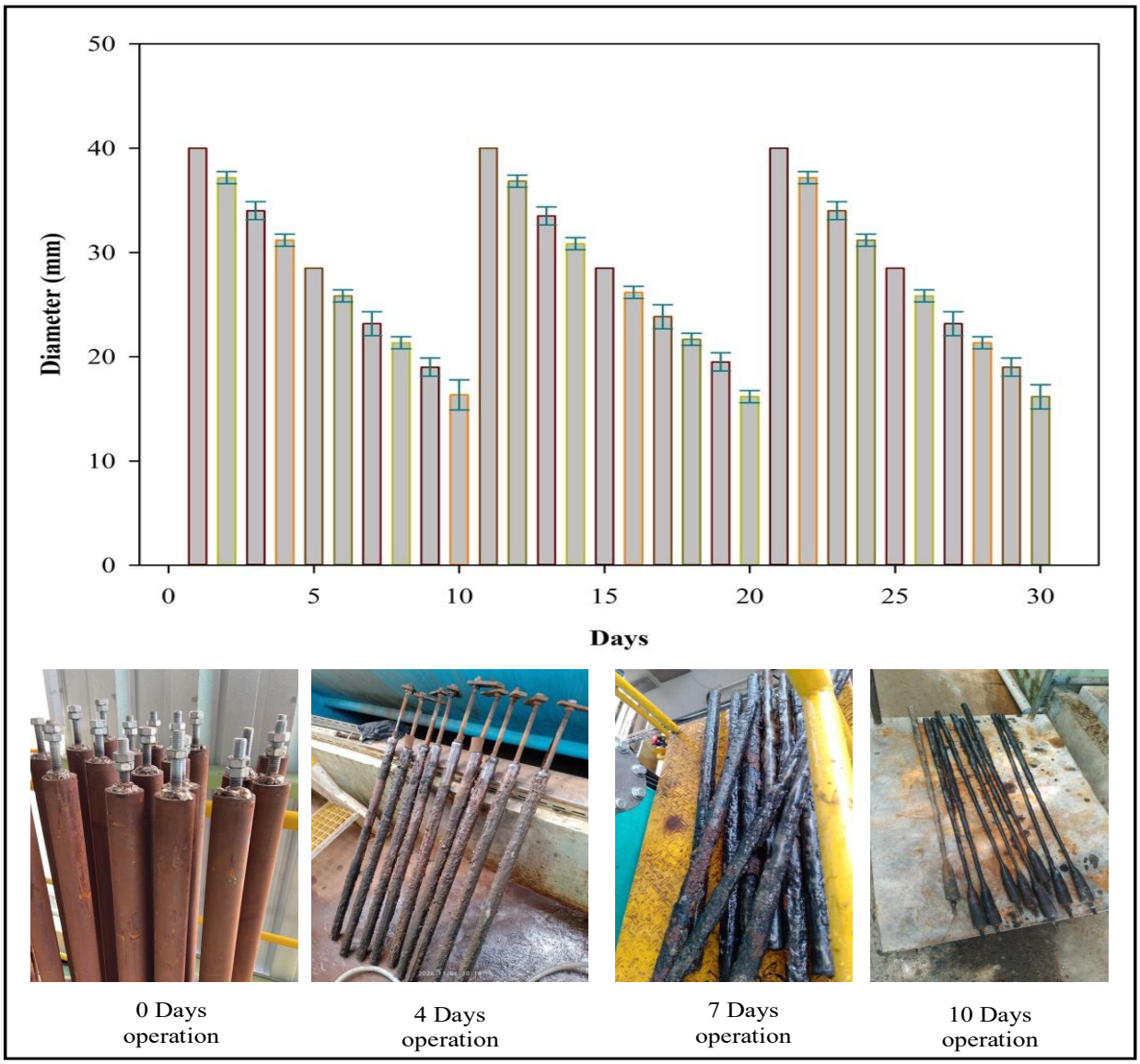
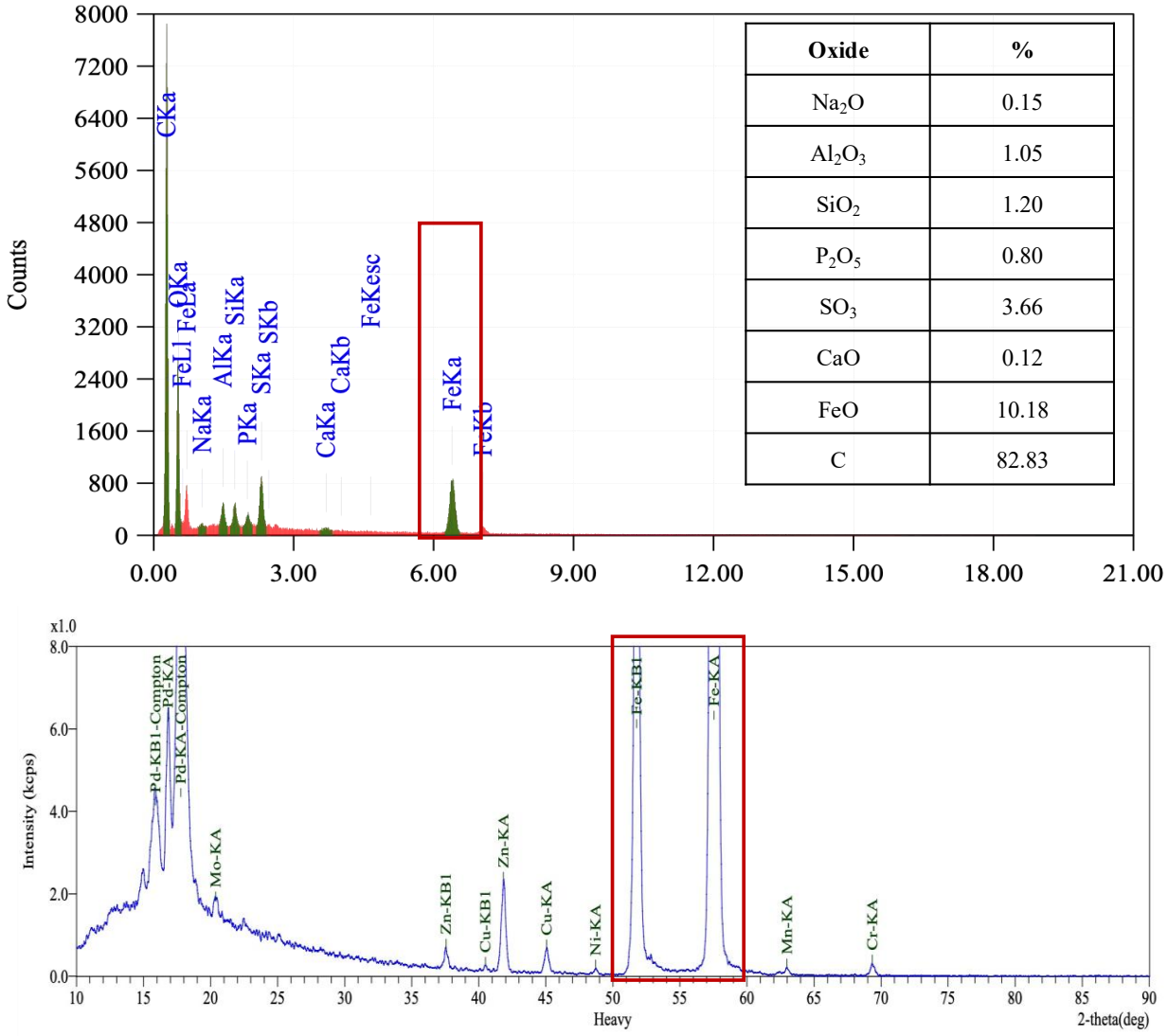


Fig. 9. Electrode reduction during full-scale operation

3.5. Sludge Characteristic

Elemental composition analysis via X-ray fluorescence (XRF) reveals that the solid produced from electrocoagulation is predominantly composed of iron oxide (Fe_2O_3) at 7.75%, underscoring the iron electrode's primary function as an in situ coagulant source. The iron predominance is attributed to the anodic dissolution process, which generates Fe^{2+}/Fe^{3+} ions that subsequently undergo hydrolysis to form hydroxide flocs, such as $Fe(OH)_3$ and various iron oxyhydroxides. Because of their active hydroxyl groups and substantial surface area, which facilitate the adsorption and coprecipitation of pollutants, these flocs are recognised as the principal components in the

electrocoagulation mechanism. In addition to iron, elements such as silicon, aluminium, calcium, and sulfur were detected, indicating the presence of inorganic particles and dissolved ions trapped within the floc structure during the treatment process. The presence of calcium and sulfur also suggests potential interactions with sulphate ions or the formation of secondary inorganic compounds during the coagulation process, as evidenced by the formation of SO3 at 3.04% from the XRF analysis (Fig. 10).



Oxide	%
Na_2O	0.15
Al_2O_3	1.05
SiO_2	1.20
P_2O_5	0.80
SO_3	3.66
CaO	0.12
FeO	10.18
C	82.83

Fig. 10. EDX and XRF analysis of the EC-CAF slurry

Fig. 11 (A and B) illustrates that morphological characterisation via SEM reveals that the solid produced from electrocoagulation exhibits an amorphous structure with irregular aggregate shapes and a rough, porous surface texture. This morphology is reminiscent of "fluffy" flocs, a characteristic feature of iron hydroxide formed through

electrocoagulation. The observed morphology suggests the formation of particle aggregates through colloid destabilisation and sweep flocculation mechanisms. Energy-dispersive X-ray spectroscopy (EDX) analysis, integrated with SEM, indicates that FeO (10.18 %) is the major component, followed by SO3 (3.66 %), as described by the overlay scatter diagram in **Fig. 11 D** and **E**, confirming the presence of iron hydroxide as the primary matrix.

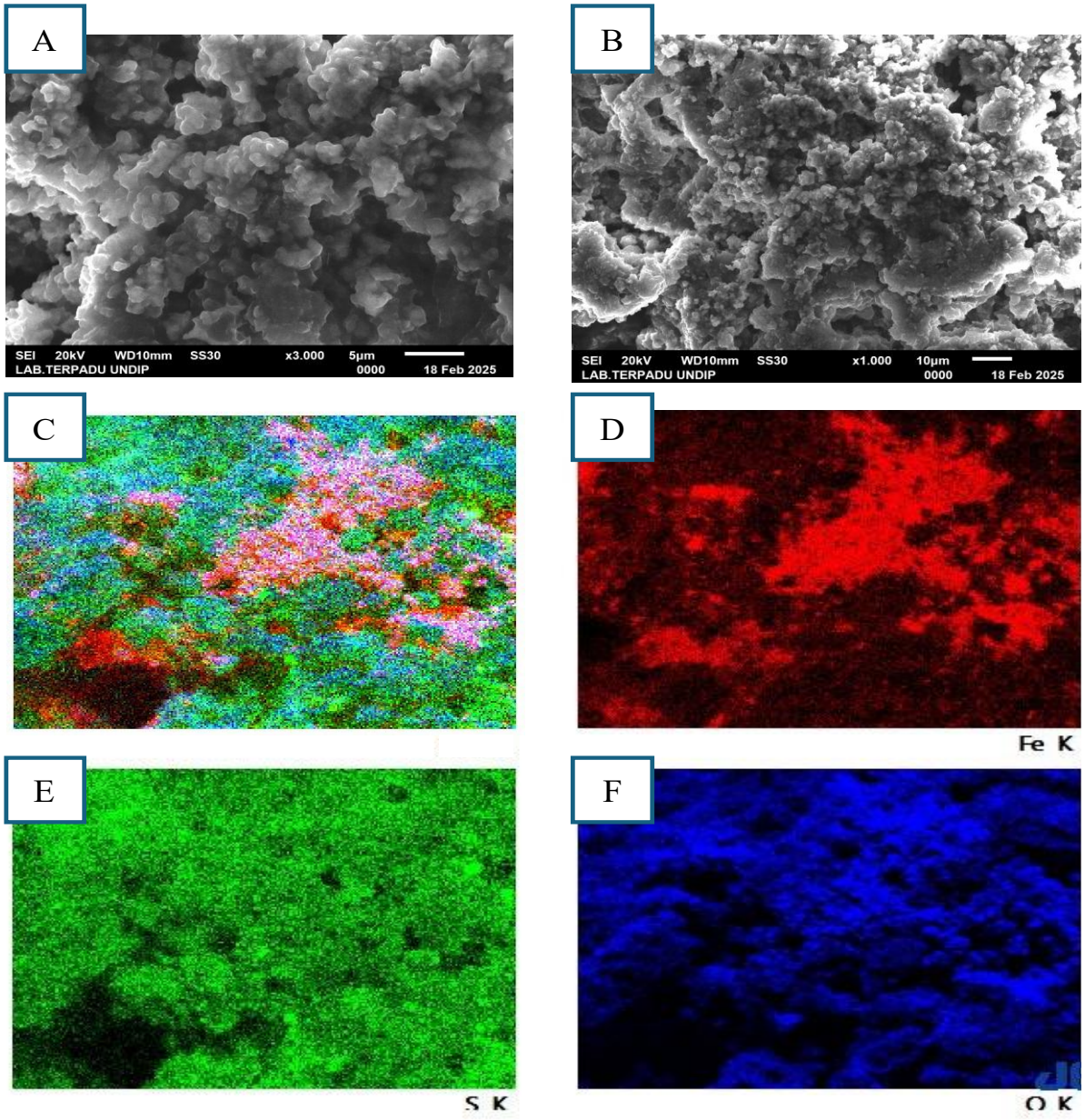


Fig. 11. SEM results (A, 3000 ×; B, 1000 ×); overlay (C); scatter diagram Fe (D), S (E), and F (O).

The XRF and SEM–EDX analyses suggest that the formation of iron hydroxide flocs is the primary mechanism in the electrocoagulation process. These flocs function as amorphous and porous adsorbent media capable of adsorbing and entrapping suspended particles and organic compounds through sweep flocculation and surface adsorption,

respectively. However, this process appears to be more effective in removing particulate and colloidal pollutants than in eliminating dissolved contaminants, such as ammonia and sulphate, based on the identified chemical characteristics. Consequently, integrating electrocoagulation with advanced processes, such as advanced oxidation or biological treatment, is necessary to enhance the overall efficiency of wastewater treatment.

4. Conclusion

The integrated electrocoagulation–chemical air flotation (EC–CAF) system shows significant efficacy in treating waste from used oil refining, particularly in removing total suspended solids (TSS) with 88.6% efficiency. This efficiency stems from the formation of metal hydroxide flocs during electrocoagulation, enhanced by dissolved air flotation. Increased electrical input further improves the coagulation–flotation process, with TSS removal exceeding 90%. However, the EC–CAF system has limitations in removing dissolved parameters, including chemical oxygen demand (COD), ammonia (NH_3-N), and sulphate (SO_4^{2-}), with efficiencies of 12.5%, 7.6%, and 13.3%, respectively. This reduced efficiency is due to nonbiodegradable dissolved organic compounds that resist coagulation and flotation mechanisms. A high chemical oxygen demand to sulphate ratio (>8) and low BOD/COD ratio (0.17) indicate resistance to degradation. While increasing the electrical energy consumption to 3.60 kWh/m³ enhances coagulant and microbubble formation, it does not significantly improve the removal of dissolved compounds. A Faraday efficiency of 53%–66% suggests an unoptimized electrocoagulation process. Sludge characterisation reveals iron-based compounds with an amorphous and porous morphology, which are effective for particulate pollutants but less effective in binding dissolved ions. The EC–CAF technology effectively removes suspended solids as a pretreatment; however, integration with biological or advanced oxidation processes is necessary to enhance the treatment of dissolved organic compounds.

CRediT authorship statement

Rachmad Ardhianto: Writing, original draft, reviewing and editing, resources, funding acquisition, and data curation. **Valencia Safir Maharani**: Writing, editing, methodology, and conceptualisation. **Nadia Paramita**: Writing, editing, methodology, and conceptualisation**. Bening Laksa Intan**: writing, editing, and methodology**. Eqiu Alkahfi Tarif**: writing, reviewing, and editing.

Declaration of Competing Interest

The authors declare that they have no competing financial interests or personal relationships that could influence the results of this study.

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5. Reference

[1] X. Tang, P.E. Eke, M. Scholz, S. Huang (2009) Processes impacting on benzene removal in vertical-flow constructed wetlands. Bioresour. Technol. 100:227–234.

[2] J.M. Bayona, C. Domínguez, J. Albaigés (2015) Analytical developments for oil spill fingerprinting. Trends in Environ Analytical Chem. 5:26–34.

[3] J.F.P. e J.C. Afonso (2012) New Strategies For Treatment And Reuse Of Spent Sulfidic Caustic Stream From Petroleum Industry. 35:1447–1452. https://doi.org/https://doi.org/10.1590/S0100-40422012000700027.

[4] K. Ghezali, N. Ben Tahar, G. Otmanine, N. Guettaf (2021) Environmental assessment and compliance of wastewater from Arzew oil refinery for future development and reuse. J. Environ Treat Tech 10:18–23. https://doi.org/10.47277/JETT/10(1)23.

[5] X. Dai, C. Chen, Y. Chen, S. Guo (2020) Comprehensive Evaluation of a Full-Scale Combined Biological Process for the Treatment of Petroleum Refinery Wastewater using GC-MS and PCR-DGGE Techniques. Int. J. Electrochem. Sci. 15:2013–2026. https://doi.org/https://doi.org/10.20964/2020.03.19.

[6] M.H. El-Naas, M.A. Alhaija, S. Al-Zuhair (2014) Evaluation of a three-step process for the treatment of petroleum refinery wastewater. J. Environ. Chem. Eng. 2:56–62.

[7] K.T. Amakiri, A.R. Canon, M. Molinari, A. Angelis-Dimakis (2022) Review of oilfield produced water treatment technologies. Chemosphere 298:134064.

[8] T. Al-Khalid, M.H. El-Naas (2018) Organic contaminants in refinery wastewater: characterization and novel approaches for biotreatment. Recent Insights in Petroleum Scie and Eng 371

[9] B.H. Diya'uddeen, W.M.A.W. Daud, A.R.A. Aziz (2011) Treatment technologies for petroleum refinery effluents: A review. Proc Safety and Environ Protect 89:95–105.

[10] H. Mehdizadeh, J. Shayegan (2003) The Effect of Sulfate Concentration on Cod Removal and Sludge Granulation in UASB Reactors 16: 1–10. https://www.ije.ir/article_71409_f8515642b60ea7d4eb7c876428301c7e.pdf.

[11] S. Vendramel, J.P. Bassin, M. Dezotti, G.L. Sant'Anna Jr (2015) Treatment of petroleum refinery wastewater containing heavily polluting substances in an aerobic submerged fixed-bed reactor. Environ. Technol. 36:2052–2059.

[12] B. Singh, P. Kumar (2020) Pre-treatment of petroleum refinery wastewater by coagulation and flocculation using mixed coagulant: Optimization of process parameters using response surface methodology (RSM). J Water Proc Eng 36:101317.

[13] M. Dehghani, M.H. Alizadeh (2016) The effects of the natural coagulant Moringa oleifera and alum in wastewater treatment at the Bandar Abbas Oil Refinery. Env health Eng and Manag J 3:225–230.

[14] E.E. Ebrahiem, M.N. Al-Maghrabi, A.R. Mobarki (2017) Removal of organic pollutants from industrial wastewater by applying photo-Fenton oxidation technology. Arabian J. Chemistry 10:S1674–S1679.

[15] Y. Zhang, X. Quan, S. Chen, Y. Zhao, F. Yang (2006) Microwave assisted catalytic wet air oxidation of H-acid in aqueous solution under the atmospheric pressure using activated carbon as catalyst. J. Hazard. Mater. 137:534–540.

[16] R. Girón-Navarro, I. Linares-Hernández, L.A. Castillo-Suárez, V. Martínez-Miranda, E.A. Teutli-Sequeira (2024) Electrocoagulation vs. coagulation–flocculation applied to real wastewater and set up to pilot/full scale. Environ Techno Reviews 13: 722–753. https://doi.org/10.1080/21622515.2024.2419562.

[17] A.A. Shah, S. Walia, H. Kazemian (2024) Advancements in combined electrocoagulation processes for sustainable wastewater treatment: A comprehensive review of mechanisms, performance, and emerging applications. Water Res. 252: 121248. https://doi.org/10.1016/j.watres.2024.121248.

[18] O.F. Saeed, K.W. Hameed, A.H. Abbar (2023) Treatment of vegetable oil refinery wastewater by sequential electrocoagulation-electrooxidation process, J. Environ. Manage. 342:118362. https://doi.org/10.1016/j.jenvman.2023.118362.

[19] Q.K. Hameed, W.T. Mohammed (2024)Treatment of Wastewater from Oil Refineries by a Combination of Electrocoagulation with Photocatalytic Processes Using Immobilized Nano-Zinc Oxide Photocatalyst. J. Eco Eng. 25:109–123. https://doi.org/10.12911/22998993/192671.

[20] R. Ardhianto, G. Samudro, S. Mangkoedihardjo, S.T. Isratul Izmi (2025) Integrated approach to textile wastewater treatment: Investigating electrocoagulation, MBBR, and adsorption synergy. J. Water Proc Eng 77:108364. https://doi.org/https://doi.org/10.1016/j.jwpe.2025.108364.

[21] R. Ardhianto, A.D. Anggrainy, G. Samudro, A. Triyawan, A.Y. Bagastyo (2024) A study of continuous-flow electrocoagulation process to minimize chemicals dosing in the full-scale treatment of plastic plating industry wastewater. J. Water Proc Eng 60:105217. https://doi.org/https://doi.org/10.1016/j.jwpe.2024.105217.

[22] E.K. Tetteh, E.O. Ezugbe, S. Rathilal, D. Asante-Sackey (2020) Removal of COD and SO42-from oil refinery wastewater using a photo-catalytic system-comparing TiO2 and zeolite efficiencies. Water (Switzerland) 12. https://doi.org/10.3390/w12010214.

[23] T.S.A. Singh, S.T. Ramesh (2014) An experimental study of CI Reactive Blue 25 removal from aqueous solution by electrocoagulation using Aluminum sacrificial electrode: Kinetics and influence of parameters on electrocoagulation performance, Desalination Water Treat. 52:2634–2642. https://doi.org/10.1080/19443994.2013.794714.

[24] M. Cherifi, S. Hazourli, S. Pontvianne, F. Lapicque, J.P. Leclerc (2016) Electrokinetic removal of aluminum and chromium from industrial wastewater electrocoagulation treatment sludge, Desalination Water Treat. 57:18500–18515. https://doi.org/10.1080/19443994.2015.1094427.

[25] J. Zhang, L. Liu, Y. Liang, J. Zhou, Y. Xu, R. Xiuxiu, Y. Lu, Z. Xu, Q. Guangren (2015) Enhanced precipitation of cyanide from electroplating wastewater via self-assembly of bimetal cyanide complex, Sep. Purif. Technol. 150:179–185. https://doi.org/10.1016/j.seppur.2015.07.008.

[26] M. Ingelsson, N. Yasri, E.P.L. Roberts (2020) Electrode passivation, faradaic efficiency, and performance enhancement strategies in electrocoagulation—a review, Water Res. 187:116433. https://doi.org/https://doi.org/10.1016/j.watres.2020.116433.

[27] S.Y. Guvenc, K. Dincer, G. Varank (2019) Performance of electrocoagulation and electro-Fenton processes for treatment of nanofiltration concentrate of biologically stabilized landfill leachate. J. Water Proc Eng 31 https://doi.org/10.1016/j.jwpe.2019.100863.

[28] A. Kundu, N. Gupta, A.S. Kalamdhad (2024) Optimization and kinetic analysis of electrocoagulation-assisted adsorption for treatment of young landfill leachate, J. Environ. Manage. 366:121779. https://doi.org/https://doi.org/10.1016/j.jenvman.2024.121779.

[29] A.L. Piaggio, G. Smith, M.K. de Kreuk, R.E.F. Lindeboom (2024) Application of a simplified model for assessing particle removal in dissolved air flotation (DAF) systems: Experimental verification at laboratory and full-scale level, Sep. Purif. Technol. 340:126801. https://doi.org/https://doi.org/10.1016/j.seppur.2024.126801.

[30] N. Bensalhia, K. Elhadeuf, A. Boulkaibet, N. Bougdah, N. Metnani, F. Bouzahzah, O. Meddour, M. Bouaoud (2026) Treatment of petroleum wastewater by electrocoagulation using recycled aluminum from cans as electrodes: optimization of operating parameters by Box–Behnken design, Reaction Kinetics. Mechanisms and Catalysis 139:1187–1211. https://doi.org/10.1007/s11144-026-03046-8.

[31] G.K. Akkaya (2022) Treatment of petroleum wastewater by electrocoagulation using scrap perforated (Fe-anode) and plate (Al and Fe-cathode) metals: Optimization of operating parameters by RSM, Chemical Engineering Research and Design 187: 261–275. https://doi.org/https://doi.org/10.1016/j.cherd.2022.08.048.

[32] N.R. Kadhim, M. Abdulredha (2021) Petroleum effluent treatment using electrocoagulation: A case study of phenols removal, IOP Conf. Ser. Mater. Sci. Eng. 1184:12009. https://doi.org/10.1088/1757-899X/1184/1/012009.

[33] I. Linares-Hernández, C.E. Barrera-Díaz, G. Roa-Morales, V. Martínez-Miranda, L.A. Bernal-Martínez, V. Lugo-Lugo (2010) Removal of Non-Biodegradable Compounds in a Complex Industrial Wastewater by Electrocoagulation - Activated Sludge Processes, ECS Trans. 29:227. https://doi.org/10.1149/1.3532320.

[34] C.A. Basha, E. Chithra, N.K. Sripriyalakshmi, Electro-degradation and biological oxidation of non-biodegradable organic contaminants (2009) Chemical Engineering Journal 149:25–34. https://doi.org/https://doi.org/10.1016/j.cej.2008.09.037.

[35] R. Ardhianto, A.Y. Bagastyo (2019) Personal Care Wastewater Treatment with Electro-coagulation and Electro-oxidation, E3S Web of Conferences 125. https://doi.org/10.1051/e3sconf/201912503008.

[36] R. Ardhianto, A. Yusrina, M. Rangkuti (2025) Optimizing chemical oxygen demand and total nitrogen removal from tofu wastewater using a multi-rod helical electrocoagulation system in a tubular reactor, J. Eco. Eng. 26:95–109. https://doi.org/10.12911/22998993/202092.

[37] R. Ardhianto, A. Yusrina, A.Y. Bagastyo (2025) Iron-Based Electrocoagulation Pretreatment to Enhance Magnesium-Struvite Recovery from Livestock Wastewater Using a Tubular Reactor, Int. J. Environ. Res. 2041. https://doi.org/10.1007/s41742-025-00993-3.

[38] A. Kumar, D. Basu (2025) Optimization and prediction of SO42- removal in electrocoagulation process with rotating electrodes using Taguchi and ANN approach. J. Environ. Chem. Eng. 13:116934. https://doi.org/https://doi.org/10.1016/j.jece.2025.116934.

[39] N. Mohammadzadeh, B. Dahrazma, S.F. Saghravani, P. Mohsenzadeh, A.H. Mahvi (2020) Removal of sulfate by electrocoagulation with discontinuous electrodes in a continuous system, Desalination Water Treat. 193:414–423. https://doi.org/https://doi.org/10.5004/dwt.2020.25817.

[40] A. Chibani, S. Ncib, A. Barhoumi, W. Bouguerra, E. Elaloui (2023) Box-Behnken design optimization of sulfate reduction from natural water by electrocoagulation process, Phosphorus Sulfur Silicon Relat. Elem. 198:164–171. https://doi.org/10.1080/10426507.2022.2134372.

[41] N.C. Archilha, C.S.A. Canto, S.M. Ratusznei, J.A.D. Rodrigues, M. Zaiat (2010) E. Foresti, Effect of feeding strategy and COD/sulfate ratio on the removal of sulfate in an AnSBBR with recirculation of the liquid phase, J. Environ. Manage. 91:1756–1765. https://doi.org/https://doi.org/10.1016/j.jenvman.2010.03.016.

[42] H.A. Moreno-Casillas, D.L. Cocke, J.A.G. Gomes, P. Morkovsky, J.R. Parga, E. Peterson (2007) Electrocoagulation mechanism for COD removal, Sep. Purif. Technol. 56:204–211. https://doi.org/10.1016/j.seppur.2007.01.031.

[43] S.A. Sajjadi, A. pakfetrat, M. Irani (2018) Removal of Remazol Black B Dye by Electrocoagulation Process Coupled with Bentonite as an Aid Coagulant and Natural Adsorbent, Iranian Journal of Health, Safety and Environ 5:1058–1065. http://www.ijhse.ir/index.php/IJHSE/article/view/327/pdf.

[44] I. Amri, Z. Meldha, S. Herman, D. Karmila, Mhd. Fadilah Ramadani, Nirwana (2023) Effects of electric voltage and number of aluminum electrodes on continuous electrocoagulation of liquid waste from the palm oil industry, Mater. Today Proc. 87:345–349. https://doi.org/https://doi.org/10.1016/j.matpr.2023.03.621.

[45] M.H.R.B. Khan, N. Islam, T. Ahmed, A. Ahsan, M. Al Farabe, M.A. Haque, K.T. Shabib, M. Shafiquzzaman, M. Imteaz, M.T. Alresheedi (2026) Optimization of voltage and retention time for enhanced textile wastewater treatment using electrocoagulation with stainless-steel electrodes, Clean Technol. Environ. Policy 28:1–21. https://doi.org/10.1007/s10098-025-03347-x.

[46] M. Nasrullah, M.N.I. Siddique, Z. Wahid (2014) Effect of High Current Density in Electrocoagulation Process for Sewage Treatment, Asian Journal of Chemistry 26:4281–4285. https://doi.org/10.14233/ajchem.2014.16134.