

ELECTROCOAGULATION FOR OIL AND GREASE WASTEWATER TREATMENT USING ALUMINUM ELECTRODES

الترويب الكهربى لمعالجه الزيوت والشحوم فى المخلفات السائلة باستخدام لأقطاب الالومونيوم

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المخلص:

تم إجراء تجارب الترويب الكهربى بنظام الدفعة لتقييم أزاله الأوكسجين الكيمياءى الممتص والزيوت والشحوم من المخلفات السائلة باستخدام اقطاب الالومونيوم . تم دراسة تأثير معايير التشغيل مثل شدة التيار , التركيز الابتدائى للأوكسجين الكيمياءى الممتص, ومدة التفاعل وتركيز كلوريد الصوديوم على كفاءة أزاله الأوكسجين الكيمياءى الممتص والزيوت والشحوم. وقد وجد من البحث أن زيادة شدة التيار زاد من كفاءة أزاله الأوكسجين الكيمياءى الممتص. تركيز الأوكسجين الكيمياءى الممتص الابتدائى كان له تأثير قليل على نسب الإزالة. النتائج أظهرت أن أفضل أداء تم تحقيقه كان باستخدام أقطاب الالومونيوم عند تيار شدته 1.3 أمبير و 11.6 فولت مع مدة تفاعل 15 دقيقة وتركيز أوكسجين كيمياءى ممتص ابتدائى 1500 مج/لتر و تركيز كلوريد الصوديوم 0,50 جم /لتر. تحت هذه الظروف، كفاءة أزاله الأوكسجين الكيمياءى الممتص وصلت إلى 95.07% وتكلفه استهلاك الطاقه والكيمواويات واقطاب الالومونيوم تعادل 0,84 جنيه / متر مكعب لكل جم أوكسجين كيمياءى ممتص يتم أزالته. وبالمثل تكلفه معالجه المتر المكعب من المخلفات السائلة لمحطات غسيل السيارات تعادل 0,32 جنيه لكل جم أوكسجين كيمياءى ممتص يتم أزالته بعد 7,5 دقيقة عند 11.6 فولت و 1.3 أمبير باستخدام اقطاب الالومونيوم بكفاءه ازاله 71.1% .

Abstract

Batch electrocoagulation (EC) experiments were carried out to evaluate the removal of COD and O&G from wastewater using aluminum electrodes. The effects of operating parameters such as current intensity, initial COD concentration, contact time and NaCl concentration on COD and O&G removal efficiency had been investigated. It was found that increasing current intensity increased COD and O&G removal efficiency. Initial COD concentration had a little effect on removal efficiencies. Results showed that the best performance was obtained using aluminum electrode at a current intensity of 1.3 Ampere and 11.6 volt in 15 min contact time for 1500 mg/L initial COD concentration and 0.5 gm/L NaCl concentration. Under these conditions, COD removal efficiency reached 95.07% and the cost of energy, chemicals and material consumption was 0.84 LE/m³/gm COD removal. Similarly, the cost of treatment of cubic meter for carwash station wastewater equals .32 LE/gm COD removal after 7.5 min at 11.6 volt and 1.3 Ampere, using aluminum electrodes with removal efficiency of 71.1% .

Key words:

Electrocoagulation, Electrodes, Oil and grease and COD removal efficiency

1. Introduction

The O&G contained in the wastewater aggregate and foul the sewer

system and produce unpleasant odor (2). Environmentally problematic oily wastewaters are produced in large volumes every day. O&G, are common pollutants

found in wastes from a vast range of industries such as petroleum refineries, petrochemical, metal manufacture, machining and finishing, food processors, textile and carwash stations(3).

Main pollutant in oilfield wastewater is oil which may range between 100 and 1000 mg/l or still higher depending on the efficiency of emulsification and nature of raw oil (4).Oily wastewaters can be classification into three categories: free-floating oil, unstable oil/water emulsions, and highly stable (steady) oil/ water emulsions. Free floating oil can be readily removed by mechanical separation whilst unstable and stable oil-water (O/W) emulsions must be mechanically or chemically broken and detached gravitationally (3).

Carwash wastewaters can be harmful to humans and environment if released without treatment to surface water bodies as they contain a lot of pollutants such as detergents, oil, grease, solvent-based solutions, road grime, heavy metals, etc., that can be toxic to fish, organisms and the accumulation of these pollutant species in the aqueous bodies leads to undesirable effects on human life and on the environment (5).

The environmental requirements in the Egyptian law 93 for year 1962 and the modified law 44 for year 2000 recognize that water should be spent on sewage system does not exceed COD 1100 mg/l, oil-grease 100 mg/l and pH be in the range between 6 to 9.5.

Although there were a lot of techniques available, including a variety of filters, gravity separation, air flotation, biological process, membrane bioreactor, carbon adsorption, chemical coagulation, electrocoagulation, electro flotation etc., for separation of oil-water emulsions, some of the researchers stated that there is still a lack of efficient universal technique in treating oily wastewaters (4, 6).

There are two kinds of coagulation: conventional coagulation and electrocoagulation. Conventional (chemical) coagulation refers to the addition of chemicals such as Alum [$Al_2(SO_4)_3 \cdot 18H_2O$] to an aqueous solution to join small dispersed particles into bigger agglomerates which can be removed by some other method such as sedimentation, air floatation, or filtration (3, 7). Conventional coagulation involves a number of drawbacks such as the high amount of require coagulant, corrosion problems with reducing pH and problems with produced sludge (3).

The rule of electrocoagulation (EC) has been popular knowledge for over 120 years with electrochemical methods first being used for water and wastewater treatment in 1887. In 1906 Dietrich invented the first electric water purifier which used aluminum electrodes whilst J.T. Harries received a patent in 1909 for wastewater treatment by electrolysis with sacrificial aluminum and iron anodes (3). The electrocoagulation of drinking water was first implemented on a large scale in the US in 1946(8).

The EC unit is environmentally friendly so that it does not produce corrosion or any pollutants. This technique has some merits when compared to conventional methods such as simple equipment, easy to apply, less retention time required and less sludge production (5, 9).Furthermore, Electrocoagulation is efficient in removing suspended solids as well as O&G. Many investigators found that it removes metals, colloidal solids, particles containing arsenic, dyes, paper mills, breaking oil emulsions in water, phosphate, boron and bacteria, viruses and cysts (9→11).

The nature of the electrode material is a main issue in electrochemical treatment and the appropriate selection of electrode material is very essential. The most popular electrode materials used for electrocoagulation technique are aluminum

and iron because they are cheap, readily available and proven effective since their dissolution in aerated media produce trivalent species (12).

Rupesh et al. said that oil removal efficiency was 90% at 4.72 pH within 30 min treatment time for 50 mg/l concentration of oil and 94.44% of oil removed in 30 min at 4 mg/l of salinity using Aluminum electrode (4). In the second place, Fouad treated oil-water emulsions. In his work the initial O&G concentrations investigated were 200, 300 and 400 ppm, pH values used were 3, 5, 7, 9 and 11(11). Maha et al., treated oil tanning effluent and proved that iron (Fe) and aluminum (Al) electrodes were considered as sacrificial electrodes in different combinations. They found that under best operating conditions such as 20 mA/cm² current density, 5% effluent concentration, 1.0 g/l NaCl concentration and Fe/Fe electrodes, % COD removal, energy consumption and operating cost were 89.65%, 1.279 kW h/m³ and 6.28 US \$/m³, respectively (15). Bensadok et al., (2011) focused on study the effect of NaCl concentration between 0.5 and 2.0 g/L on the removal efficiency and they proved that the greatest removal efficiency was obtained with the use of both cathode and anode made of aluminum (Al-Al system) (12).

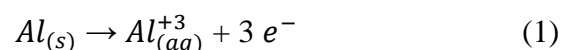
This paper is primarily aimed at development the technology of O&G removal by electrocoagulation. In the present study, the effect of numerous operating conditions such as initial COD concentration, current intensity, NaCl concentration, and contact time on the removal efficiency of COD have explored and discussed to identify the optimum operational conditions and low cost using aluminum electrodes.

2. Theory of Electrocoagulation

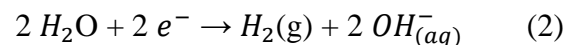
As shown in Fig. 1, aluminum is usually used as electrodes and upon the application of a direct current their cations are produced by dissolution of sacrificial anodes. The metal ions produced are hydrolyzed in the electrochemical cell to create metal hydroxide ions according to anodic and solution reactions and the solubility of the metal hydroxide complexes formed depends on pH and ionic power. Insoluble flocs are produced at pH range between 6.0 and 7.0. Positive metal species react with negatively charged particles in the water to form destabilized colloids and then flocs. The in situ generation of coagulants means that electrocoagulation processes do not require the addition of any chemicals. The gases generated at the cathode during the electrolysis of water and metal dissolution according to cathodic reaction permit the resulting flocs to float and it may be removed by any skimming technique (4, 5, 7, 9, 16, and 17).

The oxidation - reduction reactions involved in the electrochemical cell are as follow:

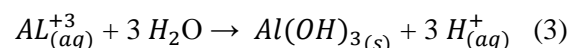
Anodic (oxidation) reactions:



Cathodic (reduction) reactions:



In the solution:



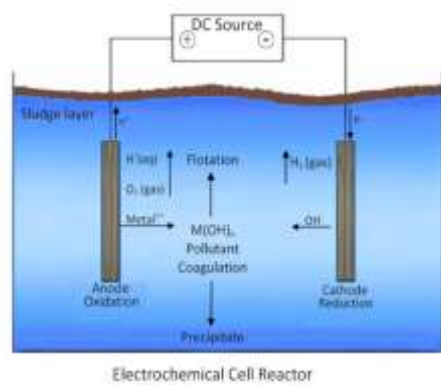
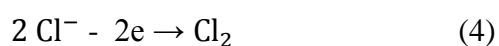


Fig. 1

When the concentration of NaCl salt in solution increased, conductivity of the solution and the current intensity increased. The higher ionic power would generally cause an increase in current intensity at the same cell voltage (8). It was found that the addition of NaCl significantly boosted the oil removal but further increase in NaCl decreased the oil removal (4). The presence of NaCl reduced the size of gas bubbles, especially hydrogen Gas. Since the buoyancy of smaller bubbles was lower than bigger bubbles, they rise slowly to the surface with high opportunities for collision with oil drops. This leads to a progress in the oil removal process (4).

When chlorides were presented in the solutions the products from anodic discharge of chlorides were Cl_2 and OCl^- . The OCl^- itself is a strong oxidant, which capable of oxidizing organic molecules present in wastewater (9). It decreased passivation effect and raised the current efficiency. The following reactions (1, 2, and 3) explain the formation of hypochlorite (15).

When we add NaCl there are three equations:



The quantity of electrode material dissolved or consumed during the electrocoagulation process depended heavily on the current intensity as explained by the Faraday's law as follow (5, 7, 8).

$$m = \frac{M I t}{n F vol} \quad (7)$$

where: m is the mass of dissolved metal (gm/L); M is the molecular weight (g/mol) ($M_{Al}=26.98$ g/mol); I is the current intensity (Ampere); t is the contact time (second); n is the number of electrons involved in the oxidation reduction reaction ($n_{Al}=3$); and F is the Faraday's constant (96,485 C/mol) and vol is the sample volume (liter).

3. Materials and methods

3.1. Experimental set-up

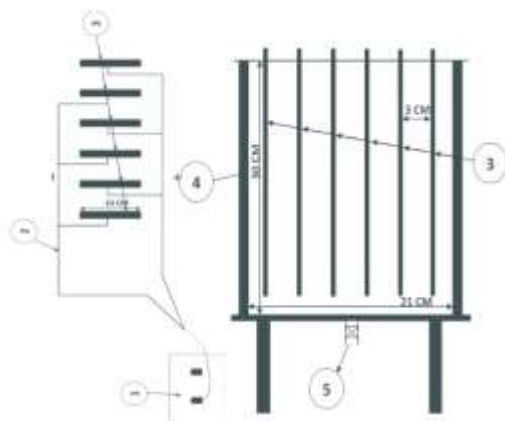
Experiments were implemented in a batch electrochemical rectangular glass cell shown in Fig. 2 that had the following dimensions: 21 cm long, 15 cm wide, and 30 cm height. The total volume of 6 liters of wastewater was treated in the electrochemical cell with 20 cm wetted depth and 10 cm free board. Six parallel aluminum electrodes made of rectangular metal sheets with dimensions of 30 cm height, 10 cm wide, and 1 mm thickness were used. Three electrodes of them were connected parallel as anodes and the other three connected as cathodes. These electrodes connected in a monopolar mode separated by a space of 3 cm and an immersed height of 16 cm (because if the spacing between electrodes <10 mm it impeded movement of liquid and hindering removal efficiency)(18). The electrode gap was kept constant in all experiments. The immersed area of one electrode was $160cm^2$. The electrodes were dropped to the wastewater sample and connected to digital multi meter, KEW SNAP model-2007, for measurement the current and the potential between the electrodes. The D.C. power supply output had three different

current conditions: 1.3 A, 1.6A and 1.9 A with the volts of 11.6 V, 14 V and 16.4V, respectively.

3.2. Synthetic oil–water emulsions

Emulsions were set by adding dosages of dirty vehicles motor oil (Mobil Oil) to 1 liter of tap water and violently mixed for 3 min. The mixture showed a uniform white color. After preparation the emulsion, it was left for 30 min to observe its stability. The emulsion was prepared by initial oil and grease concentrations of 400 mg/l, 550 mg/l and 700 mg/l that gave initial COD 1100 mg/l, 1280 mg/l and 1500 mg/l respectively. Therefore COD value was taken as a measure of O&G.

The initial conductivity ranged from 365 μ s to 381 μ s.



(Plan) (Elevation)

Fig. 2. A schematic diagram:

(1) DC power supply, (2) Anode and cathode connections, (3) six electrodes of aluminum, (4) Electrocoagulation cell, and (5) drainage valve.

In order to study the effect of conductivity, NaCl was added to the Emulsion in the fourth set with initial COD concentration of 1500 mg/l. The NaCl concentrations in that experiments were 0.50, 1.00, 1.50 gm/l. The initial pH ranged from 6.94 to 7.2.

3.3. Carwash wastewater samples

The wastewater was collected from the top of the settling tank in an automatic

carwash station situated in Mansoura city, (EGYPT).

3.4. Experimental method

First EC-cell was filled with the synthetic –water emulsion. Electrodes were submerged and then the current was passed by the regulated DC power supply. The reaction was timed, beginning when the D.C. power supply was switched on. The electrodes polarity was changed at time intervals of 15 min. Samples of 5 ml of oil–water emulsion were withdrawn from the depth of 5cm below the free surface of oil–water emulsion at regular time intervals of 15 min. The effect of the electrochemical treatment was determined by measuring COD at the regular time intervals of 15 min. After each run the electrodes were cleaned and rinsed with HCl(10% concentration) to remove the oxides formed at the anode surface and then dried. Anodic dissolution was determined by measuring weight of sacrificial anode before and after experiments.

3.4. Analytical measurement

The experimental parameters measured were COD, O&G concentration, conductivity, TDS and pH. Analysis was carried out by the standard method for the examination of water and wastewater (22nd edition, 2012) (1). The COD was measured by the closed reflux, colorimetric method and O&G was measured by hexane extractable method according to standard methods (1). The removal efficiency was determined as $(C_0 - C)/C_0$.

In order to accomplish the aims of this study five sets of runs were planned. Each set of first three sets contained three experiments with different calculated O&G concentration (400, 550, 700 mg/lit.). The first three sets were conducted with Al electrodes (Al/Al) under the previously mentioned output current conditions of D.C. power supply. The fourth set of

experiments was conducted with emulsion samples of calculated COD concentration of 1500 mg/lit and NaCl concentration of 0.50, 1.00, 1.50 gm/lit. Using Al electrodes and with current of 1.3 A and 11.6V. The last set of experiments was conducted with carwash wastewater samples.

4. Result and discussion

4.1. Effect of electrode material & current intensity

4.1.1. Aluminum electrodes (Al/Al system)

Results of the first 3 sets of runs with Al electrodes were depicted in figures (3 - 5). From it can be noticed that after 60 min, for initial COD concentration of 1500 mg/l the percentage of COD removal was 52.87%, at 11.6 volt and 1.3 ampere, 84.87%, at 14 volt and 1.6 ampere and was 95.73 % at 16.4 volt and 1.9 ampere. As shown in figures (3 - 5), the COD removal efficiencies after 60 min. were 47.63 - 52.87 % at current of 1.3 A, 82.36- 84.87 % at 1.6 A and 90.73- 95.73 % at 1.9 A.

It is commonly noted that pH increases during an electrochemical process because the process leads to the formation of metal hydroxide according to previous equations (19). pH increasing was in the range of 0.0 to 0.5 using aluminum electrodes.

With the initial COD concentration of 1500 mg/l after 60 min, the COD removal efficiency increased from 52.87 % to 84.87 % by increasing the current voltage from 11.6 volt to 14 volt. When the applied current voltage was increased from 14 volt to 16.4 volt, the COD removal efficiency increased from 84.87 % to 95.73 % as seen in figure (3→5).

Based on Faraday's law, increasing applied voltage (or current intensity) resulted in an increasing amount of metal hydroxide flocs for the removal of colloidal particles (17).

It was also recognized that the rate of bubble-generation increased and the bubble size decreased with increasing

current intensity; both of these facts were beneficial in terms of high pollutant removal efficiency by H_2 flotation as mentioned in eq.2 (14, 16).

Initial COD concentration had a low effect on COD removal efficiency. The efficiency of COD removal for COD of 1500 mg/l is slightly better compared to COD of 1280 mg/l and 1100 mg/l.

4.2. Effect of contact time

The time range from 0 to 60 min was studied to show how time affected the removal efficiencies. As the time of electrolysis increased comparable changes in the removal efficiency of COD was observed.

COD removal efficiency increased with increasing contact time. According to the Faraday's law, the amount of aluminum released to the EC system using Al electrodes was affected by the residence time which leads to an increase in Al ions freed to the system.

COD removal efficiency increased fast at first 45 min especially at 14 and 16.4 volt and increased slowly at last 15 min in all applied volts as shown in figures (3→5).

4.3 Sludge production

The effluent with aluminum electrodes was found very clear and stable. About two thirds of the sludge floated on the top and came out from a sludge outlet, while the other third was generated after sedimentation (2).

Volume of sludge from each batch increased with increasing initial concentration and current intensity. Minimum and maximum sludge volumes per batch were 63cm^3 and 567cm^3 .

4.4. Effect of salinity (NaCl)

At using emulsion of initial COD concentration of 1500 mg/l and NaCl of 0.50 gm/l in (Al/Al) system, COD removal efficiency reached to 95.07% and became

constant after 15 min, at 11.6 volt and 1.3 Ampere, as shown in figure (6).

Increasing NaCl dosage to 1 gm/l and 1.5 gm/l did not effect on COD removal efficiency at 15 min, but the COD removal efficiency slowly increased up to 99.1% after 15 min. Initial conductivity was 1446 μ s, 2550 μ s and 3630 μ s in case of 0.5gm/l, 1.0gm/l and 1.5 gm/l.

4.5 carwash station samples

COD removal efficiency reaches to 78.12% after 15 min and became constant for carwash station samples, at 11.6 volt and 1.3 Ampere, using Aluminum electrodes as shown in Fig. 7. Under previous condition, COD decreased from 3200 mg/l to 925 mg/l with removal efficiency of 71.10% after 7.5 min and outlet pH is 9.34 for carwash station samples and become in the limits of the environmental requirements in the Egyptian laws. Conductivity decreases from 3980 μ s to 3770 μ s and volume of sludge reaches to 472.5cm³.

It is shown in Fig. 8 that COD decrease from 3300 to 625 mg/l with 81.1 % COD removal at 11.6 volt and 1.3 Ampere after 15 min. By increasing current intensity to 14 volt and 1.6 Ampere the COD decreased to 375 mg/l with 88.6 % COD removal, but increasing current intensity to 16.4 volt and 1.9 ampere the COD decreased to 350 mg/l with 89.4 % COD removal

5. Total cost

One of the most important parameters that greatly affect the application of any technique of wastewater treatment is the cost. The cost include only energy, chemicals and material consumed during the operation of EC cell.

The electrical energy consumption increased with increasing current. Since the current is a main variable in controlling the performance of the electrocoagulation, it is preferable to decrease cell voltage rather than decrease current to minimize the energy consumption (4). Operating time is the significant performance parameter in the electrocoagulation process as higher operating time results in higher energy requirement (15). Electrical energy

consumption was calculated using the following equation:

$$E = \frac{VIt}{(COD_i - COD_f)vol}$$

Where E is the energy consumption (kwh/g COD); V is the current voltage (volt); I is the current intensity (ampere); t is the contact time (hour); COD_i, COD_f is initial and final chemical oxygen demand (mg/l) respectively; and vol is the sample volume (liter).

$$\begin{aligned} \text{Total Cost (LE/g COD removal/m}^3\text{)} \\ = E \text{ (kwh/g COD)} \times \text{Price (LE/kwh)} + m \\ \text{(kg/m}^3\text{)} \times \text{metal Price (LE/kg)} + \text{salt price} \\ \text{(LE/m}^3\text{)} \end{aligned}$$

Where the price of kwh of electricity equals 0.25 LE; price of kg aluminum equals 31.75 LE; and price of NaCl; for 0.5gm/l equals 0.16 LE ; for 1.0gm/l equals 0.32 LE ; for 1.5 gm/l equals 0.48 LE.

Experimental mass of aluminum is 0.8 of theoretical mass as shown in table (1).

Total costs of cubic meter for each gm COD removal after different interval times were calculated, total costs decrease with decreasing current intensity and with increasing for initial COD concentration. Minimum and maximum total cost after 1 hr are 2.6 and 4 LE/g COD removal/m³ as shown in table (2).

The low cost results were at 11.6 volt and 1.3 Ampere, for initial COD concentration of 1500 mg/l, adding 0.5 gm/l NaCl using aluminum electrodes. Total cost was 0.84 LE/g COD removal/m³, after 15 min with removal efficiency of 95.07%. As shown in table (3). Under previous condition, total cost of treatment of cubic meter for a carwash station equals 0.32 LE/gm COD removal after 7.5 min with removal efficiency of 71.1% (final COD 925) and outlet pH is 9.34 and subject to the environmental requirements in the Egyptian law.

Fig. 3 COD removal with Al electrodes at 11.6 volt and 1.3 A.

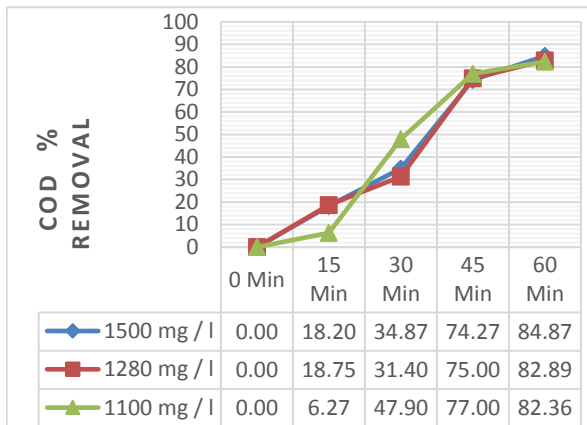


Fig. 4 COD removal with Al electrodes at 14 volt and 1.6 A

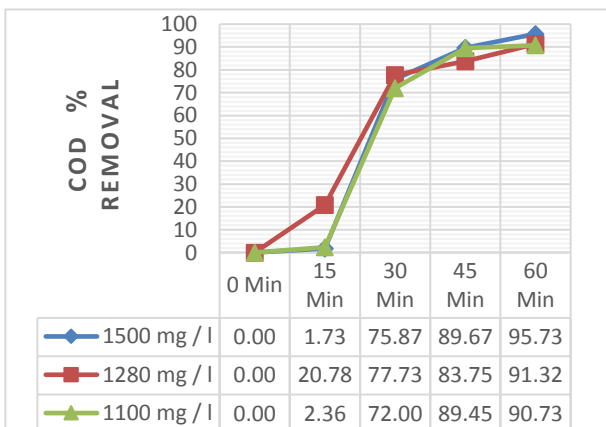


Fig. 5 COD removal with Al electrodes at 16.4 volt and 1.9 A.

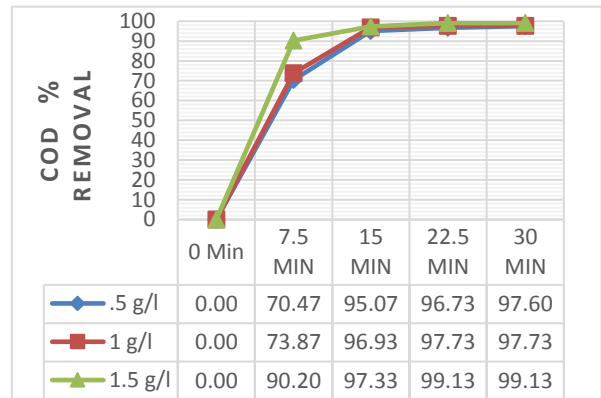


Fig. 6 Effect of NaCl dosage on COD removal at 11.6 volt and 1.3 A for initial COD concentration of 1500 mg/l.

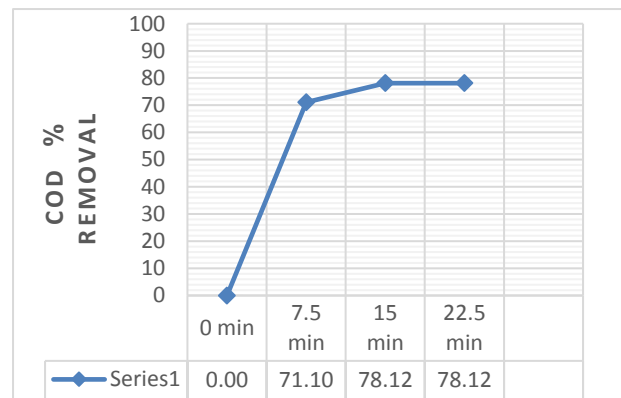


Fig. 7 COD removal at 11.6 volt for carwash station sample (Initial COD =3200 mg/l).

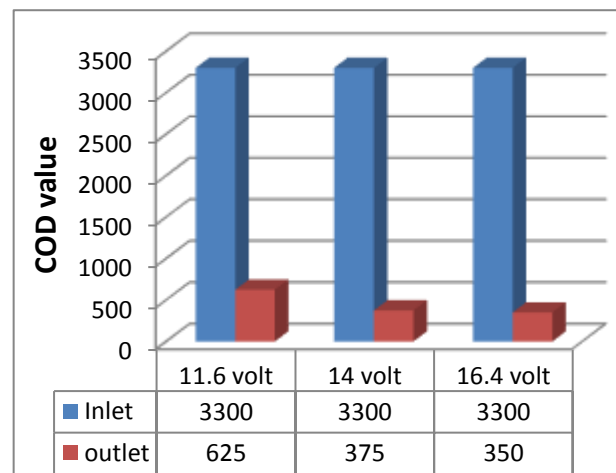


Fig. 8 Effect of current voltage on COD removal for carwash station samples at reaction time 15 min.

Table (1) Estimation of theoretical and experimental mass dissolved of AL electrodes at different volts and contact times

Current voltage and Intensity	Mass dissolved (gm/l) after (1 hr)		Mass dissolved (gm/l) after (45 min)		Mass dissolved (gm/l) after (30 min)	
	Theo.*	Experi.**	Theo.*	Experi.**	Theo.*	Experi.**
11.6 volt 1.3 Ampere	0.0727	0.058	0.0545	0.0435	0.036	0.029
14 volt 1.6 Ampere	0.0895	0.0713	0.0671	0.05347	0.0447	0.03565
16.4 volt 1.9 Ampere	0.1062	0.0847	0.0797	0.0635	0.053	0.04235

* Theoretical value using Faraday law in eq(7)

**→Experimental value by measuring the loss of metal during the present study

Table (2) Total Cost of treatment of cubic meter for each gm COD Removal using Aluminum Electr

Volt and Ampere	Initial COD concentration (mg/l)	COD removal % after(1 hr)	Total cost(LE) after(1 hr)
11.6 volt 1.3 Ampere	1500	52.87	2.6
	1280	48.05	2.8
	1100	47.63	3
14 volt 1.6 Ampere	1500	84.87	3
	1280	82.89	3.14
	1100	82.36	3.3
16.4 volt 1.9 Ampere	1500	95.73	3.6
	1280	91.32	3.8
	1100	90.73	4

Table (3) Total Cost of treatment of cubic meter for each gm COD Removal using NACL dosages for initial COD concentration of 1500 mg/l

NACL dosage (gm/l)	COD removal % (after 15 min)	Total cost (LE) (after 15min)	COD removal % (after 30 min)	Total cost (LE) (after 30min)
.5	95.07	0.84	97.6	1.51
1	96.93	1	97.73	1.68

1.5	97.33	1.16	99.13	1.83
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6. Conclusion

1. The results of this study showed that electrocoagulation could be applied in the treatment of carwash wastewater.
2. At using (Al / Al) system the maximum COD removal efficiency after 60 min. was 90.73 – 95.73 % at current intensity of 1.9 A and 16.4 V.
3. The current intensity is an important operating factor influencing the performance of electrocoagulation process and initial COD concentration has a little effect.
4. Results showed that the economic performance was obtained using aluminum electrode at a current intensity of 1.3 A and 11.6 volt in 15 min. contact time for 1500 mg/L initial COD concentration and 0.5gm/L NaCL concentration. Under these conditions, COD removal efficiency reached 95.07% and total cost was 0.84 LE/m³/gm COD removal.
5. Total cost of treatment of cubic meter for carwash station sample equals 0.32 LE/gm COD removal after 7.5 min at 11.6 volt and 1.3 Ampere, using aluminum electrodes with removal efficiency of 71.1% (final COD 925) and outlet pH is 9.34 .

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