Performance Evaluation of a Combined Electrocoagulation–Electrooxidation Process for the Treatment of Petroleum Refinery Wastewater

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Abstract. The present study investigates the application of a combined electrocoagulation-electrooxidation (EC-EO) process for the treatment of wastewater generated from Al-Dewaniya petroleum refinery plant in Iraq. The EC-EO process was examined in terms of its ability to simultaneously produce coagulant and oxidant agents by using a parallel plate configuration system composed of stainless steel plates as cathode and pair of aluminum and graphite plates as anode at two different current concentrations (1.92A/l and 0.96A/l). The results showed that the best conditions for treatment of Al-Dewaniya petroleum refinery wastewater using the combined approach were current concentration of (0.96A/l), current density of (12mA/cm²), NaCl concentration of (2g/l), pH of (7), and electrolysis time of (60 min). In this case COD removal efficiency (93.75%), phenol removal efficiency (96.20%), TDS removal efficiency (6.88%) were obtained with lower specific energy consumption (29.45 kWh/kg COD) and lower aluminum consumption (0.587 x10⁻³ kg/h). The combined process showed to be better than EC process in term of COD and phenol removal efficiencies as well as aluminium consumption. In addition, it was better than EO in term of energy consumption. The combined process also gave buffering effect regarding to pH hence no need for controlling the pH during the operation.

1. Introduction
The treatment of wastewaters generated by petroleum refinery industry represents an essential environmental pollution challenge due to the large amount and the variety of contaminants existing in these effluents. Water utilize at large quantities especially in thermal cracking and catalytic cracking, distillation, desalting, hydro-treating, and cooling systems [1]. The generated wastewater from petroleum refining processes is approximately 1.6 times the quantity of the processed crude oil. Besides, around 80-90 % of the quantity of water utilized in the petroleum refinery releases as wastewaters, thus treatment of these effluents offers an economic benefit for oil refineries in addition to the avoiding of environmental pollution issues by reusing of water again [2]. Wastewaters from refinery processes are commonly contained high concentrations of aromatic and aliphatic hydrocarbon resulting in dangerous and harmful effects on the aquatic life, plants, and ground water sources [3]. Therefore, it is necessary to develop active and economical methods for treating petroleum refinery wastewater. Various approaches have been applied in this field like adsorption, coagulation, ultrafiltration, biodegradation, and electrochemical processes [3]. Electrochemical methods such as
Electrocoagulation (EC) and electrooxidation (EO) have the advantage of the minimum amount and number of chemicals can be used in addition to being easily controlled.

Electrocoagulation (EC) is based on coagulants generation via electrically dissolving anodic electrodes either aluminum or iron to their respective ions and production of hydrogen gas at the cathode. The metal ions create flocculates that trap pollutants then these flocculates are floated by hydrogen gas [4].

For instance, by applying a voltage or electrical current between aluminum anode and stainless steel cathode of an electrocoagulation cell, aluminum ions are produced due to the dissolution of aluminum anode (Eq.1). At the same time, oxygen liberates at the anode as a side reaction (Eq.2). While, at the cathode, hydroxyl ions generate due to the reduction of water with liberation of hydrogen gas (Eq.3). In the solution, hydroxyl ions react with aluminum ions to make flocs of amorphous Al(OH)$_3$(s)(Eq.4).

\[
\text{At the anode:} \quad (\text{Al})_s \rightarrow \text{Al}^{3+}(\text{aq}) + 3e^- \quad (1)
\]
\[
2\text{H}_2\text{O} \rightarrow O_2(g) + 4\text{H}^+ + 4e^- \quad (2)
\]
\[
\text{At the cathode:} \quad 2\text{H}_2\text{O} + 2e^- \rightarrow \text{H}_2 + 2\text{OH}^- \quad (3)
\]
\[
\text{In solution:} \quad \text{Al}^{3+} + 3\text{OH}^- \rightarrow \text{Al(OH)}_3(s) \quad (4)
\]

EC was applied effectively to treat textile wastewater [6], remove phenolic compounds [7], remove heavy metals [8], decolorize reactive dye solutions [9, 10], and clarify suspended clay solutions [11]. The advantages of EC involve higher removal efficiency, compact treatment facility, and complete automation with low cost. In this method; the treated water is colorless, odorless, clear and having low sludge formation in comparison with the chemical coagulation. The gas bubbles generate at the cathode surface permit easily separation the pollutants by floating. However, this method suffer from disadvantages like the requirements of regular replacement the sacrificial electrodes, reducing the efficiency due to formation of a resistant film layer on the cathode, and its deficiency for removal of tenacious dissolved organic pollutants [12].

Electrooxidation (EO) in the treatment of wastewaters can be carried out through various ways. For example, when ions of chloride exist in the solution, indirect electrooxidation process occurs in which the organic pollutants such as phenol, dyes, glucose, and aniline can be destroyed by chlorine and hypochlorite that generated anodically in the electrooxidation process via numerous reactions. Eqs.5-12 represents the anodic and cathodic reactions as well as the bulk reactions during the indirect electrooxidation[13,14]:

Anodic reactions:
\[
2\text{Cl}^- \rightarrow \text{Cl}_2 + 2e^- \quad (5)
\]
\[
2\text{H}_2\text{O} \rightarrow \text{O}_2 + 4\text{H}^+ + 4e^- \quad (6)
\]

Cathodic reactions:
\[
2\text{H}_2\text{O} + 2e^- \rightarrow 2\text{OH}^- + \text{H}_2 \quad (7)
\]
\[
\text{OCl}^- + \text{H}_2\text{O} + 2e^- \rightarrow \text{Cl}^- + 2\text{OH}^- \quad (8)
\]

Bulk reactions:
\[
\text{Cl}_2 + \text{H}_2\text{O} \rightarrow \text{HOCI} + \text{H}^+ + \text{Cl}^- \quad (9)
\]
\[
\text{Cl}_2 + \text{OH}^- \rightarrow \text{HOCl} + \text{Cl}^- \quad (10)
\]
\[
\text{HOCI} \rightarrow \text{H}^+ + \text{OCl}^- \quad (11)
\]
\[
\text{R} + \text{OCl}^- \rightarrow 2\text{CO}_2 + \text{Cl}^- + \text{H}_2\text{O} \quad (12)
\]

Organic Pollutants can also be destroyed through a process called direct anodic oxidation, in which contaminants can be degraded directly on anodes via producing physically adsorbed “active oxygen”.

In the Anodic oxidation, no chemicals or oxygen are added also no secondary pollutants could be produced as well as complicated accessories may not be required. In the anodic oxidation process, anode material is the most important component. The most anodes that have been used in the anodic oxidation are graphite, glassy carbon, carbon fiber, stainless steel, Ti/Pt–Ir, Ti/RuO\textsubscript{2}, and boron-doped conductive diamond (BDD) [15]. In spite of its highly cost, BDD has shown higher over potentials for oxygen and hydrogen evolution in addition to its stability and efficiency during the electroooxidation process [16]. These properties cause its successful application in oxidation of organic pollutants [17], electrosynthesis [18], decolorization of solutions [19], degradation of surfactant [20], and oxidation of benzoic and carboxylic acids [21]. However, in the presence of chloride ions, graphite is preferred for generation active chlorine due to its high overpotential for chlorine liberation in comparison with other anodes [22]. The advantages of EO method involve ease of operating conditions, simple equipment, total mineralizing of organic contaminants, low cost of electrode maintenance, and forming disinfecting compounds. However, EO method has some weaknesses like it’s ineffectiveness in removing of suspended solids, and the deficiency of the process due to formation of a resistant film layer on the cathode [23].

EC is considered as an efficient and fast technique for treating of wastewaters having suspended solids. However, it is an inefficient technique in the case of removing persistent dissolved organic pollutants. On the other hand, EO has the ability to degrade persistent organic pollutants completely but in the case of effluents containing suspended solids, it is a slow and energy-consuming method, hence a combination of these two processes gives a practical hybrid method that takes into account advantage of the synergistic effects [24, 25]. Applications of combined treatment have been successfully achieved in removal of different pollutants either by simultaneous EC and EO treatment with a suitable electrode configuration or by sequential EO treatment of an electro-coagulated effluent [25-29]. To the best of our knowledge, a combination of electrocoagulation (EC) and electro-oxidation (EO) in a batch electrochemical reactor for treating petroleum refinery wastewater has not been reported. In our previous work [30], we studied the application of electrooxidation using graphite anode and stainless-steel cathode for treating wastewater produced from Al-Dewaniya petroleum refinery plant. Our results showed good performance of the indirect electrooxidation process in removal of COD and phenol. However, pH was decreased to be acidic during the operation and a slightly removal of TDS was observed. Later on, we investigated the possibility of electrocoagulation process in removal of COD from the same source of wastewater using aluminum electrode as anode and stainless steel as cathode. Good performance was obtained for this system related to COD removal. However, phenol removal was not exceeded 70% and final pH was increased rapidly to be alkaline during the operation [31].

It could be exciting to combine the two processes (EC and EO) in-situ using the same reactor for removal both insoluble and soluble pollutants from petroleum refinery wastewater. Therefore, the main objective of this study was to develop a hybrid process called the EC-EO process that combines both the electrocoagulation and electro-oxidation processes, and to examine its performance in treating wastewater generated from Al-Dewaniya petroleum refinery plant in comparison with the electrocoagulation and electro-oxidation as individual processes. Aluminum and graphite were selected as electrode materials due to their low cost, availability, and efficiency [32].

2. Experimental work
A sample of petroleum refinery wastewater taken from the feeding tank to the biological treatment unit in Al-Dewaniya petroleum refinery plant was delivered and stored in a closed container at 4 °C until use. Table 1 shows the properties of this sample in addition to the properties of the effluent resulted from the biological treatment unit in the settling tank.
Table 1. Properties of the waste water generated in Al-Dewaniya petroleum refinery plant and the treated wastewater from the biological unit

| Parameter   | COD (mg/l) | pH | T.D.S (mg/l) | Cl- (mg/l) | SO4^2- (mg/l) | Turbidity NTU | Phenol (mg/l) |
|-------------|------------|----|--------------|------------|---------------|--------------|--------------|
| Feeding tank| 160        | 7.8| 1610         | 931        | 560           | 4.17         | 0.17         |
| Settling tank| 65         | 7.5| 1680         | 119        | 400           | 6.44         | (0.01-0.05)  |

A rectangular Perspex glass electrochemical cell having a working volume (1.25 L) was used for the electrochemical treatment experiments. It has dimensions (120 mm width x 120 mm length x 120 mm height) and provided with top cover (150 mm × 150 mm × 28 mm thickness) which contains slits for electrodes fixation as well as holes for inserting probes of the conductivity and pH meters. Figure 1 shows the schematic diagram of the electrochemical system.

Electrodes in the electrochemical cell were arranged and connected electrically in a parallel plate configuration where two anodes and three stainless steel cathodic plates have been utilized for current concentration 1.92A/l whereas one anode and two stainless steel cathodic plates have been used for current concentration 0.96A/l. In the electrocoagulation experiments, the anodes were aluminum plates with dimensions (130mm x 80mm x 3mm (thickness)). While in the electrooxidation experiments, the anodes were graphite plates with dimensions (130mm x 80mm x 3mm (thickness)). At the combined process, each anode was composed from pair electrodes; graphite plate with dimensions (130mm x 40mm x 3mm (thickness)) and aluminum plate with dimensions (130mm x 40mm x 3mm (thickness)). In each process, the stainless steel cathodic plate has dimensions of (130mm x 80mm x 3mm (thickness)). The inter-electrode gap was fixed at 15 mm.

A constant current throughout the experimental process was provided via a digital power supply UNI-T (UTP3315PF). For achieving a proper mixing, the sample of each experiment (1.25L) was agitated with a magnetic stirrer then the required amount of NaCl (2g/l) was added before transferring the solution to the electrochemical cell. NaCl was used to raise the conductivity of Al-Dewaniya petroleum refinery wastewater used in this research from 2.92mS/cm to 6.19 mS/cm by addition 2g/l.
NaCl hence decreasing the power consumption. It has been found that chloride ions could significantly reduce the adverse effect of other anions such as HCO$_3^-$, SO$_4^{2-}$[33]. All experiments were performed at constant temperature 30±2 °C using water bath (Memmert, type: WNB22, Germany). The pH of electrolyte was measured by pH meter (HNNA Instrument Inc, PH211, Romania) then adjusted for suitable experimental conditions using 1M NaOH or 1M HCl. TDS and conductivity were measured by using (HM Digital Inc. model COM-100, Korea). At every 10 minutes of the electrolysis time (60 min.), samples were taken and analyzed to knowledge chemical oxygen demand (COD) and phenol concentration. COD determination was achieved by taken a sample (2ml) of the effluent then digested by an oxidizing agent (K$_2$Cr$_2$O$_7$) for 120 min at 150°C in a COD thermos-reactor (RD-125, Lovibond). The digested sample was then cooled down to room temperature and analysed in spectro-photometer (MD-200, Lovibond). Phenol concentration was determined based on Method 8047 assigned by Hach Company/Hach Lange GmbH, USA [34]. Each run was repeated twice, and the mean values of COD and phenol concentrations were taken in this work.

The removal efficiencies of COD and phenol were calculated according to Eq. 13, where $C_i$ is the initial concentration (mg L$^{-1}$) and $C_f$ is the final concentration (mg L$^{-1}$) [35]:

\[
RE\% = \frac{C_i - C_f}{C_i} \times 100
\]  

(13)

The specific energy consumption (SEC) represents the amount of the consumed energy in the process for a kilogram of COD that requires digesting. EC in (kWh/kg) may be acquired with the use of eq. 14 [36]:

\[
SEC = \frac{E.I.t \times 1000}{(COD_f - COD_i) \times V}
\]  

(14)

Where SEC represents the specific energy consumption (kWh/kg COD), E represents the applied cell voltage (Volt), t represents the electrolysis time (h), I represents the current (A), V represents the volume of the effluent(L), and COD$_f$ and COD$_i$ represent the final and initial chemical oxygen demand (mg L$^{-1}$). Aluminium consumption was calculated by taking the difference in weights of the aluminium electrode before and after each run.

3. Results and discussion

In our two previous works [30, 31] for the treatment of the same sample of petroleum refinery wastewater using electrooxidation (EO) process alone in the first work and electrocoagulation (EC) process alone in the second work, we found that the preferred conditions were similar in each processes which gave higher removal of COD. They were current density (12mA/cm$^2$), NaCl concentration (2g/l) and pH (7). Based on these preferred conditions, the combined experiments at two values of current concentrations (1.92A/l and 0.96 A/l) were performed.

3.1. Comparison among the processes at current concentration 1.92A/l (two anodes/ three cathodes) Figure 2 displays the abatement of COD with time for the three operations (EC, EO, and EC-EO) at current concentration(1.92A/l) using effluent with COD (160 ppm) and addition of NaCl at a concentration of (2g/l) then applying a current density of (12mA/cm$^2$) for 60min. It was observed that the electrooxidation process gives higher abatement of COD followed by the combined (EC-EO) process then electrocoagulation process.

Figure 3 shows the pH profile in each process during the electrolysis operation. It can be seen that pH value was maintained approximately constant in the combined process while it was increased gradually in the electrocoagulation and decreased during the first 30min then increased beyond this period with respect to electrooxidation process. Hence the combined process has the effect of buffering the pH during the electrolysis which added further advantage of using the combined approach. Similar observation was found by Redah [37] in their work on the treatment of fuel washing wastewater taking from south Baghdad gas power station/2 by combined electrocoagulation-electrooxidation followed by electrodialysis process. Similar trend was also observed by Daghrir, et al.[38] in their work for treatment of restaurant wastewaters by hybrid process combining electrocoagulation and electro-
oxidation processes. Increasing of pH with time during the electrocoagulation was observed also by previous studies [39, 40]. Similar trend of pH variation during electrooxidation was also observed by Souza and Ruotolo [41] in their work on the electro-chemical treatments of effluents of oil refineries with the use of the boron-doped diamond anodes. TDS profile during the electrolysis is depicted in Figure 4 where electrocoagulation gave higher removal of TDS in comparison with other processes.

![Figure 2](image2.png)

**Figure 2.** COD profile with time for the three different process at current concentration 1.92A/l

![Figure 3](image3.png)

**Figure 3.** pH profile with time for the three different process at current concentration 1.92A/l

Table 2 illustrates the removal efficiencies of COD, phenol, and TDS for each process. It can be seen that electrooxidation process gives higher removal efficiencies of COD and phenol while electrocoagulation gives higher removal of TDS. The results showed that the combined process has the intermediate effect between each single process. However, using electrocoagulation alone will not lead to a complete removal of phenol where its removal efficiency had not exceeded 76.47%. Using a
combined process has another advantage where more TDS removal could be obtained in comparison with electrooxidation. Similar behaviors were observed by Redah [37]. Results showed that EO has higher specific energy consumption while EC has the lower value. EC-EO has the intermediate value (63.26 kWh/kg COD) with aluminum consumption lower than EC. From an economic point of view, the combined approach is better since the final COD and phenol concentrations are below the allowable limits for environment discharge [30].

![Figure 4. TDS profile with time for 3 distinct processes at current concentration 1.92A/l.](image)

### Table 2. TDS, COD, and phenol removal efficiencies for each EC, EO, and EC-EO processes at current concentration (1.92A/l)

| Type   | TDS% at 60min | COD RE% at 60min | COD<sub>i</sub> (ppm) | phenol | SEC (kWh/kg COD) | Al Consumption (kg/h) |
|--------|---------------|------------------|------------------------|--------|-----------------|-----------------------|
| EC     | 14.7          | 96.875           | 5                      | 0.17   | 0.04            | 76.47                 |
| EO     | 5.97          | 100              | 0                      | 0.17   | 0.0007          | 99.60                 |
| EC-EO  | 10.15         | 97.5             | 4                      | 0.17   | 0.004           | 97.65                 | 63.26                |
|        |                |                  |                        |        | 48.31           | 1.916 x10⁻³            |
|        |                |                  |                        |        | 72              | -----                 |
|        |                |                  |                        |        | 1.153 x10⁻³     |                       |

3.2. Comparison among the processes at current concentration 0.96A/l (one anode/ two cathodes)

Figure 5 illustrates the abatement of COD with time for the three operations (EC, EO, and EC-EO) at current concentration (0.96A/l) using effluent with COD (160 ppm) and addition of NaCl at concentration of (2g/l) and operating at constant current density of (12mA/cm²) for 60min. It is clear that electrooxidation process gives higher abatement of COD followed by combined (EC-EO) process then electrocoagulation process. However, the profile of each process are different from those observed at current concentration of 1.92A/l. Similar observations with respect to pH, and TDS profiles were obtained as in the case of higher current concentration (1.92A/l) as shown in Figures 6,7, respectively. Table 3 reports the removal efficiencies of COD, phenol, and TDS for each process. It was observed that electrooxidation process gives higher removal efficiencies of COD and phenol while electrocoagulation gives higher removal of TDS. The results showed that the combined process has the intermediate effect between each single process in a similar behavior observed at current concentration of (1.92A/l). Also using electrocoagulation alone will not lead to the complete removal of phenol where its removal efficiency had not exceeded 73.53%. It was concluded that increasing the current...
concentration will not lead to achieve a complete removal of phenol using electrocoagulation alone. Therefore the combined system is the preferred from environmental and economic issues (lower energy consumption than electrooxidation with high removal efficiency).

Figure 5. COD profile with time for the three different process at current concentration 0.96A/l

Figure 6. pH profile with time for the three different process at current concentration 0.96A/l

Results showed that EC-EO process operated at current concentration of 0.96 A/l has lower specific energy consumption as well as lower aluminum consumption in comparison with that operated at 1.92 A/l. In comparison with EC, the combined process is better in term of phenol removal and aluminum consumption with a slightly increase in the specific energy consumption. results of the final COD and phenol concentration using either current concentrations are better than the performance of the biological treatment unit (Table 1) hence adopting the present system is a promising step for treating the effluents of Al-Dewaniya petroleum refinery plant at low cost.
Figure 7. TDS profile with time for three distinct processes at current concentration 0.96A/l.

Table 3 TDS, COD, and phenol removal efficiencies for each EC, EO, and EC-EO processes at current concentration (0.96A/l)

| Type   | TDS% at 60min | COD RE% at 60 min | CODf (ppm) | phenol | SEC (kWh/kg COD) | Al Consumption (kg/h) |
|--------|---------------|-------------------|-------------|--------|------------------|-----------------------|
| EC     | 9.43          | 90.63             | 15          | 0.17   | 0.045            | 73.53                 | 25.16                 | 0.97 x10^{-3}         |
| EO     | 4.56          | 96.87             | 5           | 0.17   | 0.0015           | 99.12                 | 35.31                 | -----------           |
| EC-EO  | 6.88          | 93.75             | 10          | 0.17   | 0.0065           | 96.20                 | 29.45                 | 0.587 x10^{-3}       |

4. Conclusions
The present results showed that the combination of the two processes (electrocoagulation and oxidation) in situ gave many advantages firstly COD and phenol removal efficiencies in the combined process were better than EC process alone, secondly TDS removal efficiency for the combined was higher than EO process alone, thirdly energy consumption for the combined was lower than EO as well as the aluminum consumption was lower than EC. Finally, the combined process gave buffering effect regarding to pH hence no controlling on the pH during the operation is required. Accordingly, it can be concluded that EC-EO process can be considered as an efficient alternative method for treatment of petroleum refinery wastewater.

Using a combined approach with current concentration of (0.96A/l), current density of (12mA/cm2), NaCl concentration of (2g/l), pH of(7), and electrolysis time of (60 min) lead to get COD and phenol removal efficiencies 93.75%,96.20% respectively with specific energy consumption of (29.45 kWh/kg COD) and aluminum consumption of (0.587 x10-3kg/h). Therefore, application of EC-EO process with current concentration of 0.96 A/l is recommended for treating the effluent from Al-Dewaniya petroleum refinery plant in Iraq at low specific energy consumption and low consumption of aluminum with good removal efficiencies of COD and phenol.

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