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Comparative Electrocoagulation of Total Petroleum Hydrocarbons from Oil-Contaminated Water Using Aluminum and Iron Electrodes

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Abstract

Oily wastewater is the main source of pollution in maritime ecosystems, electrocoagulation technology was used to treat water contaminated with total petroleum hydrocarbon. An design of experiment program was used to obtain the best experiments with pre-dtermined factors (pH, voltage, and time) . Use iron plates and aluminum plates to find out which one is more efficient in treating plluted water, the highest percentages of removal of TPH using iron plates (95) under the conditions pH(5), Voltage (25v) , time (60min) , while when using aluminum plates TPH removal was 92 at pH(7) Voltages (15v) , and time (45min) .

Keywords: Electrocoagulation; Iron plates ; Aluminum plates ;TPH removing

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Introduction

The creation of society and the growth and prosperity of cities are fundamentally dependent on water. As a result, one of the most important procedures is treating water to make it safe for human consumption, lessen the harm caused by liquid waste from industrial and human activity, and allow for the potential for reuse (Almayahi et al ., 2022) .

The operations of extracting and refining crude oil require a lot of water. Significant The wastewater that is released has a lot of various toxins , such as heavy metals , dyes, solid particles , bacteria , nutrients, (Zaboon et al., 2022) , and petroleum hydrocarbon , This has caused alteration to the physical, chemical, and biological properties of water (AlJaberi et al ., 2020) . The environment is negatively impacted by the intense concentration of aromatic and aliphatic compounds in refinery process effluents (El-volumes of generated water are released from upstream during the extraction of crude oil from subterranean reservoirs. More oily effluent is discharged downstream when crude oil is refined . Aquatic biota is suffered when industrial discharges and oil spills end up in bodies of water. Water pollution still poses a serious threat to all living things, as it degrades the ecosystem, the economy, and the health of the inhabitants (Tjale et al., 2022) .

Naas et al ., 2009) . In refinery effluent treatment, oil-water separation and coagulation are commonly employed initially, with biological treatment coming later (Naser et al., 2021) . However, these methods produced a great deal of sludge and had a number of practical and technical shortcomings (Oladoye et al., 2021).

The EC generation of metal ions (such Al and Fe) during the electrocoagulation process serves as a destabilizing agent and neutralizes the electric charge, which removes contaminants. Like tiny magnets, oppositely charged particles unite to form a mass. This method, which has been shown to be quite successful in eliminating pollutants from water (Sahu et al., 2014) , Electrocoagulation equipment is simple, easy to operate , produces little sludge, and low cost The most common electrode material used in the electrocoagulation process is iron. When iron is used as anode, upon oxidation in the electrolytic system, it produces iron hydroxide, Fe(OH)n where n is either 2 or 3 (Dawood et al., 2017).

Electrocoagulation, which relies heavily on the adsorption process that occurs through the electrolysis treatment of various pollutants, such as the removal of pigment (Adeogun and Balakrishnan, 2016), heavy metals (AlJaberi , 2018) , detergent (AlJaberi and Mohammed , 2018), and oil content (Riyanto and Hidayatillah , 2014) . This process is dependent on the electrocoagulation reactor's autocatalytic behavior (Hmood et al., 2019), which creates electro-coagulants as adsorbent without the need to add any external materials (Un and Ocal , 2015)

Materials and methods Materials

Acrylic basin (chain) , Iron plates , aluminum plates , Electrical wires , DC Power Supply(China) , pH-meter (China) , Glass wool(India) , Magnetic stirrer (China) , Silica gel 100-200 mesh(India) , HCl(India), NaOH(India), Dionize water, Crude oil(Iraq), Hexane(Germany), Aluminum oxide Al2O3(India) , Chloroform(India) , Sodium sulfate anhydrous Na2So4(India) .

Methode

Sample preparation

In this study , Testing the effectiveness of electrocoagulation technology in removing total petroleum hydrocarbon from water. Laboratory polluted water was prepared by adding 0.1 crude oil to 10ml hexane then added to 3L dionize water

Coagulation process

Electrical current is passed to the coagulation basin after plates (iron or aluminum) are placed inside it, and the pH of the initial sample is modified using HCl and NaOH. Transfer the 3 L samples to the aquarium, then run the magnetic stirrer until the experiment is complete. Finally, the sample is allowed to settle and a certain amount of it is taken for examination before and after treatment.

The percentage of removal of total petroleum hydrocarbon is calculated using the following equation

$$RE\% = \frac{C_i - C_F}{C_i} * 100 \quad (1)$$

C_i initial concentration , C_f final concentration of pollutants .

Design of experiment;

Because it may be applied to optimization procedures, (RSM) is a statistical technique that is widely employed in experimental designs. The primary parameters (pH, voltages, and time) were optimized using the DOE version (8.0.0.5, State-Ease, Inc., USA). In order to determine the effective variables that affect the coagulation efficiency of cobalt, nickel, and lead as a response (dependent variables), Center Composite Design and response surface methodology were utilized in this work. Many researchers have used (RSM) to remove pollutants from wastewater (Chasib *et al.*,2021)

To estimate the experimental error variance in this investigation, RSM employed a popular type of CCD called the Center Composite Face Design (CCFD), which consists of 2k factor points (k implies factors=3), 2k focus points, and one duplicate at the center point.

The components that were not subject to control were pH (X1), voltages (X2), and time (X3).According to the table (Table 1), these components have three levels: -1 (low), 0 (middle), and +1 (high). Preliminary experiments yielded the actual values for the coded components' levels, which are represented by the codes in Table 1.

Table 1: The three levels of experimental factors.

		Real value of coded levels		
Design Variable (factors)	Symbol	-1	0	+1
pH		5	7	9
V(volt)		15	20	25
Time (min)		30	45	60

To establish the connection between the components (X1, X2, and X3) and the examined responses (Y1, Y2, and Y3), the second-order empirical polynomial equation (Eq. 2) is utilized.

$$Y = f(x) = \beta + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{j=i+1}^{k} \beta_{ij} x_i x_j \dots \dots (2)$$

The response equation, denoted as Y (after TPH elimination),β0 ; fixed coefficient , βi ; linear model coefficient , βii ; coefficient of square model , and βij ; quadratic model coefficient . The number of variables that are independent of each other k. The coded values of the independent variables are Xi and Xj.

1. Results and discussion ;

statistical evaluation:

As indicated in Table 2, a total of 15 experiments were conducted to investigate the parameters of the independent variables (pH, voltages, and time) in the coagulation process. To determine the correlation of the experimental data and to obtain the regression equation for each response, a variety of models, including square, linear, and two-way interactions, can be employed. Since the square model had the lowest F and P values and the best representation of the correlation between all responses and the experimental data, it was based on the experimental data. Consequently, this model was used. However, as the linear model did not yield adequate data to estimate the model coefficients, it was not suggested for this investigation. The final square model is displayed in terms of the factors encoded in equations (Eqs. 3and 4) for each response (Y TPH by Fe plates , Y TPH by Al plates):

(At Fe plates) $Y_{TPH} = -144.962 + 6.652X_2 + 2.532X_3 - 0.512X_1X_2 - 0.254X_1X_3$ ---(3)

(At Al plates) $Y_{TPH} = 236.337 + 0.962X_1X_2 - 0.083X_3^2$ ---(4)

Where the responses (dependent variables) to the TPH removal efficiency are (Y TPH by Fe plates, Y TPH by Al plates). The encoded factors (independent variables) for the samples' pH, voltages, and time are X1, X2, and X3.

Analysis of variance (ANOVA). An ANOVA analysis was performed on each response's final quadratic model findings to determine the "goodness of fit." There are certain statistically insignificant values in the empirical equations (equation 3 and equation 4) with the lowest F value; as a result, these terms must be removed from the response equations. For the models (TPH removal by Fe plates, TPH removal by Al plates), the P value (P>Prob) in terms of each confidence equation is more significant than the probability (p-value) and the 95% confidence loss (Voltages, time, pH*voltages, and pH*time) are important model terms in the TPH removal by Fe plates, but (pH*Voltages and time*time) are important model terms in the TPH removal by Al plates. as it is smaller than 0.1.

Their interaction R^2 is a metric that quantifies the extent to which the experimental variable can account for variation in the observed species' values. The better the model adopts the answer, the closer R^2 is to 1 (Ghafari *et al* ., 2009) . TPH removals by Fe plates had a R^2 value of 0.9171, and TPH removals by Al plates had a R^2 value

Table 2: Experimental design and responses

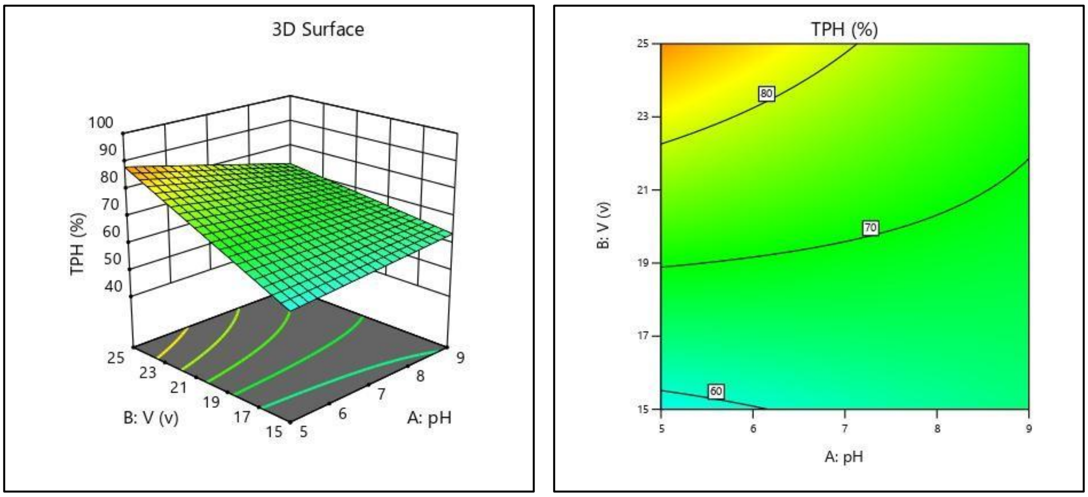
Experimental design coded				**Result**	
Run No.	pH	Voltage (v)	Time (min)	**TPH** removal% by iron plates	**TPH** removal% by aluminum plates
1	5	15	30	45	85
2	5	25	60	95	51
3	7	20	60	78	57
4	9	25	30	76	69
5	9	25	60	66	76
6	5	15	60	73	80
7	9	20	45	71	78
8	9	15	60	59	49
9	7	25	45	88	84
10	7	15	45	64	91
11	5	25	30	80	68
12	7	20	45	64	82
13	9	15	30	67	65
14	7	20	30	65	61
15	5	20	45	71	74

of 0.9031. This, of course, shows that the observed and expected response values have a strong reliance and association. The adjusted R^2 for the TPH removals by Fe plates was 0.8550, and the adjusted R^2 for the TPH removals by Al plates was 0.7287. These values are in close proximity to the R^2 of each response equation, indicating that the estimate of the experimental data is deemed satisfactory.

Coagulation process analysis. The effects of the experimental factors were examined using the design-expert software as 3D surface and contour plots for the model (Eqs. 3 and 4), as well as the graphs shown in figures (Figs. 1, 2, 3, 4, 5, and 6) that illustrate the responses of the experimental variables and can be used to identify the primary interaction between the variables.

TPH removal by Fe plates : The highest percentage of removal of total petroleum hydrocarbons using iron plates is 95% at a pH of 5 and a voltage of 25V in a time of 60 minutes. The observed decrease of TPH concentration versus pH can be explained by taking into account the sacrificial electrode material, where the anode is oxidized, leading to the release of Fe^{+2} into water, and water molecules are decomposed at the same time at the cathode, leading to the generation of hydrogen gas and oxygen molecules. Because of The presence of oxygen oxidizesFe^{+2} , which is soluble in water, to Fe^{+3} , and depending on the pH of the water, it produces gelatinous hydroxide precipitates. Consequently, hydrocarbon molecules, which are hydrophobic, neutral, and non-polar compounds, collect on the surfaces of ferric hydroxide agglomerates and are subject to physical absorption (Moussavi *et al.*, 2011)
. The increase in absorbed metal ions increases with increasing time until equilibrium is reached and reaches its maximum value at 60 minutes (Salih *et al.*, 2012).

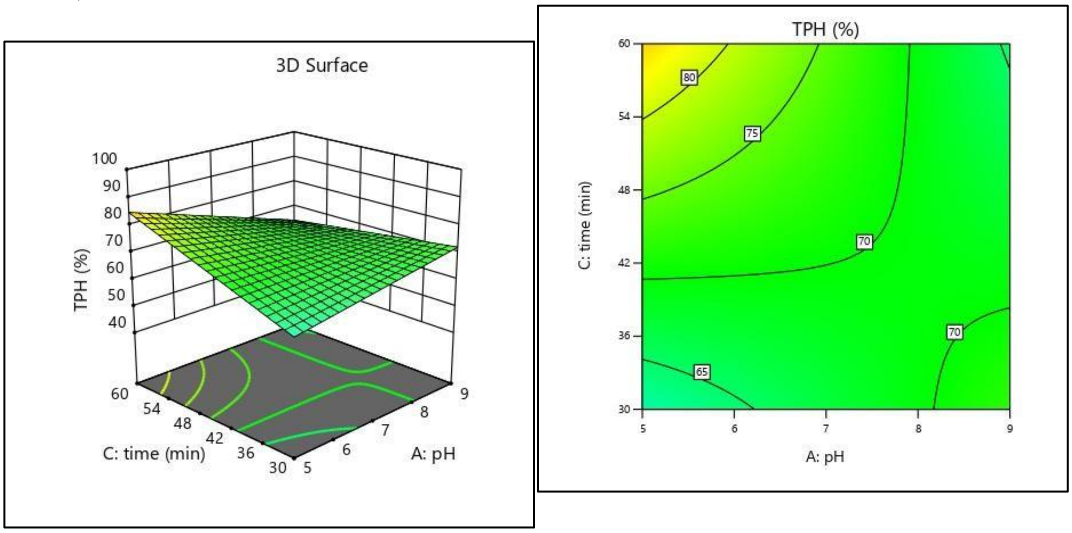
The three-dimensional surface drawing and the layer drawing (Figure.1) show the relationship between pH and voltage in removing total petroleum hydrocarbons from polluted water, as the removal process increases with the range of pH (5-6) and voltage (23-25v). The removal rate decreases at pH (7.5-9) and voltage (15-20).



a **b**

Figure (1): (a) 3D drawing and (b) a layered drawing showing the relationship between pH and voltage in removing total petroleum hydrocarbons from laboratory polluted water using iron plates

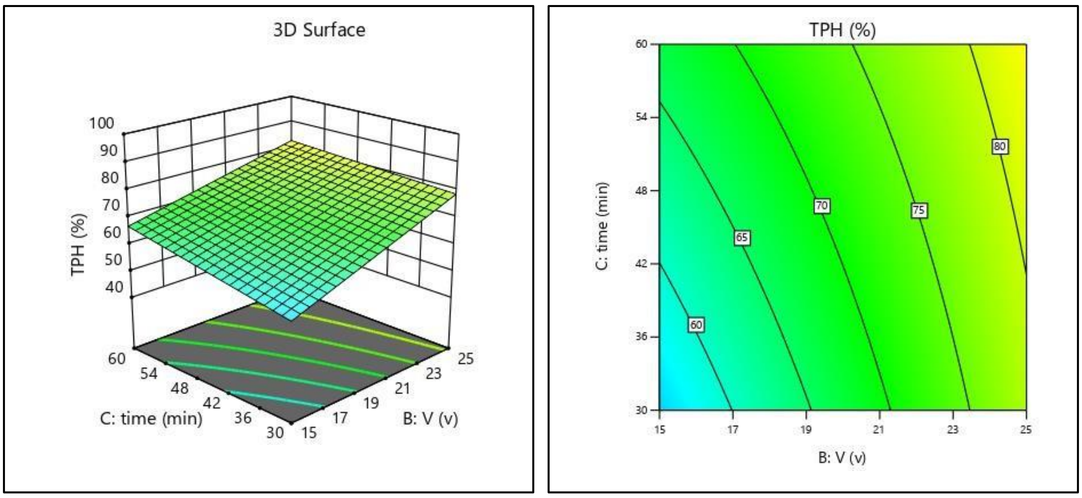
While the three-dimensional and layered drawing in Figure (2) shows the relationship between pH and experiment time in removing total petroleum hydrocarbons, where the removal rate increases at pH (5-6) and at test time (45-60) minutes, but the removal rate decreases at pH (8-9) and experiment time (30-40).



a b

Figure (2): (a) 3D drawing and (b) a layered drawing showing the relationship between pH and time in removing total petroleum hydrocarbons from laboratory polluted water using iron plates.

With regard to the relationship between voltage and experiment time in the efficiency of removing total petroleum hydrocarbons, the removal rate increases at an electrical voltage difference (20-25v) and an experiment time of (42-60) minutes. However, the removal rate decreased at an electrical potential difference of (15-19) and a time of (30-40) minutes. As shown in Figure (3).

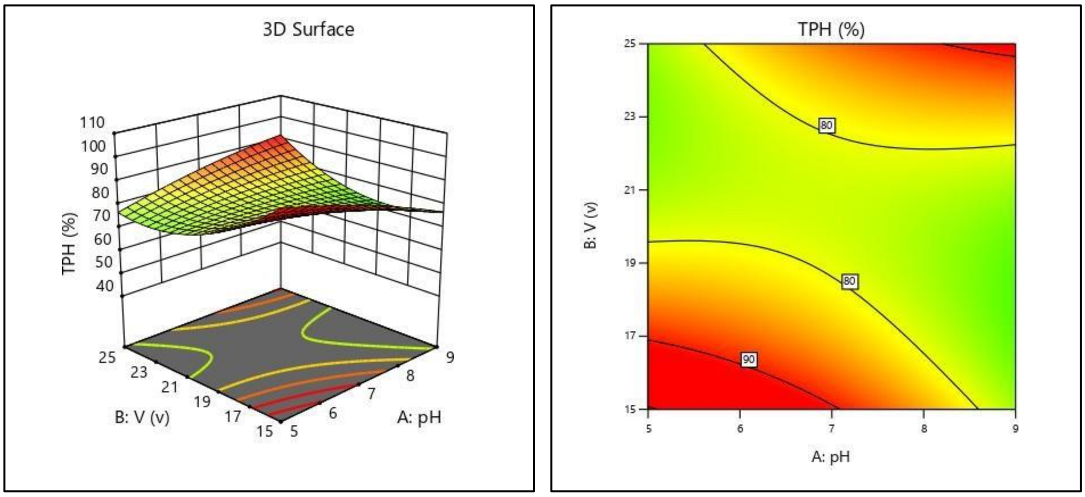


a b

Figure (3): (a) 3D drawing and (b) a layered drawing showing the relationship between voltage and time in removing total petroleum hydrocarbons from laboratory polluted water using iron plates.

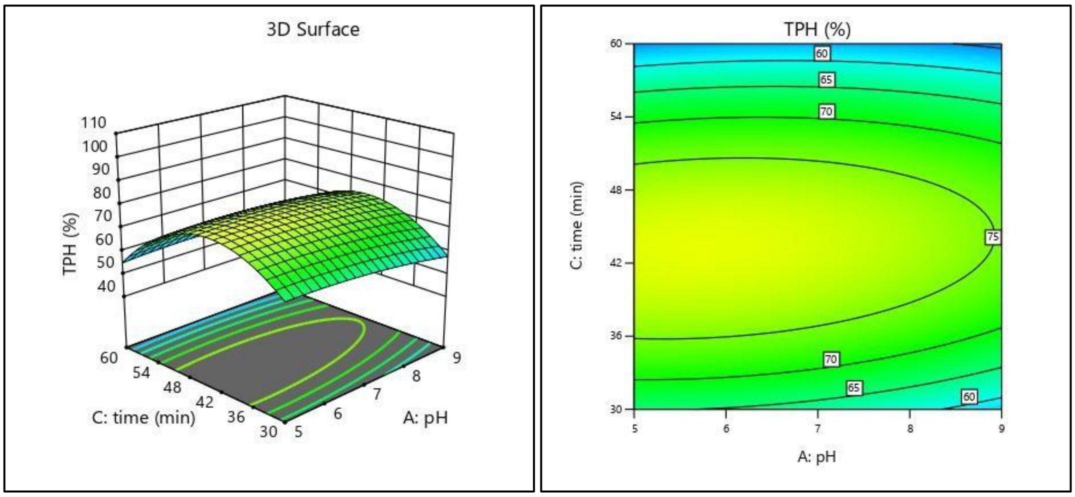
TPH removal by Al plates : The highest removal percentage of total petroleum hydrocarbons using aluminum plates is 91% at a pH of 7 and a voltage of 15v in a time of 45 minutes . This is due to the electrochemical dissolution of the aluminum electrode, and this is especially important on the cathode surface. When the pH value increases, the amount of hydroxide ions in the solution increases, and thus the efficiency of removing petroleum compounds decreases. These results are consistent with other researchers (Moussavi *et al.*, 2011 ; kobaya *et al* ., 2006) .
The three-dimensional surface drawing and the layer drawing (Figure 4) show the relationship between pH and voltage in removing total petroleum hydrocarbons from polluted water, as the removal process increases with the range of pH (5-7) and voltage (15-17v). The removal rate decreases at pH (8-9) and voltage (21-25v).

While the three-dimensional and layered drawing in Figure (5) shows the relationship between pH and experiment time in removing total petroleum hydrocarbons, where the removal rate increases at pH (5-8) and at test time (36-50) minutes, but the removal rate decreases at pH (8-9) and experiment time (30-35).



a **b**

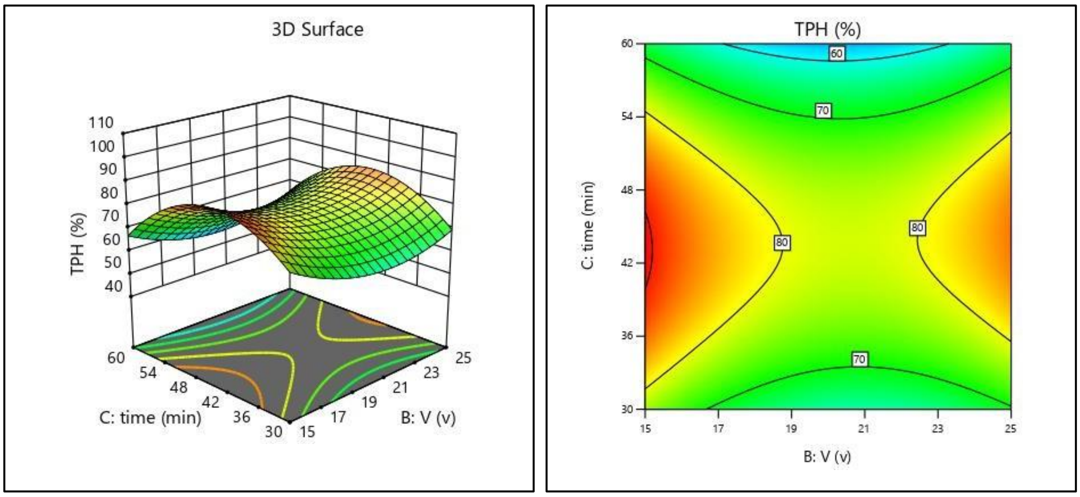
Figure (4): (a) 3D drawing and (b) a layered drawing showing the relationship between pH and voltage in removing total petroleum hydrocarbons from laboratory polluted water using aluminum plates.



a **b**

Figure (5): (a) 3Ddrawing and (b) a layered drawing showing the relationship between pH and time in removing total petroleum hydrocarbons from laboratory polluted water using aluminum plates.

With regard to the relationship between voltage and experiment time in the efficiency of removing total petroleum hydrocarbons, the removal rate increases at an electrical voltage difference (23-24v) and an experiment time of (35-50) minutes. However, the removal rate decreased at an electrical potential difference (17-22v) and a time of (30- 34) minutes. As shown in Figure (6).



a **b**

Figure (6): (a) - Three-dimensional drawing and (b) a layered drawing showing the relationship between voltage and time in removing total petroleum hydrocarbons from laboratory polluted water using aluminum plates.

Conclusion: Using electrocoagulation technology to treat water contaminated oil is very good, as the removal rate is at a lower cost. The total petroleum hydrocarbons were removed from the water using both plates (aluminum, iron), but the removal rate from the aluminum plate was higher than using the iron plate under different conditions (pH, Voltages, and time). TPH removal using iron plates was 95% and the TPH removal using aluminum plates was 91%.

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المقارنة بين كفاءة ألواح الألمنيوم وألواح الحديد في إزالة الهيدروكاربونات النفطية من المياه الملوثة باستخدام التخثير الكهربائي

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الملخص : تعتبر مياه الصرف الصناعي الزيتية المصدر الرئيسي للتلوث في النظم البيئية المائية ، وتم استخدام تقنية التخثير الكهربائي لمعالجة المياه الملوثة بالهيدروكربونات البترولية الكلية. حيث استخدم برنامج تصميم التجارب للحصول على أفضل النتائج مع العوامل المحددة مسبقاً (الرقم الهيدروجيني، فرق الجهد ، والوقت). استخدم ألواح الحديد وألواح الألمنيوم لمعرفة أيهما أكثر كفاءة في معالجة المياه الملوثة ، أعلى نسب إزالة لإجمالي الهيدروكاربونات باستخدام ألواح الحديد (95%) تحت الظروف التالية ، الرقم الهيدروجيني (5)، الجهد (25v)، الوقت (60min)، بينما عند استخدام ألواح الألمنيوم فإن نسبة إزالة المركبات الهيدروكاربونية كانت (92%) عند الرقم الهيدروجيني (7) والجهد (15v) والوقت (45min).

الكلمات المفتاحية: التخثير الكهربائي؛ ألواح الحديد ؛ ألواح الألمنيوم ؛ TPH