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Electrocoagulation Treatment of Oily Wastewater in the Oil Industry

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Abstract

Electrocoagulation has turned out to be a rapidly growing area of wastewater treatment due to its ability to remove contaminants that are generally more difficult to remove by filtration or chemical treatment systems. These pollutants may include emulsified oil, petroleum hydrocarbons, suspended solids, and heavy metals. In this novel technology, metal cations are released into the wastewater through dissolving metal electrodes. Simultaneously, beneficial side reactions can help in removing flocculated material from the water.

Oily wastewater represents a dangerous threat when discharged to the environment; therefore treating it becomes vital in oil industry. This research has investigated electrocoagulation as a simple, effective and economic technique for treatment of such wastewater. Bench scale reactor was used to evaluate the factors that may affect the treatment of oily wastewater. Aluminum was used as a sacrificial anode. Electrodes were arranged at different configurations to select the optimal one. Other tested operation parameters include time of treatment, current density, distance between the electrodes, and the electrolyte concentration of the emulsion. The experimental results indicated that electrocoagulation was very efficient and able to achieve 94% turbidity removal in less 60 min, pH:7, current density of 45 (A/m^2).

Also the effect of initial concentration of oil (100-1000 ppm) in wastewater, applied voltage (10-20 v), the dose of NaCl electrolyte (0-1 g) and temperature (35-60 ^oC) were optimized. What distinguishes this work is the the type of the electrodes configuration. Circular horizontal Al/Al electrodes (Ø12 cm) with 17 holes (Ø7 mm) have been found to give the best electrocoagulation environment and thus optimum results. The associated electrical energy consumption is 1.6 kW h/m^3 for flotation times of 60 min.

Keywords Electrochemistry; Electrocoagulation; emulsion; Oily water; Wastewater Treatment

معالجة المياه الملوثة في الصناعة النفطية بتقنية التخثر الكهربائي

الخلاصة:

لقد تزايد بسرعة استخدام تقنية التخثر الكهربائي في مجال معالجة المياه الملوثة نظرا لقدرتها على إزالة الملوثات التي يصعب عادة إزالتها بواسطة الانظمة التقليدية مثل انظمة الترشيح أو المعالجة الكيميائية للنفط المستحلب ومجموع الهيدروكربونات البترولية والمواد العضوية المقاومة للحرارة والمواد الصلبة العالقة والمعادن الثقيلة. يتم في هذه النوع من التكنولوجيا، توليد كاتيونات معدنية في الماء المعالج من خلال التحلل الكهربائي لاقطاب معدنية. وفي الوقت نفسه، يمكن أن تساعد التفاعلات الجانبية المفيدة في ازالة المواد المخثرة من الماء. وتشكل المياه العادمة النفطية تهديدا خطيرا عند تصريفها للبيئة وبالتالي فان معالجتها يصبح حيويا في الصناعة النفط. تم في هذا البحث دراسة عملية التخثر الكهربائي باعتباره أسلوب بسيط وفعال واقتصادي لمعالجة اغلب انواع المياه العادمة المعروفة. تم استخدام مفاعل مختبري لتقييم العوامل التي قد تؤثر على معالجة المياه الملوثة باستخدام اقطاب معدنية من الالمنيوم / او الحديد كقطب انود متحلل. تم ترتيب اقطاب الخلية الكهروكيمياوية في تكوينات مختلفة لتحديد الأمثل منها في المعالجة. كما شمل البحث دراسة عدد من المعلمات العملية مثل اختبار تاثير وقت المعالجة، كثافة التيار، المسافة بين الاقطاب، تاثير درجة الحرارة وتركيز المستحلب النفطي المستخدم. لقد بينت النتائج التجريبية أن التخثر الكهربائي كان فعالا جدا وقادر على تحقيق إزالة عكورة بنسبة 94٪ في أقل من 60 دقيقة، عند درجة حموضة: 7، وكثافة تيار : 45 امبير/م^2. كما تم دراسة الاختيار الامثل لكل من التركيز الأولي للنفط المستخدم (100-1000) ملغم / لتر في المياه العادمة، الفولتية المطبقة (10-20) فولت وجرعة الملح المضاف (0-1) غرام ودرجة الحرارة المستخدمة (35-60) درجة مئوية. ان مايميز هذا العمل هو نوع الترتيب الخاص للالكترودات. اذ تم استخدام الكترودات من صفائح الالمنيوم الدائرية (بقطر 12 سم) بترتيب افقي ومثقبة بسبعة عشر ثقبا (قطر كل ثقب 7 ملم) والتي اعطت افضل نتائج. كان استهلاك الطاقة المصاحب للعملية حولي 1.6 كيلوواط ساعة لكل متر مكعب من السائل المعالج لزمن تطويف مقداره 60 دقيقة.

الكلمات الدالة: الكيمياء الكهربائية، التخثر الكهربائي، المياه الملوثة، الدهون، معالجة المياه، النفط المستحلب.

1. Introduction

The recent advance in industry has caused serious deprivation of water quality released to the environment. This is appeared both through negligence in treatment of wastewater before releasing and accidental contamination through spills in marine environments. This originated in the formation of enormous amount of wastewater encumbered with abundant unacceptable toxic compounds for both the environment and human health as well. There are varieties of technologies available for reduction of toxicity

which includes many processes for treating wastewater in order to meet international treatment standards [1-7].

Oil production is one of the most key industrial activities of modern society and its derivatives have many industrial applications. Because of the need to meet the rising demand for oil production, the mining of oil has increased significantly in recent decades. However, this causes damage to the environment, with the main perpetrator being produced water. Oil and its byproducts can be considered as the most important pollutants, as a result of the increasing amounts that have been extracted and processed [5]. Oil in wastewater may be existed in many forms including floating, dispersed and emulsified with the latter being the most difficult to treat. Remediation of produced water is a critical matter in view of the high volume of daily effluent water [6].

Several traditional processes can be applied to treat oily wastewater which may include employing physical, chemical and biochemical techniques. Examples of these processes are membrane, heating, centrifugation, adsorption, ozonation, filtration and coagulation [7]. However, conventional techniques remain inadequate and have many disadvantages including high amount of required coagulant, long running time, high cost, corrosion problems and excessive produced sludge. On the other hand, biological processes are generally less expensive than the chemical counterparts and often have been engaged to treat the organic elements in industrial wastes. Their efficiency decreased as the concentration of pollutant increased.

More recently, several electrochemical methods have been examined for the treatment of wastewater containing oils. These include electro-flotation, electro-deposition and electro-coagulation. However, there has been an increasing awareness in the application of electro-coagulation in the treatment and purification of industrial wastewater [8-13].

Oily Water

Oily water is a broad term and it is used to label all water which contains varying amounts of oil and grease in addition to a variety of other suspended materials. These suspended materials may include sand, clay, besides a range of dissolved colloidal substances, such as detergents, salts, and metal ions. The treatment of oily water can be considered as an

intricate process depending highly on the efficiency to meet the environmental standards for disposal and the characteristics necessary for reuse [1].

In the oil industry, oily water occurs in many phases of production, transportation and refining, as well as during the usage of oil derivatives. However, the production phase is the largest source of this contamination. During the production process, oil is commonly mined along with water and gas. The accompanying water may grasp half of the volume produced, or even higher, approaching 100% at the end of the wells productive life. The release or reinjection of this co-produced water is only allowed after the removal of most of the oil and suspended solids. The composition of this produced water is very complex. Depending on its origin it may comprise a variety of substances such as salts, hydrocarbons, oils, and metals [2].

This extreme production of water has become a foremost apprehension in the oil industry. Before disposal into the environment or use, it is necessary to treat this water because the large amounts of pollutants cannot be discharged into the receiving environment.

Electrocoagulation

Electrolysis is a chemical decomposition in which oxidation and reduction reactions take place when electric current is applied to an electrolytic solution [14] . The electrocoagulation (EC) is a novel technology that has been successfully used to treat industrial wastewater containing different contaminants such as arsenic, phosphate, boron, dyes and viruses [14-19]. Furthermore, it has been also effective in breaking oil emulsions in water and treating paper mill effluent, olive mill effluent, landfill leachate and dairy effluent. However, few studies have investigated the removal of hydrocarbons using EC process. EC is based on dissolution of the electrode material used as an anode. The so-called “sacrificial anode” yields metal ions which preform as coagulant agents in the aqueous solution. For many reasons, EC is an alternative method to the conventional chemical coagulation. EC is capable of reducing the need for chemicals due to the fact that the electrodes facilitate the coagulant. However, many individuals still use chemical coagulants to attempt to enhance treatment. Conventionally, chemical coagulation involves the use of alum, ferric chloride, or ferrous sulfate. These chemicals can be very expensive depending on the volume of water treated. When applying the coagulant, it

performs a similar function as the electrodes, neutralizing the charge of the particulates, thereby allowing them to agglomerate and settle down at the bottom of the tank. In addition, EC is capable of reducing waste production from wastewater treatment and also reduces the time necessary for treatment.

In an EC process, the coagulating ions are generated involving three stages:

(a) Electrolytic reactions at electrode surfaces.

(b) Formation of coagulants in aqueous phase.

(c) Adsorption of soluble or colloidal pollutants on coagulants, which are removed by sedimentation or flotation.

"Sacrificial electrode" dissolved from the anode creating corresponding metal ions. These ions hydrolyze to polymeric iron or aluminum oxyhydroxides, as the coagulating agents. The aluminum and iron hydrolysis products destabilize the pollutants present in the solution, and allowing agglomeration and further separation from the solution by settling or flotation. Destabilization is mainly achieved by means of two different mechanisms;

a) "Charge neutralization" of negatively charged colloids by cationic hydrolysis products.

b) "Sweep flocculation", where impurities are trapped and detached in the amorphous hydroxide precipitate produced.

In view of the above, several parameters such as the coagulant dosage and pH have an influence on the relative importance of charge neutralization and "sweep flocculation". Micro-bubbles (H_2 and O_2) released at the electrodes surfaces bring about electroflotation by adhering to agglomerates and carrying them to the surface of the water. The most important factors influencing the efficiency of the EC process are the electrode materials used, applied voltage, operating time, and solution chemistry, including initial pH and the chemical composition of the aqueous solution being treated. The solution temperature, type of salt used to raise conductivity, electrode gap, and passivation of the anode, also have an impact on the removal efficiency and economic durability of a given EC application. Finally, other un-tried parameters such as the effect of using ultrasonic source are also important to be studied

2. Experimental

Schematic diagram for the experimental set up is given in Figure (1) [19]. The EC tests were done in a batch system using a 2000 ml Pyrex beaker by horizontal circular Al/Al electrodes. The electrodes were connected to a digital DC power supply having an input of 220 V and variable output of 0–20 V, with variable current 0–2 A. Effective area of electrode used was 220 cm^2. The optimum gap between the electrodes was 6 mm. Before each experiment, electrodes were washed with acetone to remove surface grease-oily and pollutants were washed, then matters on electrode surfaces were cleaned by dipping for 1 min into a diluted HCl solution then washed with pure water for the removal of the residuals on their surfaces and dried by oven. Oil for this study was obtained using common lubricant oil from a local market. A synthetic emulsion mixture of oil/water was prepared by mixing an appropriate amount of oil (with density 0.89 g/L and 28 ^{o}API) in 10 liter of water to obtain 100 NTU turbidity. This mixture was then subjected to vigorous mechanical stirring for 15 minutes to form a stable oil/water emulsion. The desired amount of oil mixed with fresh tap water was used under ultrasound agitation for 30 min, for obtaining very stable or soluble synthetic oil wastewater. This quantity will be stored to carry out a number of experiments. All tests were performed at 35 ^{o}C and stirring speed was 250 rpm. Later on, oily solution conductivity was increased by adding proper amounts of electrolytes: NaCl. Methods of analysis samples were periodically taken out from the reactor and then turbidity measurements of the reaction solutions were immediately performed. Residual oil measurement was performed by using a Turbidity meter (HANNA instruments Hi93703). Each experiment lasted 40 min. To achieve mixing of solution during the treatment a magnetic stirrer (BOECO MSH-300N) was used. Temperature and pH measurements were obtained using multi-meter (HANNA Hi9828). Samples were taken from the cell using a pipette tube. Turbidity removal efficiency was calculated from the ratio of the concentration change to the initial concentration expressed in percentage as given in Equation 1. Also Equation energy consumption was estimated from electrical power definition (see Equation 2).

$$T(\%) = \frac{C_o - C_t}{C_o} \quad \ldots\ldots\ldots\ldots(1)$$

$$Energy = Current \times Voltage \times time \quad \ldots\ldots\ldots\ldots(2)$$

Where, T, C_o and C_t are turbidity removal efficiency, initial turbidity concentration and the concentration at a specific time, t respectively.

Fig. (1) The apparatus set up.

3. Results and Discussion

In this study, electrocoagulation using aluminum sacrificial anode was used for the treatment of oily emulsion. In order to evaluate the decreasing of turbidity and increasing oil removal from the emulsion, various important electrochemical factors were investigated: applied voltage, the behavior of the current during the operation, the initial concentration of oil in the wastewater, initial pH, gaps between the electrodes, the added NaCl electrolyte, volume of wastewater and temperature.

3.1. Effect of time on oil separation efficiency:

In this research, turbidity was used as a measure of the oil concentration in the wastewater. The change in turbidity with time during the electrocoagulation process is depicted by Figure 2. As seen in the figure the oil separation efficiency has a direct relationship with the time. Increasing the treating time caused considerable decrease in water turbidity. During this period of time of 60 min, the turbidity has been reduced from 100 NTU to 6 NTU with an efficiency of 94%.

Figure (2) indicates that the electrocoagulation process comprises of two stages which are destabilization and aggregation [23]. The first stage is fast, whereas the second stage is

relatively slow. The turbidity decreased from an initial value of 100 NTU corresponding to separation efficiency of 94%. This value was obtained after 60 min of operation. However, 80% separation efficiency was achieved after 30 min. According to Faraday's law the time of current passage increases the quantity of electricity applied to the cell which leads to more aluminum ions to liberate at the anode. These ions with their positive charges will neutralize the oil droplets that are negatively charged. This effect enhances the separation of the neutralized oil droplets and makes them float on the surface. On the other hand, more hydrogen gas bubbles will be created at the cathode by the increase of the treatment time. These hydrogen bubbles will push the oil drops upwards by bouncy force thus increasing the efficiency of oil separation and decreasing turbidity of the emulsion.

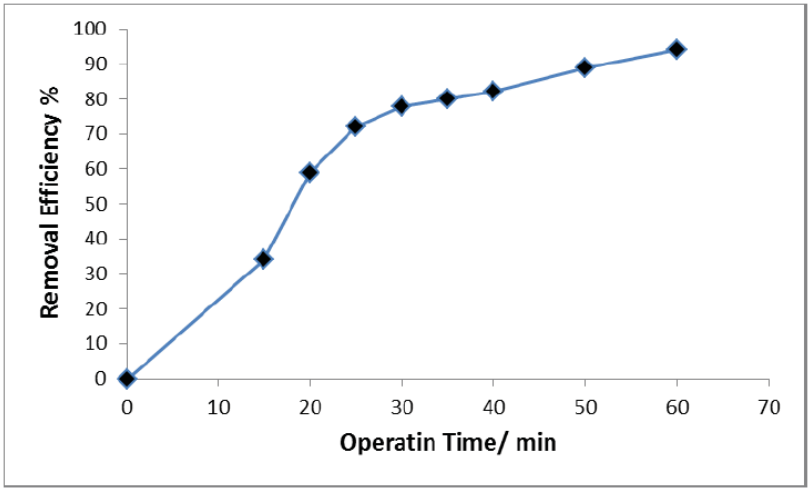

Fig. (2) The relation between time and removal efficiency.

3.2. Effect of distance between electrodes on oil separation efficiency:

The relation between oil separation efficiency and the distance between the electrodes is shown in Figure 3 carried out at 35 ^{o}C and initial oil concentration of 100 NTU and applied voltage of 15v. The inter-electrode distance is an important variable in order to optimize the operating costs of electrolysis systems. Researchers report that when the conductivity of the effluent is high, a larger spacing between the electrodes is possible. On the other hand, when conductivity is low as the case in this work, the spacing should be smaller [20].

In this study, the inter-electrode distance was varied (4 mm, 6 mm and 8 mm) while the other factors remained unmodified. Our results indicate that the inter-electrode distance did affect the performance. Increasing the distance between electrodes has led to reduce removal efficiency, in this case, the ohmic resistance has decreased because of the shortening of the path of the electric current between the anode and the cathode [22]. In addition, the energy consumption also increases due to the higher resistivity of the solution [21].

The results in Figure (3) show an improvement in the efficiency of removal for lower inter-electrode distance between 4 and 8 mm. A distance of 6 mm has been chosen as an optimum distance since the use of larger distances would involve greater energy consumption. Previous results have shown that increasing the inter-electrode distance to about 15 mm has reduced considerably the removal efficiency [18]. Similar results have been reported from other researchers [22].

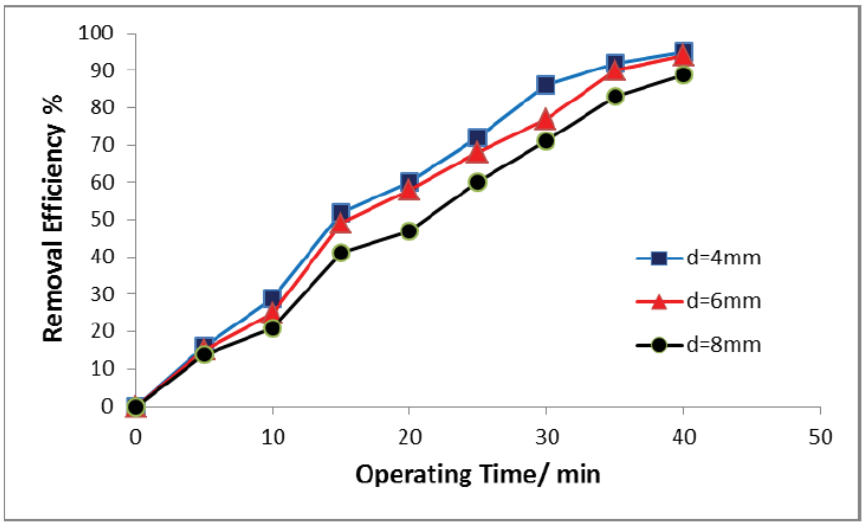

Fig. (3) The effect of changing electrodes distances (with V= 15 v,T=35 ^{o}C).

3.3. Effect of applied Voltage on oil separation efficiency:

The using of different applied voltages (10, 15, 20 v) with fixed electrodes distance of (6 mm) has been investigated here. Figure (4) reveals that the removal efficiency was direct correlation with the applied voltage. The applied voltage is a vital factor in the EC process as it regulates the coagulant dose rate, bubble production rate, size and growth of the produced

flocs. Thus increasing the applied voltage would obviously increase the anode dissolution rate, leading to an increase in the number of metal hydroxide flocs. The applied voltage in the electrocoagulation reactor controls the amount of coagulant ions released from the anode. So, increasing the applied voltage will increase the number of coagulant ions and consequently increase the oil separation efficiency. Another effect of the high applied voltage is the increase of rate of generation of the hydrogen bubbles and the decrease of their size. However, exceeding the optimum voltage would lead to wasting energy due to heating up of the water and decrease in current efficiency. The same results of current density effects were reported by [23].

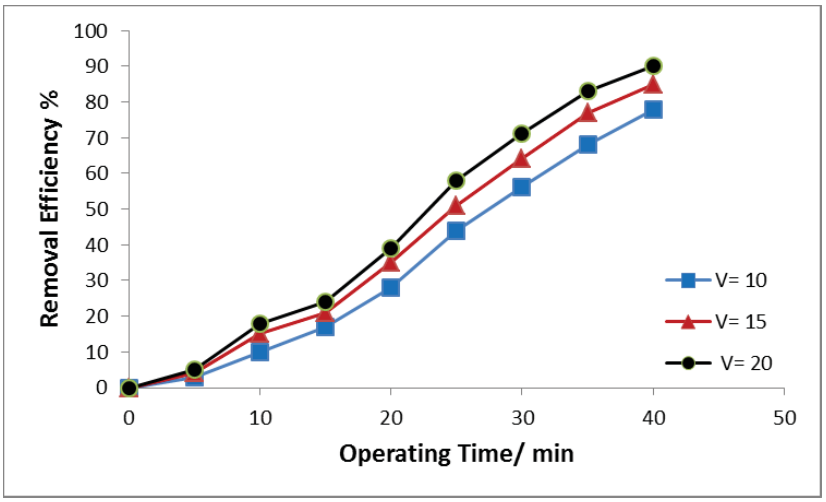

Fig. (4) The effect of changing the applied voltage (with d-6 mm,T=35 ^{o}C).

3.4. The current variation of EC process with time:

Figure (5) shows the behavior of the current during the EC process for three different applied voltages (10, 15, 20 v) at 35 ^{o}C and inter-electrode distance of 6 mm and initial concentration of 100 NTU. It is clear that there has been a continuous low decrease in the current density over the 40 min period of time. A 10% decreased in the current value in this case is not very serious compared with our previous results with zinc contaminated wastewater where over 50% reduction in the current was observed [19].

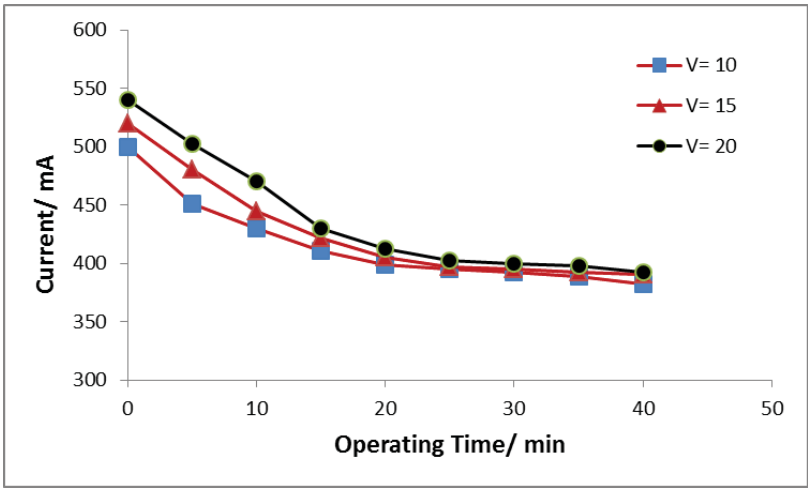

Fig. (5) the variation of the EC current with time (with d=6 mm,T=35 ^{o}C).

3.5. Effect of NaCl concentration on oil separation efficiency:

The effect of adding NaCl concentration on the separation of oil from oily wastewater was studied as shown in Figure (6). From this figure it can be seen that increasing NaCl concentration leads to an increase in the removal efficiency with decreasing in treatment time required to achieve the highest removal percent. The presence of NaCl causes the formation of positive sodium ions and negative chloride ions. Thus, introducing NaCl to the water increases the conductivity of the emulsion solution and decreases the fraction of the applied potential that is consumed to overcome the resistance of the emulsion and accordingly increase the fraction of the applied potential directed to the electrochemical coagulation. However, increasing the electrolyte concentration involves high Cl^- concentration in the emulsion. These negatively charged will compete with the oil droplets in the emulsion towards migration to the anode which decreases the separation efficiency of oil [24]. The same results of sodium chloride effect were reported by [25] and [26].

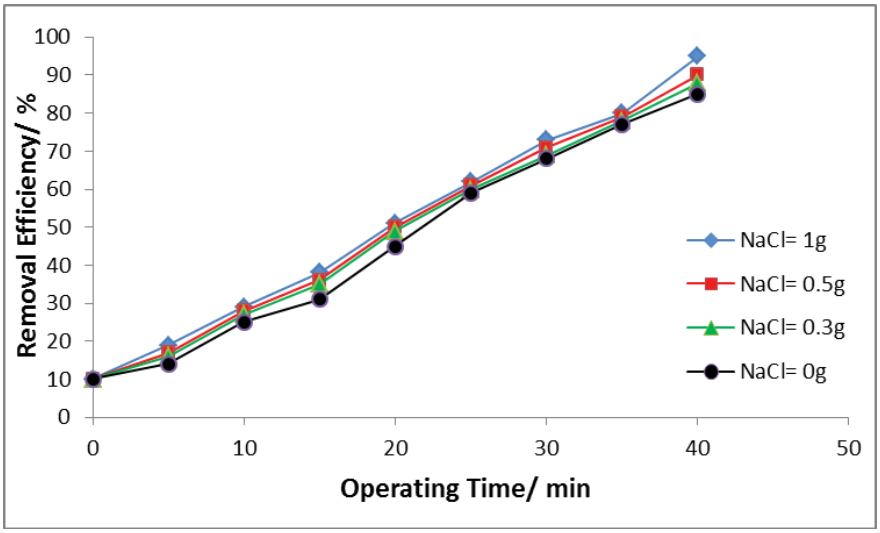

Fig. (6) The effect of adding NaCl (with d=6 mm, V=15 v, T=35 ^{o}C).

3.6. Effect of temperature on oil separation efficiency:

This important operating parameter has received little consideration from researchers [27, 29]. Temperature is a complex factor and its effect has touched by many topics such as the rate of the EC reactions, solubility of metal hydroxides, liquid conductivity, kinetics of the bubbles formation, and the movement of the ions produced. Figure 7 shows the effect of changing temperature on the variation of oil efficiency separation with time. The data specify that oil separation increased slightly with increasing temperature. This result is in agreement with that reported by [28, 29]. They stated that the efficiency achieved with aluminum electrodes increased with temperature up to the 60 °C, above which the efficiency has decreased. In addition, Becher [30] showed that an increase in the temperature resulted in a decrease in the oil emulsion stability due to an increase in the adsorption rate, because the Van der Waals forces and Brownian motion increase and decrease the viscosity of the continuous phase film. This would contribute to increase coalescence rate, through an increased probability of film rupture.

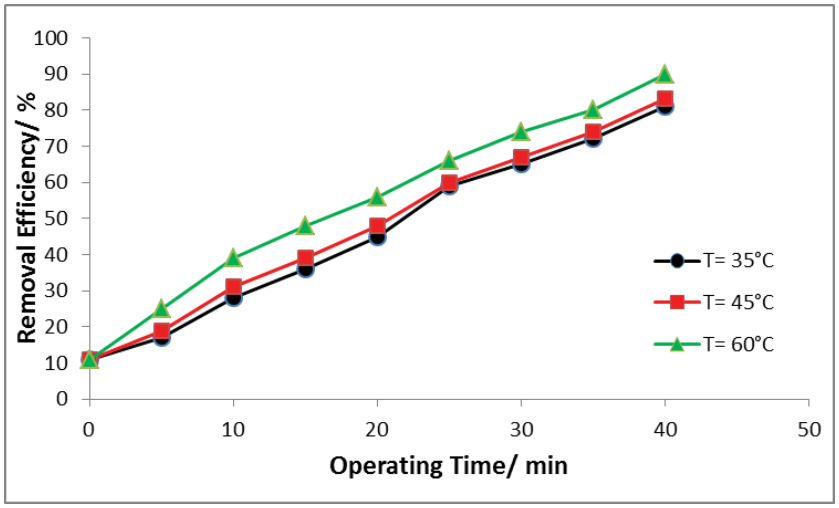

Fig. (7) The effect of changing the temperature (with d= 6mm, V=15 v).

4. Conclusion

In this study, the efficiency of the electrocoagulation process applied to the treatment of oily wastewater emulsion was investigated. It was observed that the electrocoagulation treatment achieves a fast and effective removal of oil turbidity. The treatment efficiency was found to be a function of the initial pH, inter-electrode distance, applied voltage, NaCl electrolyte dose, temperature and operating time under the optimal values of the process parameters.

The results showed removal efficiencies of 94% turbidity with an energy consumption of 1.6 kWh/m^3 with electrode consumption of 0.05 g could be achieved at a current density of 45 A/m^2 with an operation time of 60 min and initial pH of 7.0.

This technique thus seems to be a promising alternative for the treatment of oil-in-water emulsions of the petroleum industry.

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