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Research Article

Electrochemical Removal Of Oil From Oil Refinery Wastewater.

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Abstract

Recently, COD and oil concentrations in the water have increased, as a result of reduced water volumes and increased industrial waste being dumped into the river. Increased concentrations of these pollutants lead to health and environmental problems. As the water treatment plants use the usual methods of water treatment and could not reduce the concentration of oil and COD to the limit set by the World Health Organization, so an effective way to treat these pollutants became absolutely necessary. In this study, electrochemical method was used to treat water contaminated with oil and COD, using aluminum and iron electrodes as positive electrode and cathode electrode. A coagulation cell with a volume of 2 liters was also used in the work. Several factors affecting the process of treating oil and carbon dioxide pollutants were studied, and these factors were as follows: Submerge depth, Number of electrodes, the distance between the electrodes. From the results, it was found that the optimum removal was 94.2% for oil, and 99.5% for COD. It was achieved when the number of electrodes was 4 aluminum and 4 iron, the distance between the electrodes was 2 cm, the depth was 12 cm, and the function variables were acidic equal to 7, time 50 min, and voltage 10.5 V. The concentration of sodium chloride is 0.5 g/l and the electrode material is aluminum as anode and cathode and the initial oil concentration is 95 mg per liter, initial COD concentration is 710 mg per liter and the consumption of energy was estimated to be 12 kWh/m, the TSS was 73 mg/l . The results demonstrated that the electrochemical is a feasible technique for treatment of oily contaminated petroleum refinery wastewater.

Keyword: oil , COD , removal , oily wastewater , Electrochemical.

1. Introduction

Large amounts of water are used by petroleum refineries for various purposes, such as desalting, distillation, and hydro treating (Yavuz et al., 2010); hence, facilities create a huge quantity of wastewater. The characteristics of the wastewater generated by petroleum refineries depend on the process configuration (Rasheed et al., 2011). For instance, the high concentrations of aromatic petroleum hydrocarbons and aliphatic acid in the wastewater are often the result of (El- Naas et al., 2009 and Sun et al., 2008). the refining processes the refinery effluents are often contaminated with various types of toxic substances, such as cyanide, oil, benzene, and sulfide. These substances can lead to harmful effects on plants. The wastewater is also very high in chemical oxygen demand (COD) and biological oxygen demand (BOD) (Rasheed et al., 2011 and Verma et al., 2013), They typically have negative impacts on plants, the ocean, rivers, and sources of surface and ground water (El- Naas et al., 2009 and Sun et al., 2008). Prior to the final biological treatment and purification stage, facilities are required to lower the concentration of organic pollutants in their wastewater to a value sensitive to microbial destruction. These pretreatment units typically include adsorption, coagulant and coagulant aides, ultrafiltration, and electrochemical methods (El- Naas et al., 2009). Electrochemistry (EC) is one of the new methods of water and wastewater treatment (Chafi et al., 2011). The EC process has several advantages, such as easy handling. short treatment

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time, low sludge formation and no chemicals required (Gengec et al., 2012). EC is a technology effectively used to treat refinery effluents containing various pollutants such as arsenic, phosphate, boron, dyes and viruses. In addition, it was effective in breaking up emulsions in water and paper, mill effluents, landfill leachate and dairy effluents (Bensadok et al. 2008; Irdemez et al. 2006; Katal et al., 2011; Khoufi et al. 2007; Li et al. 2011; Azadi Aghdam et al. 2016; Phalakornkule et al. 2010; Sengil .2006: Tchamango et al. 2010; Wan et al. 2011; Will Maz et al. 2008; Amani Ghadim et al. 2013).Few studies have explored the removal of hydrocarbons using the EC process. EC procedure used for the removal of hydrocarbons from contaminated groundwater. Effects of pH, electrode material, and treatment time are investigated, and the removal efficiency of 99.5 % of was achieved in no significant difference was found when using stainless steel or aluminum electrodes. In an EC process, coagulant ions are generated in situ, with three steps involving (a) electrolytic reactions at electrode surfaces, (b) formation of coagulants in aqueous phase, and (c) adsorption of soluble contaminants or colloidal on coagulants, which are removed either carried out sedimentation or flotation (Kobya et al., 2003). Iron (Fe) and aluminum (Al) have been widely used as sacrificial electrodes in the EC process (Gengec et al.,2012 and Kobya et al., 2006). Sacrificial electrode dissolves from the anode creating corresponding metal ions. These ions hydrolyze to polymeric iron or aluminum ox hydroxides, which are the coagulating agents (Gengec et al., 2012). The electrocoagulation was more effective to eliminate TDS and presented higher removal of turbidity compared to chemical coagulation. Based on the analysis, the electrocoagulation is more efficient that the textile effluent treated can be reused or rejected without risk in the environment.(Gzar et al., 2020).

Electrochemical (EC) is one of the novel methods for water and wastewater treatment. Treatment of wastewater by EC has been practiced for most of the 20th century with limited success and popularity. In the last decade, this technology has been increasingly used in South America and Europe for treatment of industrial wastewater containing metals. It has also been noted that in North America EC has been used primarily to treat wastewater from pulp and paper industries, mining and metal- processing industries. In addition, EC has been applied to treat water containing foodstuff wastes, oil wastes, dyes, suspended particles, chemical and mechanical polishing waste, organic matter from landfill leachates, defluorination of water, synthetic detergent effluents, mine wastes and heavy metal-containing solution. Typically, empirical studies are done on EC to define major operating parameters for broad classes of contaminated water or waste streams (Mollah, et al., 2001).In the present study

Study the removal oil and COD of oily wastewater by using an electrochemical, Aluminum and iron electrodes are used to represent the anode and cathode of an electrochemical cell,These two types of electrodes are arranged in a way that allows them to form an electrochemical cell,For each arrangement of electrodes, the removal efficiency of COD and oil has been studied and Various parameters such as pH, time, voltage, NaCl concentration, and the initial concentration were studied to determine best removal conditions.

3. Results and Discussion:

The Parameters affecting Electrochemical are:

3.1 Effect of pH

The effect of pH on COD and oil removal efficiency is shown in Figure (1). The maximum removal efficiencies of oil and COD are 90.5 % and 93.3% respectively.

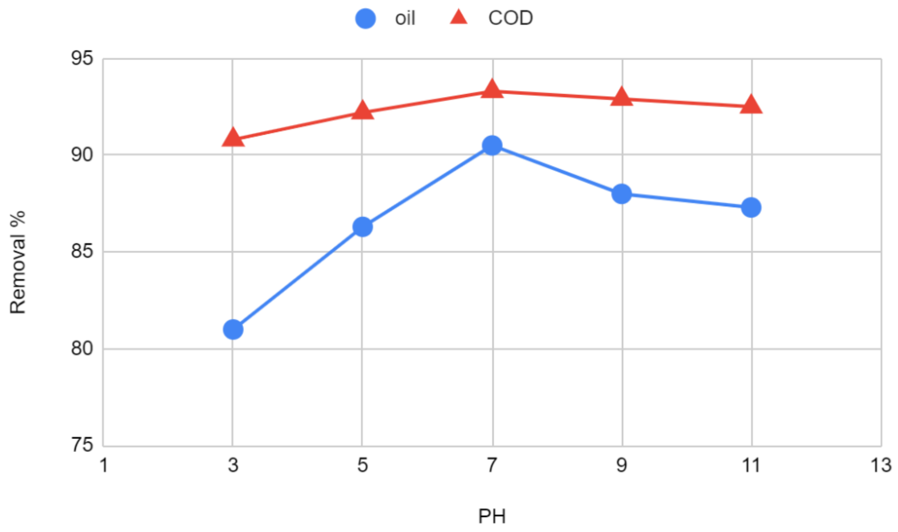

Figure 1: Effect of pH on the removal of COD and oil.

It is obvious that oil and COD removal effectiveness were significantly affected by pH value, an increase in pH value can be noticed when pH value was acidic in the carried out tests. This increment can be attributed to the emission of oversaturated carbon dioxide due to the development of hydrogen at the cathode . Furthermore, one can notice that in basic conditions, pH value of the sample reduced throughout treatment process, this took place because of hydroxide ions precipitation with the cation. The aforementioned results emphasized that EC process can be applied as pH buffer. The obtained removal efficiencies of oil and COD reduced in basic and acidic conditions. The obtained result was demonstrated by arrangement of ionic species of iron. These species were important in removing the contaminants that are insoluble and stable. Due to the predominance zone diagram of Fe (III)(Barrera-Diaz et al., 2012) , at acidic conditions the species of Fe^{3+}, $Fe(OH)^{+2}$, $Fe(OH)^{+2}$ ions are dominant and at alkaline condition $Fe(OH)^{-4}$ ion these species are soluble; moreover, the solubility removal of COD and oil . However, in neutral pH, Fe $(OH)_{3(s)}$ is stable, insoluble and available for pollutant adsorption from wastewater. Thus, Fe $(OH)_{3(s)}$, has the major role in the removal of COD and oil. The ingredients of molecules of hydrocarbon are nonpolar neutral and hydrophobic, so they can be eliminated from wastewater by generation of surface complexes by precipitate of gelatinous $Fe(OH)_{3(s)}$. Active zones on complex hydrocarbon molecules provide faces for flocs of ferric hydroxide that grow (co-precipitation) and undergo physical adsorption onto the amorphous $Fe(OH)_{3(s)}$ flocs using van der Waals forces. Based on the following reactions, the mechanism involved in the removal of petroleum hydrocarbons can be simplified Formation of ferric hydroxide (at $6 < pH < 9$):

$Fe^{3+} + 3\,OH^{-} \rightarrow Fe\,(OH)$ (1)

Adsorption of hydrocarbons (HC):

$Fe\,(OH)_3 + HC \rightarrow (Fe\,(OH)_3\text{-}HC)_{floc}$ (2)

$(Fe(OH)_3$ -HC) flocs can attach together and flocculate; thus, contaminants are separated from the wastewater by settling, flotation or filtration (Moussavi et al ,2011). This is consistent with finding of Farhadi et al (2012). The higher removal effectiveness occurred when distance distance between the electrodes is 2 cm. The highest removal at lowest distance Because the force of attraction between the particles is low and it takes longer time for the pollutant particles to unite with the coagulant particles and to form clumps. The results agreed with Abdulkhadher et al (2021) and Song et al (2007). Increasing number of electrode (4 electrodes Al and 4 electrodes Fe) led to increasing removal efficiency of pollutants because the number of coagulants generated increases with the increase of electrodes . Higher removal efficiencies in the experiments were occurred when electrode submerged depth 12 cm because the surface area increases, the coagulant dissolved from the electrode increases. Energy consumption and quantity of the produced sludge is shown in figure (2) . The maximum Sludge amount and energy consumption are 55 mg/l and 0.89 kwh/m^3 at pH value 11.

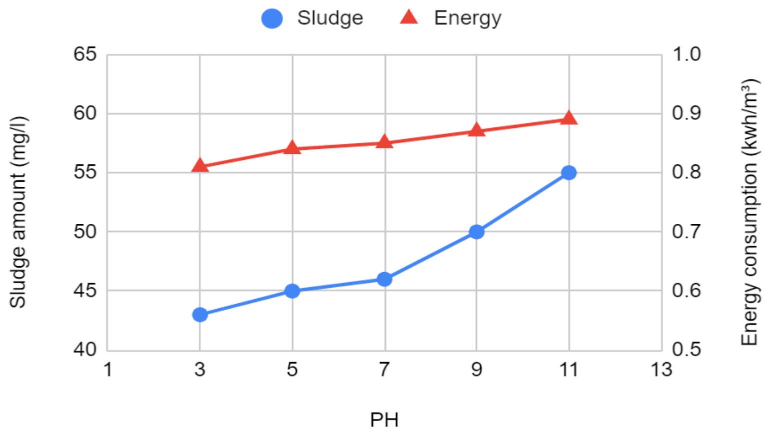

Figure 2: The effect of pH on the sludge amount and energy consumption.
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3.2 Effect of time on electrochemical

The effect of time on COD and oil removal efficiency was investigated in the EC process, and it is shown in Figure (3).

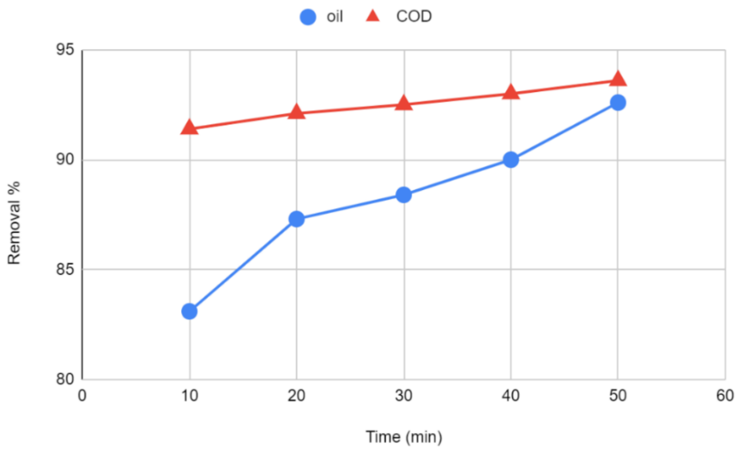

Figure 3: Effect of contact time on the removal of COD and oil.

The maximum removal efficiencies of COD and oil were 93.6 % and 92.6% respectively at 50 min time.

The electrolysis time determines the rate at which metal ions are produced from the electrodes. COD and oil removal efficiencies are directly dependent on metal concentration ions produced by the electrodes. This subject is justified by Faraday's law. According to Faraday's law (equation 4.3), theoretically, the amount of coagulant is directly proportional to the current applied to the electrolytic cell and time.

$$C = \frac{MIt}{n\,F\,V} \quad (3)$$

where *M*, *I*, *t*, *n*, *F* and *V* stand for the molecular weight of the metal (g/mol), and the current (A), time (s), metal valency (3 for Al and 2 for Fe), Faraday's constant (96500 C/mol) and wastewater volume, respectively. At the beginning of the EC process, in the experiments the COD and oil removal efficiency is low since the rate at the initial times the production of metal ions from the electrodes is low; However, when the time of electrolysis increases, the concentration of metal ions and their hydroxide clusters increase; thus, COD and oil removal efficiencies increase (Ji et al., 2015) . electrolysis is required to generate adequate amount of Al^{+3} ions and aggregated flocs need the time to be formed. It is well known that the electrolysis time is proportional with coagulant concentration in the reaction medium which leads to higher pollutant removal (Redah et al., 2016). The effect of time on the amount of sludge produced and energy consumption is shown in Figure (4). The maximum Sludge amount and energy consumption are 60 mg/l and 3.32 respectively at time 50 min.

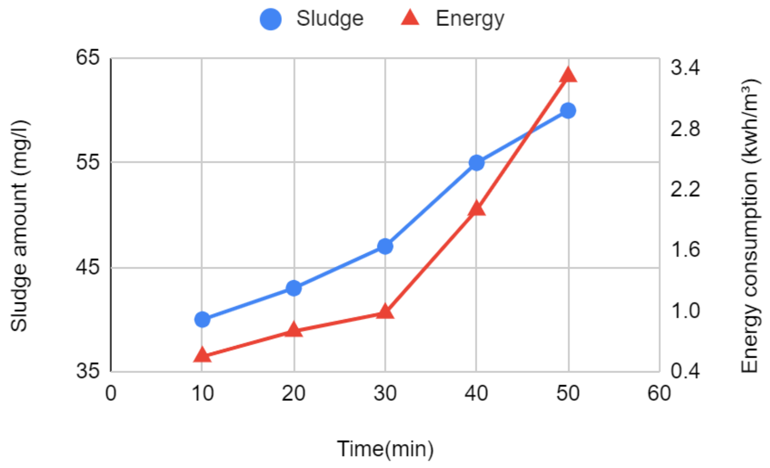

Figure 0: The effect of time on the sludge amount and energy consumption.

The results showed that sludge production and energy consumption were proportional to time. According to Faraday's law the amount of formation of metal hydroxide and coagulants is directly proportional to time; Thus, with increasing time, the resulting sludge increases. energy consumption is directly proportional to time; Thus, by increasing the time, the energy consumption is increased. In addition, the amount of sludge generated and energy consumption increase by increasing the removal of COD and oil.

3.3 Effect of voltage on electrochemical

The voltage applied to the EC system determines the coagulant dosage rate. The effect of voltage on the removal efficiency of COD and oil is shown in Figure (5). The maximum removal efficiencies of COD and oil at voltage 10.5 v are 93.6% and 93.9% respectively.

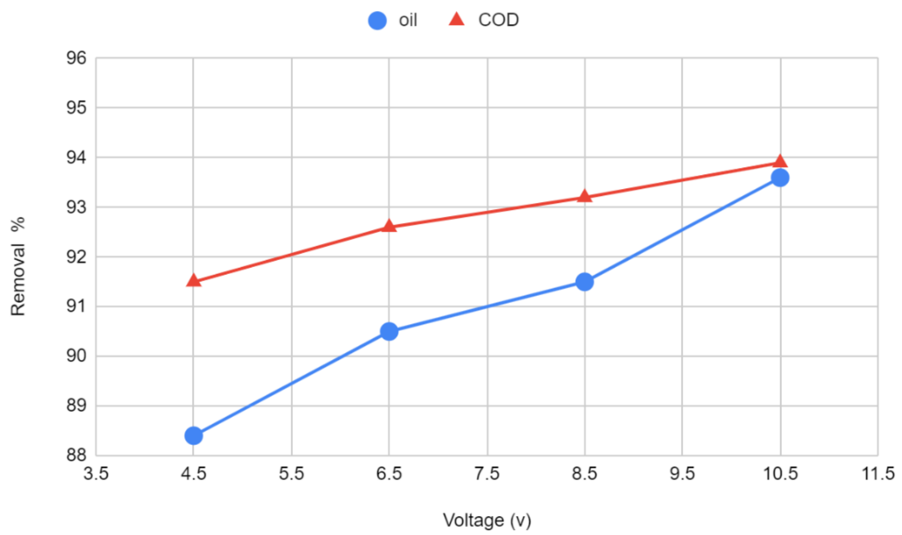

Figure 4: Effect of voltage on the removal of COD and oil.

The removal efficiencies of COD and oil increases with an increase in voltage. The applied voltage is an important parameter for the COD and oil removal since it determines the coagulant dosage rate, concentration of metal ions and their hydroxide flocks. When voltage increases, the concentration of metal ions and their hydroxide flock increase to absorb hydrocarbons. That subject is justifiable by Faraday's law. Thus, the COD and oil removal efficiencies are increased. In addition to decreasing bubble size, the bubble generation rate increases with the increasing of voltage, which is beneficial for pollutants removal agreed with (farhadi et al ., 2012). Since this has caused that the contact between hydroxide flock and pollutants increases, the flotation process is improved and pollutants are rapidly removed. These results agreed with (Safari et al., 2016). The effect of voltage on the amount of sludge produced and consumption of energy is shown in Figures (6). The maximum sludge amount and energy consumption at voltage 10.5 v are 68 mg/l and 6.9 kwh/m^3 respectively.

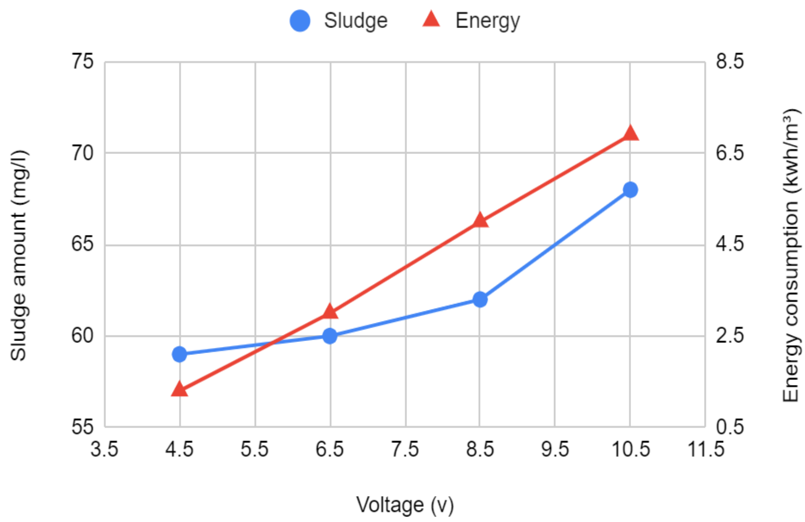

Figure 5: The effect of voltage on the sludge amount and energy consumption.

The results have shown that sludge production and consumption of energy were proportional to voltage According to Faraday's law the amount of metal hydroxide formation and coagulant is directly proportional to voltage; thus, by increasing voltage, sludge produced is increased, according to:

$$E= \frac{V.I.t}{Treated\ volume\ (l)} \qquad (4)$$

consumption of energy (E) is directly proportional to voltage; thus, by increasing voltage, consumption of energy is increased. In addition, the amount of sludge produced and consumption of energy increase by increasing the COD and oil removal. These results agreed with (Safari et al., 2016).

3.4 Effect of supporting electrolyte on electrochemical

In this section, different sodium chloride concentrations were used to evaluate the effect of the solution conductivity on EC. It should be noted that the solution conductivity affects the cell voltage, current efficiency and consumption of energy in electrolytic cell. The effect of supporting electrolyte on the removal efficiency of COD and oil is shown in Figure (7). NaCl concentration was set in the range from 0.25 to 1.25 g/L. the removal efficiency of COD and oil was in four Experiments when we used NaCl concentration 1.25g/l it was removal efficiency of COD and oil very high Hence, the experiments were performed at 0.5 g/L NaCl due to the negligible increase in removal.

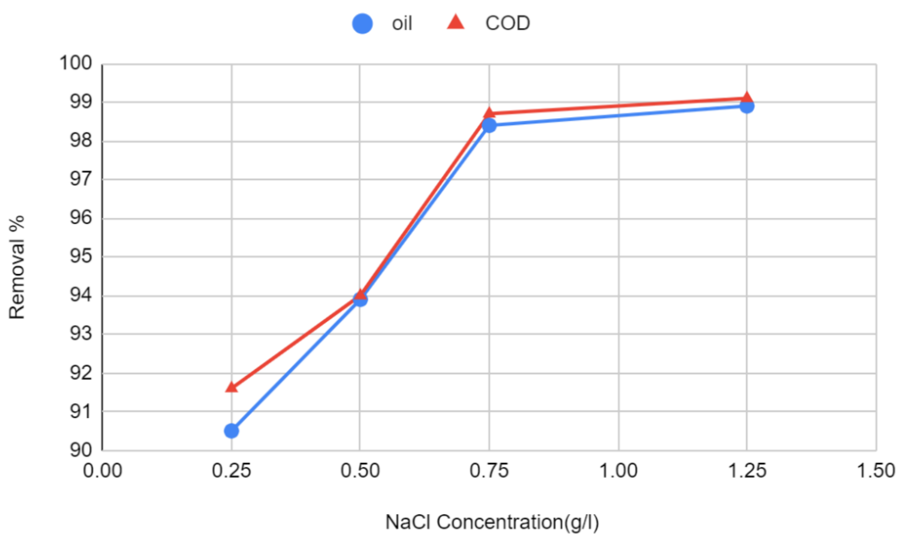

Figure 6: Effect of NaCl concentration on the removal of COD and oil.

The supporting electrolyte concentration of 0.5 g/L gave maximum removal efficiencies of COD and oil 94.1% and 93.9 % respectively.
The experiments were conducted at 0.5 g/L of NaCl which gave the best removal of pollutants. Very low removal is observed without adding NaCl to the solution, which is due to the fact that high electric conductivity is needed in order to use the EC is potential to remove the pollutants which oil refinery wastewater does not possess it. NaCI solution was selected as an electrolyte since it has several advantages, i.e., chloride ions could significantly reduce the adverse effects of other anions such as HCO and SO. The presence of the carbonate ion would lead to the precipitation of Ca^{2+} or Mg^{2+} ions that form an insulating layer on the surface of the electrodes. This insulating layer would sharply increase the ohmic resistance of the electrochemical cell and result in a significant reduction in the current efficiency and treatment conversion. Hence, it is suggested that among the present anions, there should be 20 % CI to ensure a normal operation of EC in water treatment (parsa et al., 2011). Conductivity causes an increase in current density; thus, more amount of coagulant can be introduced to the media. In addition, NaCl causes an increase in CI ions that chloride ions can remove the formed passivation layer on electrode surface. Thus, availability of metal hydroxide in the solution leads to an increase in the COD and oil removal efficiency.
Molecular chlorine is produced during the electrolysis is of chloride salts:

$$2Cl \rightarrow Cl_2 + 2\ e^- \qquad (4)$$

The produced molecular chlorine can then be hydrolyzed to hypochlorous acid and hypochlorite ions that these species are responsible for pollutants removal due to their high oxidative potentials(Janpoor et al., 2011).

$$Cl_{2\ (g)} + H_2O \rightarrow HOCI + H^+ + CI^- \qquad (5)$$

$$HCIO \rightarrow CIO^- + H^+ \qquad (6)$$

This is consistent with findings of Abdelwahab et al (2009) , when supporting electrolyte increases during the operation the removal efficiency is increased. The effect of supporting electrolyte concentration on the amount of sludge produced and consumption of energy is shown in Figure (8).

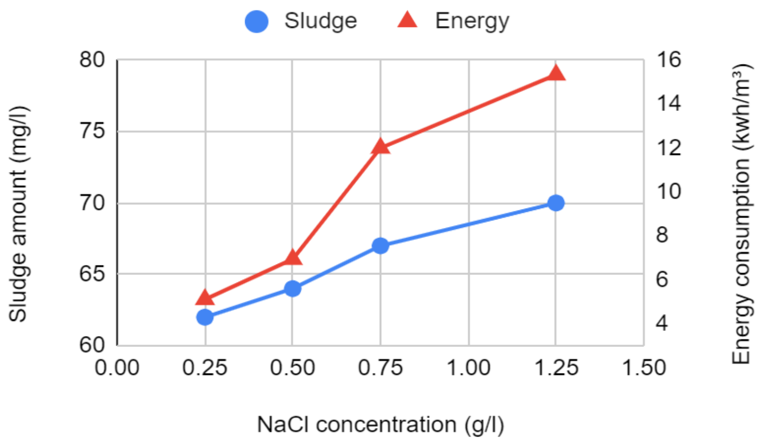

Figure 7: Effect of NaCl concentration on the sludge amount and energy consumption.

The maximum sludge amount and maximum energy consumption when using1.5 g/L NaCl concentration, are 70 mg/l and 15.33 kwh/m^3.
The results have shown that sludge production and consumption of energy are proportional to supporting electrolyte concentration. By increasing supporting electrolyte concentration, current and conductivity are increased during the process. Therefore, the amount of metal hydroxide formation, sludge production and consumption of energy are increased. In addition, the amount of sludge produced increase by increasing the COD and oil removal. These results agreed with (Safari et al., 2016).

3.5 Effect of electrode material on electrochemical

Aluminum and iron are cheap, available and proved to be effective; hence, they were applied in the current study. The effect of electrode material on the removal efficiency of COD and oil is shown in Figure (9).

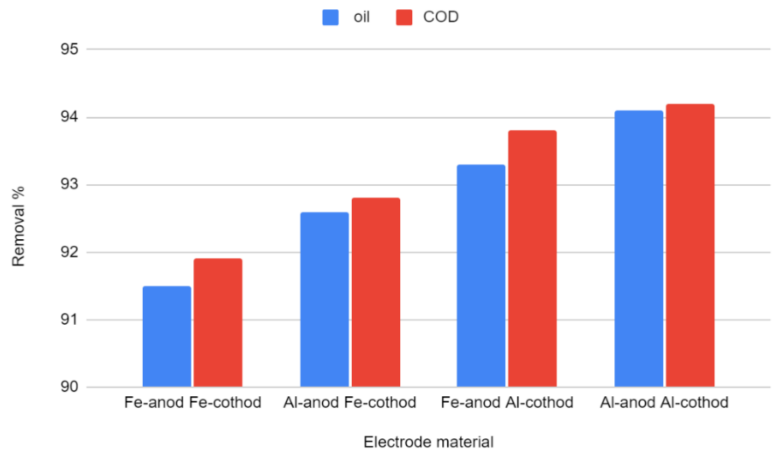

Figure 8: Effect of electrode material on the removal of COD and oil.

When the anode and cathode electrodes are both aluminum best removal efficiencies of oil and COD are 94.1 % and 94.2 % respectively.
The results reveal that the aluminum electrode as anode and cathode has the highest effect on reduction of COD and oil from wastewater. Iron electrode as sacrificial anode appeared greenish at first resulting from Fe^{+2} ions and then turned into yellow resulting from Fe^{+3} ion in the effluent. During the electrolysis of iron electrode, Fe^{+2} is the common ion produced. It can be oxidized easily into Fe^{3+} using dissolved oxygen in water. Moreover, there is Fe^{+2} in yellow particles of Fe (OH)3, and it is difficult to be settled. Furthermore, iron electrode becomes corroded at open circuit; therefore, the iron electrode is not preferred. Furthermore, effluent with aluminum electrodes was

found very clear and stable. Conductivity of aluminum electrode is more than iron electrode, which results in greater formation of coagulant of aluminum into the media. Hydroxide of aluminum is more stable than hydroxide of iron and also absorption strength of hydroxide of aluminum is more than hydroxide of iron. Hence, results using aluminum electrode for anode and cathode chosen comparing to other electrode sets, agreed with (Safari et al., 2016, Fadali et al., 2015 and Amani et al., 2014). The effect of electrode material on the amount of sludge produced and consumption of energy is shown in Figure (10).

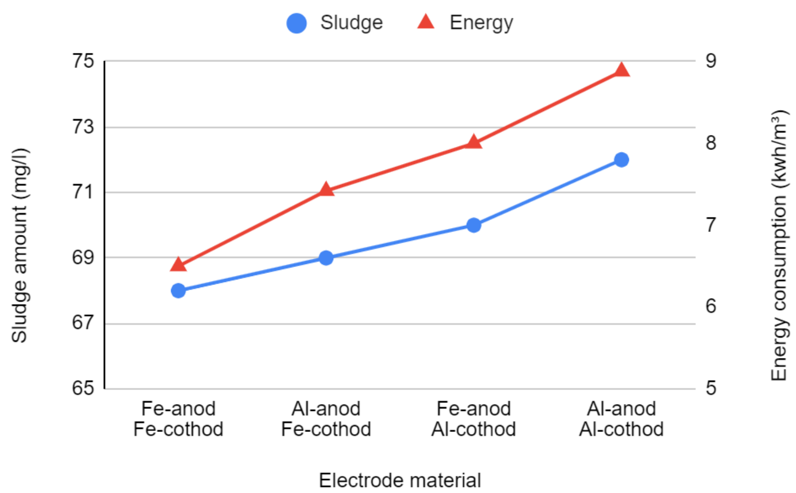

Figure 9: Effect of electrode material on the sludge amount and energy consumption.

The maximum sludge amount and maximum energy consumption when using aluminum electrodes as anode and cathode are 72 mg/l and 8.88 kwh/m^3.

The sludge formed and consumption of energy by aluminum electrodes were more than the sludge formed and consumption of energy by iron electrodes. As a result, the conductivity and coagulant formation by aluminum electrode are greater than those by iron electrode, this results agreed with (Safari et al., 2016).

3.6 Effect of initial concentration on electrochemical

The effect of initial oil and COD concentration (Co) on the removal efficiencies of COD and oil are shown in Figures 11 and 12.

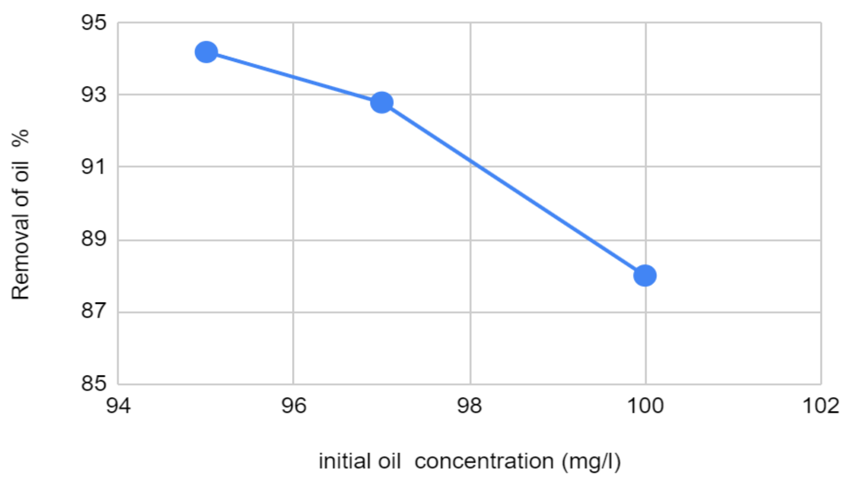

Figure 10: Effect of initial oil concentration on the removal of oil.

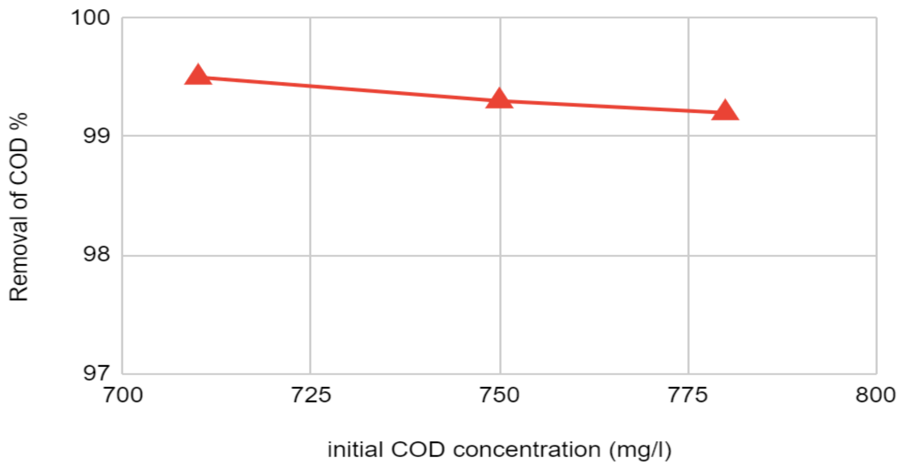

Figure11: Effect of initial COD concentration on the removal of COD.

In the experiments the Removal efficiency for higher concentration of oil and COD is lower compared to its lower concentrations when using the conditions, which are optimized for lower concentrations. Since the formation amounts of the coagulant are insufficient, the EC process needs more time and voltage to achieve the same removal efficiency as concentrations is lowered. This is consistent with the studies of (Canizares et al., 2007and Safari et al., 2016). COD removal efficiency from breakup of oil-in-water emulsions. They observed that the COD and oil removal efficiency was decreased when COD and oil concentration is increased.

The initial concentrations with removal efficiencies of COD and oil for the four experiments are presented in table 11

Table 11: The initial concentrations with removal efficiencies of COD, oil, sludge and energy.

Co oil	Co COD	PH	Time (min)	Voltage (V)	NaCl concentration (g/l)	Electrode material	C_e oil(mg/l)	Removal of oil (%)	C_e COD (mg/l)	Removal of COD (%)	Sludge (mg/L)	Energy (kwh/m³)
95	710	7	50	10.5	0.5	Al-anode Al-cathode	5.5	94.2	3	99.5	73	12
97	750	7	50	10.5	0.5	Al-anode Al-cathode	9	92.8	5	99.3	77	16.87
100	780	7	50	10.5	0.5	Al-anode Al-cathode	12	88	6	99.2	82	17.2

The results in Figure (13) have shown that sludge production and consumption of energy were proportional to the initial oil concentration.

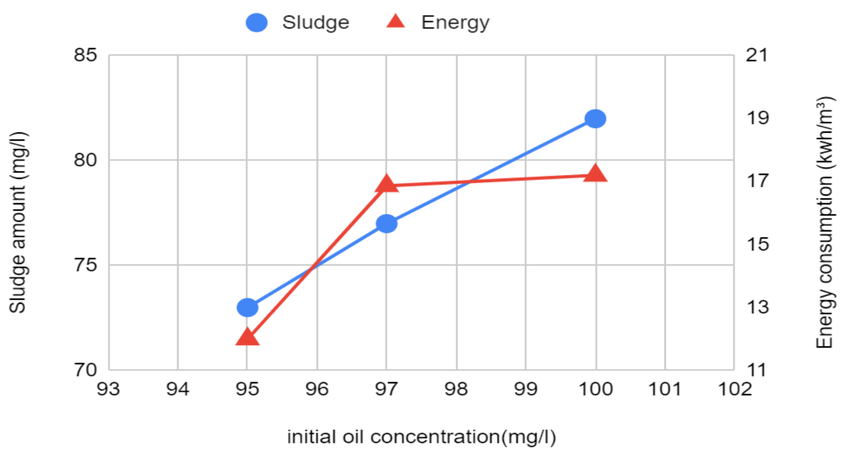

Figure 12: Effect of initial oil concentration on the sludge amount and energy consumption.

By increasing the initial concentration, current is increased during the process. Therefore, the amount of metal hydroxide formation, sludge production and consumption of energy are increased. The results agreed with (Safari et al ., 2016). Summarily, the results have confirmed that sludge production and consumption of energy are proportional to voltage, time, the initial oil concentration, as well as supporting electrolyte concentration furthermore, there was a direct relationship between the amount of sludge formed and removal efficiency of the COD and but further monitoring is required for oil and grease in order to ensure that the plant is successful in implementing Storm water Pollution Prevention Plan (SWPPP) (EPA 2014). Even though the optimized EC process was not able to treat the oil and COD to its treatment limit, it was able to remove almost 99.5 % of influent concentration; therefore, it can be used as a pretreatment for this type of wastewater.

5. Conclusions

In the current study, removal of COD and oil from oily wastewater was investigated using electrochemical (EC) treatment method. The results of this study demonstrated that EC could be successfully used to remove the COD and oil from refinery wastewater. The effect of different parameters such as pH, time, voltage, supporting electrolyte, electrode material and initial concentration are studied. Higher removals of COD and oil were achieved when electrode submerged depth 12 cm, four Aluminum electrodes with four iron electrodes, distance between the electrodes 2 cm, and CO of Oil and COD are 95 mg/l and 710 mg/l, respectively. According to this study, the following points are the main conclusions:

1. The pH has a considerable effect on the COD and oil removal The optimal pH for current experiments was 7. In addition, the pH of wastewater was nearly constant during the process due to the buffering effect of the EC.
2. The electrode arrangement of Al-Al as anode-cathode achieved the highest efficiency in reducing the COD and oil.
3. Increase in voltage, time and NaCl concentration resulted in increased removal efficiency, sludge production and energy consumption. There was almost no removal for COD and oil when no NaCl is added to the solution which is due to low conductivity of oily wastewater.
4. Augmentation in the initial concentrations decreases the COD and oil removal efficiencies but increases the sludge production and consumption of energy.
5. EC is a promising process for pretreatment of oily wastewater. Furthermore, the buffering effect of the EC process allows the wastewater to be discharged without pH adjustments.
6. Highest removal efficiency for pollutants from oily wastewater was observed at pH 7, 50 min, 10.5 V, NaC1 concentration of 0.5 g/L, aluminum as anode and cathode material, and initial oil and COD concentrations are 95 mg/l and 710 mg/l, respectively. Removal effectiveness were 94.2 % for oil and 99.5 % for the COD. Moreover, the amount of sludge produced and energy consumption were 73 mg/L and 12 KWh/m3, respectively.

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