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Petroleum refinery wastewater degradation by heterogeneous-electro-fenton process using activated carbon loaded with Iron and Cerium: A kinetic study

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Abstract

This work aims to study the kinetics of heterogeneous electro-Fenton (HEF) process used to treat petroleum refinery wastewater by a catalyst made of an activated carbon loaded with iron and cerium. Effects of HEF operating variables such as current density, dose of catalyst, and pH on the removal of COD and reaction rate constant (k_{app}) were examined. Results showed that decline of COD with time at different operating conditions obeys a pseudo-first-order kinetics with a regression fitting (R^2) not less than 0.97. Furthermore, increasing the current density was found to give higher rate of COD removal and k_{app} to a limit beyond which no longer enhancement can be achieved. Similar behaviour regarding to the dose of catalyst while increasing pH has an adverse effect. The best operating conditions that give high rate of COD removal and k_{app} were a current density of 10 mA/cm^2, a dose of catalyst equal to 1.0 g/L, and a pH of 3, in which 86.97% of COD was removed with k_{app} value equal to 0.0188538 min^{-1} which required only13.06 KWh/kg COD as an electrical energy consumption. The present system approved to have high reaction rate and could be applied to treat various types of wastewaters.

Keywords: petroleum refinery wastewater; pseudo first order kinetics; heterogeneous electro-Fenton; Advanced oxidation process.

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1- Introduction

Water is very important for life and human beings, and its availability is the main task of all industrial sectors [1]. However, a large amount of wastewater is generated from these sectors and disposed of without a proper management or treatment [2]. Petroleum refinery is one such sectors that has generated a huge amount of wastewater and faces treatment difficulties to discharge these effluents in a safe manner [3]. Refinery wastewater contains various organic and inorganic compounds, including phenols, hydrocarbons, sulfides, and heavy metals making it very dangerous to the aquatic system [1, 3]. Various types of treatment methods have been applied for petroleum refinery wastewater, including biological, chemical, and physical methods [4-10]. However, these methods are inefficient, unable to decompose the recalcitrant organic compounds, generate a large amount of sludge, and require a large and expensive space [4, 5].

Advanced oxidation processes (AOPs) are other technologies with a basic toolkit that enables them to degrade a variety of organic contaminants [11]. These processes produce highly reactive •OH, which rapidly oxidizes organic contaminants, converting them to less harmful molecules, most precisely to carbon dioxide and water [12]. In general, AOPs include the Fenton reaction, ozonation, and UV/H_2O_2 methods. These methods have the potential to increase the efficiency of petroleum refinery wastewater treatment by removing refractory or non-biodegradable contaminants that are difficult to remove with the conventional methods, providing cleaner and safer discharges to the environment [3, 13].

Electrochemical advanced oxidation processes (EAOPs) are among the most widely used groups of advanced oxidation processes in industrial wastewater treatment [14]. These technologies are characterized by their low cost, high removal rate, ease of operation and scale-up, and no generation of secondary toxic pollutants [15]. These processes include anodic oxidation (AO), electro-Fenton (EF), sonoelectro-Fenton, and photoelectro-Fenton [16-22].

EF, one of the well-known EAOPs, can produce a strong oxidizing agent (•OH), with a standard electrode potential of 2.8 V/SHE, via the Fenton reaction (Eq. 1), thereby degrading most pollutants in PRW [23].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH\bullet \quad (1)$$

Where H_2O_2 is produced via reducing O_2 supplied from air/oxygen at the cathode (Eq. 2):

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (2)$$

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Based on the properties of the catalyst involved in this process, EF can be divided into homogeneous and heterogeneous processes [24, 25]. In conventional homogeneous EF, the catalyst is an externally added soluble iron salt that is ionic in the solution, making it difficult to separate and recycle, resulting in the generation of a large amount of iron sludge as the solution pH is adjusted to a neutral value [26]. This, in turn, increases the operating cost. Furthermore, homogeneous EF suffers from low H_2O_2 generation rate, low current efficiency, a narrow pH operating range (2-4), and a long reaction time [27]. Compared with the homogeneous type, heterogeneous electro-Fenton (HEF) has many advantages such as catalyst reusability, working near neutral pH, low cost, and long service life [28]. Accordingly, the selection of a suitable catalyst has become a new research focus. Various types of HEF catalysts have been used such as chalcopyrite [29], pyterite [30], Fe-based bimetallic catalyst [31], and α-FeOOOH [32]. Among which zero-valent iron catalysts are the best due to their low cost, strong reducing ability, and good performance [33–36].

However, these catalysts are difficult to handle or store due to their aggregation property [37]. Therefore, they are preferably supported on activated carbon to solve this problem [38, 39].

Studying the kinetics of EAOPs offers numerous advantages from both an economic and engineering perspective. Kinetic data provide insight into the rate of contaminant degradation under specific operating conditions, enabling the identification of optimal EAOPs based on high removal rates and low energy consumption. Furthermore, kinetic parameters are important for scaling up a system from a laboratory scale to a pilot plant scale and then to an industrial scale, providing a basis for determining reactor size, operating time, and electrode surface area. Kinetic analysis prevents overtreatment by stopping the process at the point of diminishing returns, by linking the degradation rate to the specific energy consumption [40]. In the EAOPs, specifically electro Fenton and heterogeneous electro Fenton, kinetics of Fenton reactions happened during the process are very complex due to involve a large number of reaction steps that performed simultaneously and not macroscopically detectable [41]. It was found that in these processes, the oxidation of organic pollutant as well as their by-products is performed by •OH radicals attacking, and reaction rate can be expressed by the following equation in terms of COD as a concentration of pollutants [42-46]:

$$r = \frac{dCOD}{dt} = k[HO^{\bullet}]^{\alpha} COD \quad (3)$$

In Eq. 3; k represents the reaction rate constant, α is the reaction order of hydroxyl radicals' concentration, However, the ratio of the COD to •OH was low and the concentration of •OH radicals can be considered constant. In this case Eq. 3 can be written as follows:

$$r = \frac{dCOD}{dt} = k_{app}\, COD \quad (4)$$

Where $k_{app} = k[^{\bullet}OH]^{\alpha}$ represents the apparent rate constant. The integration of Eq. 4 subject to the initial conditions $COD=COD_o$ at t=0 leads to the following equation:

$$COD = COD_o \exp(-k_{app} t) \quad (5)$$

This equation is termed as a pseudo first order kinetic in which k_{app} is known as a pseudo-first-order kinetic constant which could be calculated from the plot of ln (COD_o/COD) versus t [47].

Many kinetics studies have been reported for treating various organic pollutants using heterogeneous electro-Fenton process [23, 41, 42, 48-53]. However, studying the kinetic of COD removal from petroleum refinery wastewater using AC loaded with Fe+Ce in a HEF process was not reported previously. AC loaded with Fe+Ce was found by our previous work [54] has good catalytic activity in removal of methylene blue due to the incorporating CeO_2 with Fe@Fe_2O_3 which played an essential role since CeO_2 has large amount of oxygen vacancy defects resulted from the redox conversion between Ce^{4+} and Ce^{3+}causing more activating of H_2O_2 to generate more •OH radicals during the process [55].

Therefore, the aim of this study is to examine the kinetic of COD removal by AC loaded with Fe+Ce as HEF catalyst using a new design of electrochemical reactor consisting of cheap electrodes and easy to scale-up. Impacts of operating factors such as current density, dose of catalyst, and pH on the removal rate of COD and pseudo first order kinetic rate constant were investigated.

2- Materials and method

2.1. Chemicals

Commercial activated carbon (AC) was purchased from Zhengzhou Kelin Company, China. It has the following specifications: total surface is 950-1200 m^2/ g, apparent density 460± 40kg/m^3, ash content 5%, moisture content 8%. Sodium borohydride ($NaBH_4$, ≥99%) was purchased from Fluka-Garantie. $FeCl_3.6H_2O$ with a purity of 97%was purchased from Loba Chemie PVT Ltd,India. Ce $(NO_3)_3.6H_2O$ with a purity of 98% was purchased from Himedia Laboratories Pvt.LTD,India. Acetone, ethanol, Na_2SO_4, HCl, and NaOH were analytical grade and used without needing for purification. All runs were performed using distilled water with a conductivity of 0.05 µS/cm.

The performance of the AC catalyst loaded with Fe/Ce (2:1) in COD removal from petroleum refinery wastewater generated at Al-Dora refinery plant located at Baghdad province, Iraq was evaluated using the heterogeneous EF. The wastewater had the characteristics listed in Table 1.

2.2. Synthesis of AC loaded Fe/Ce

Synthesis of the catalyst was performed according to our previous work [54]. Firstly, 20g of AC was sieved, and the cut of 40 to 60 mesh was isolated and used in

preparing the catalyst. The isolated amount was soaked in 5% HCl for 24 h, washed with distilled solution, filtered, followed by measuring the pH of filtrate. This cycle was repeated several times until the pH of the solution reached 6.5–6.7. The washed AC was dried at 105 °C for 24 h and stored until used as a support material. The composite $Fe@Fe_2O_3$ and CeO_2 were loaded onto AC as follows: Appropriate amounts of $FeCl_3.6H_2O$ and $Ce(NO_3)_3.6H_2O$ were dissolved in 100 mL of a 30/70, v/v, ethanol-water mixture at pH 4. The prepared solution had a total concentration of 0.07 M for Fe+Ce. Then, 4 g of AC was added to the above solution, and the resulting solution was mixed for 60 min at room temperature and 500 rpm. 50 mL of 0.5 M $NaHB_4$ was prepared and added dropwise (2 mL/min) to the AC-containing solution, mixing for another 30 min. The resulting catalyst was filtered, washed with ethanol and distilled water three times each, and dried at 70 °C for 6 h.

2.3. Electrochemical system

The heterogeneous EF system consists of an electrochemical cell, a power supply UNI-T, UTP3315TFL-II, China), an overhead stirrer (Heidolph overhead stirrer company, Germany), an air pump (HAILEA, Aco-318, China), an Ammeter (UNI-UT39C$^+$, China), and an air flow meter (Flowtech-z-7002, China).

The heterogeneous EF system is based on an electrochemical cell with a novel design. It consists of a cell body and its cover, a microporous air diffusion cathode, and a porous graphite anode. The cell body is a cylindrical Perspex vessel (outer diameter = 90 mm, inner diameter = 80 mm, length = 160 mm), while its cover is a disc made of the same material (outer diameter = 120 mm, thickness=10 mm) enclosed inside the cell body. The cover has three silts: a central silt (40 x 20 mm), a slit for inserting the cathode (40 x 15 mm), and a slit for inserting the anode (40 x 5 mm). The gap between the anode and cathode was fixed at 25 mm. The anode is a porous graphite plate (150 x 40 x 4 mm). The air-diffused cathode consists of two parts: a graphite plate and a Perspex brood cavity. The graphite plate (139 x29 x 4 mm) was machined with 300 µm holes with a total of 60 holes, which are evenly distributed at the lower section of the graphite plate (60 x 30 mm). While the rectangular brood cavity has dimensions (150 x 40x 15 mm), Fig. 1 shows the schematic of the heterogeneous EF system.

Table 1. Characteristics of wastewater

Characteristic	pH	TDS	Phenol	COD	BOD	Oil	Cl^-	Turbidity
Value	6.6	ppm	18.5 ppm	307 ppm	140 ppm	20.2 ppm	840 ppm	35 NTU

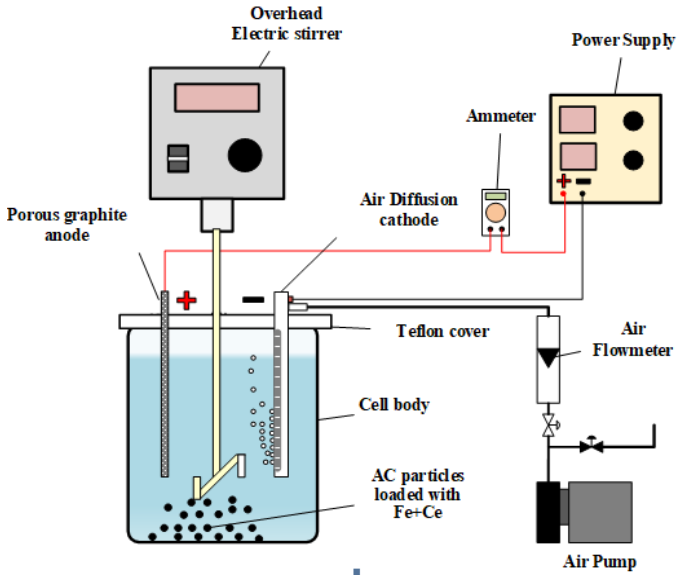

Fig. 1. Schematic of the heterogeneous EF system

2.4. Analysis and measurements

The crystalline structure and morphology of prepared catalyst was examined using X-ray diffraction (XRD) device, Panalytical X`pert Pro, Netherlands and scanning electron microscopy (SEM) device, Czech Republic.

Lovibond thermo-reactor was utilized to measure the COD via heating up the reagents to 150 °C for 2 hours. Turbidity was measured by the turbidity meter-TB 210 IR Lovibond, Germany. The removal efficiency of COD or any phenolic compound can be evaluated based on Eq. 6 [44]:

$$RE\% = \frac{C_i - C_f}{C_i} \times 100 \quad (6)$$

Where C_i stands for the initial concentration (mg/L), C_f stands for the final concentration (mg/L), and RE% stands for removal efficiency.

The specific energy consumption (SEC) for treatment process was calculated using Eq. 7 [45]:

$$SEC = \frac{U*I*t*1000}{(CODo-COD)*V} \quad (7)$$

Where SEC stands for specific energy consumed (kWh/kg COD), U, for cell voltage (Volt), I, for current intensity (A), t, for time (h), V, for water volume (L), COD_o and COD_f are the initial and final chemical oxygen demands (mg/L). The operating variables and their ranges used in the present work is illustrated in Table 2.

Table 2. Operating variables and their range

Variable	Range
Current density(mA/cm^2)	5,10,15
pH	3,6,9
Dosage of catalyst (g/L)	0.5,1,1.5

3- Results and discussion

3.1. Characterization of the catalyst

To demonstrate the success of loading process for the activated carbon, XRD and SEM tests were performed as shown in Fig. 2 and Fig. 3. Fig. 2 displays XRD results for the prepared catalyst where no sharp peak can find at 2θ values between 20º and30º confirming amorphous structure of carbon [55]. A clear peak of zero valent iron was observed at 2θ of 44.7º (JCPDS, NO. 06–0696) [56, 57]. While peaks of Fe_2O_3 was observed at 2θ of 35.6º (JCPDF, NO. 89–0691) [58]. Two peaks of CeO_2 were detected at 2θ of 33.32º, 56.76º (JCPDF, NO. 34–0394) [58, 59].

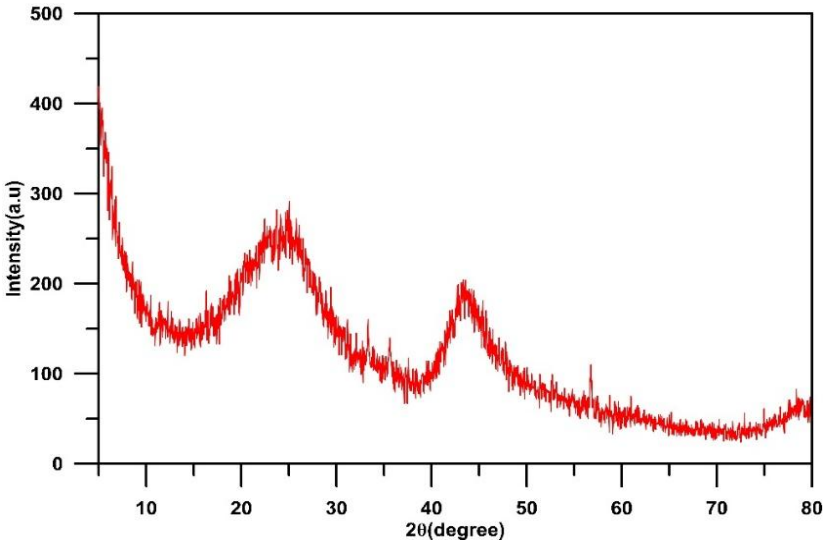

Fig. 2. Shows SEM of catalyst where results showed good immobilized of zero-valent and cerium oxide within the latex of AC. Similar observation were noted in previous works [60]

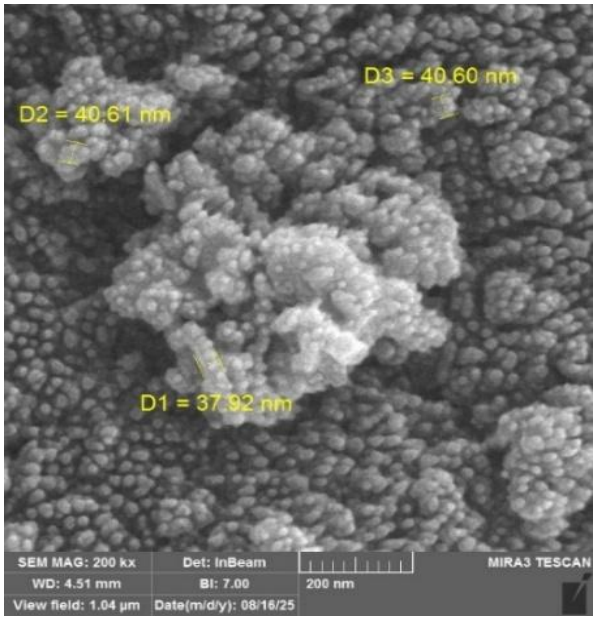

Fig. 3. SEM of the catalyst

3.2. Effect of operating variables

3.2.1. Effect of current density

Fig. 4 displays the decay in COD with time at different current densities for operating time of 120 min at constant catalyst dose (1 g/L) under initial pH of 6, while Fig. 5 represents the corresponding plot of ln (COD/COD_o) verses time. The results show that the decrease in COD with time exhibits an exponential behavior, while a straight linear relationship was found for ln (COD/CODo) with time, confirming pseudo first-order kinetics. Also increasing current density results in increasing COD removal to a maximum value beyond which start to decrease with further increasing in its value where a maximum removal efficiency of 84.5% was obtained at a current density of 10 mA/cm^2 as shown in Table 3. In fact, increasing current would accelerate the formation of H_2O_2 on the cathode according to Eq. 2 and activating the oxidation power of the catalyst to generate further $^{\bullet}OH$ radicals [61]. Also, at the same time, regeneration of Fe^{2+} by reduction of Fe^{3+} on the cathode (Eq. 8) increases with increasing current density leading to more degradation of COD [48].

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (8)$$

Conversely, high current density could lead to reduce the rate of H_2O_2 generation due to parasitic reactions (reducing O_2 to H_2O instead of H_2O_2 (Eq. 9) and H_2 generation (Eq. 10) occurred on the cathode [62]:

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad (9)$$

$$2H^+ + 2e^- \rightarrow H_2 \quad (10)$$

Table 3 shows the value of k_{app} at different current densities, with the corresponding regression coefficient (R^2) having values above 0.980. At 5 mA/cm^2, k_{app} was 0.01508 min^{-1} which increased to 0.01738 at 10 mA/cm^2 leading to an increment of 20% in its value. However, k_{app} decreased by 10% in further increasing of current density. Thus, increasing the current density improves the reaction rate to a maximum value beyond which it is not recommended to further proceed the reaction. Table 3 displays that increasing the current density led to increase the electrical energy consumption and a current density of 10 mA/cm^2 is preferred in terms of lower energy consumption with higher removal of contaminants.

A similar observation regarding the effect of current density on k_{app} was confirmed by Pormazar and Dalvand [41] in the degradation of DB80 dye using HEF with Fe_3O_4 magnetite-loaded AC. Xu et al. [23] found that the maximum reaction rate constant (0.0201/s) was obtained at a current density of 5 mA/cm^2, which then decreased with further increase in current density during the treatment of a polyacrylamide contaminant using HEF with $CoFe_2O_4$ catalyst. Tan et al. [48] also found that increasing the current density from 10 to 30 mA/cm^2 resulted in a significant enhancement in the kinetic constant from 0.0244 to 0.12 min^{-1}. However, it subsequently decreased with increasing current density during the treatment of Rhodamine B by HEF with nano-calcined pyrite.

3.2.2. Effect of Catalyst dosage

Fig. 6 shows the decay in COD with time at different catalyst dosages for operating time of 120 min at 10 mA/cm^2 and initial pH of 6. While Fig. 7 denotes the corresponding plot of ln (COD/COD_o) verses time. Similarly, the decrease in COD with time was found to follow an exponential behavior, and a straight linear relationship for ln (COD/CODo) with time was observed approving pseudo first-order kinetics for COD removal from PRW. The results show that increasing the catalyst dosage would enhance the COD removal and rate constant to a maximum value after which the removal rate and reaction rate constant decrease for further increase in the catalyst dosage. The increase in both COD removal and k_{app} with increasing in dose of catalyst up to 1g/L could be interpreted by the increase in number of active sites as catalyst quantity increased, favoring more H_2O_2 reaction to generate more reactive oxidant ($HO^{\bullet}$) [62]. The decrease in both COD removal and k_{app} as the dose of catalyst exceeded 1 g/L can be explained by the reaction of Fe^{2+} with $HO^{\bullet}$ (Eqs. 11 and 12) as well as the side reactions (Eqs. 13 and 14) resulted from $HO^{\bullet}$ reactions [48, 63, 64].

$$Fe^{2+} + HO^{\bullet} + H^+ \rightarrow Fe^{3+} + H_2O \quad (11)$$

$$Fe^{2+} + HO^{\bullet} \rightarrow Fe^{3+} + OH^- \quad (12)$$

$$2HO^{\bullet} \rightarrow H_2O_2 \quad (13)$$

$$H_2O_2 + HO^{\bullet} \rightarrow HO_2 + H_2O \quad (14)$$

Table 4 shows the K_{app} value at different catalyst dosages, with the corresponding R^2 values exceeding 0.979. At 0.5 g/L, the K_{app} value was 0.01350 min^{-1}, and it increased to 0.01738 min^{-1}at 1.0 g/L, resulting in a 29% increase in its value. However, the K_{app} value decreased by 11% with increasing catalyst dosage. Therefore, increasing the catalyst amount improves the reaction rate to a maximum value beyond which it is no longer recommended to continue the reaction from economic and recycling aspects. Table 4 shows that increasing the catalyst dosage has a slightly effect on electrical energy consumption.

Similar behaviors have been reported in previous studies; for example, Tesnim et al. [38] found that increasing the AC-NZVI dosage from 0.8 to 1.4 g/L increased pseudo-first order rate constant from 0.87 to 1.73 min^{-1}, with a slight decrease as the catalyst dosage reached 1.6 g/L during the treatment of wastewater containing an antipyrene via HEF using AC-NZVI. In another study conducted by Xu et al. [23], the pseudo-first order rate constant increased from 0.0112 to 0.0256/s with an increase in the $CoFe_2O_4$ catalyst dosage from 0.1 to 0.4

g/L during the treatment of a polyacrylamide contaminant using HEF. However, they found that increasing the catalyst dosage above 0.4 g/L resulted in a negligible improvement in the reaction rate constant. Tan et al. [48] observed that increasing the catalyst dosage from 0.5 to 1.0 g/L increased the rate constant by 1.55 times. However, the rate constant decreased with increasing catalyst amount beyond 1.0g/L during the treatment of Rhodamine B by HEF using calcined nanopyrite. Sadeghi et al. [52] observed a similar trend during the degradation of diclofenac by HEF using a catalyst made of magnetic single-walled carbon nanotubes.

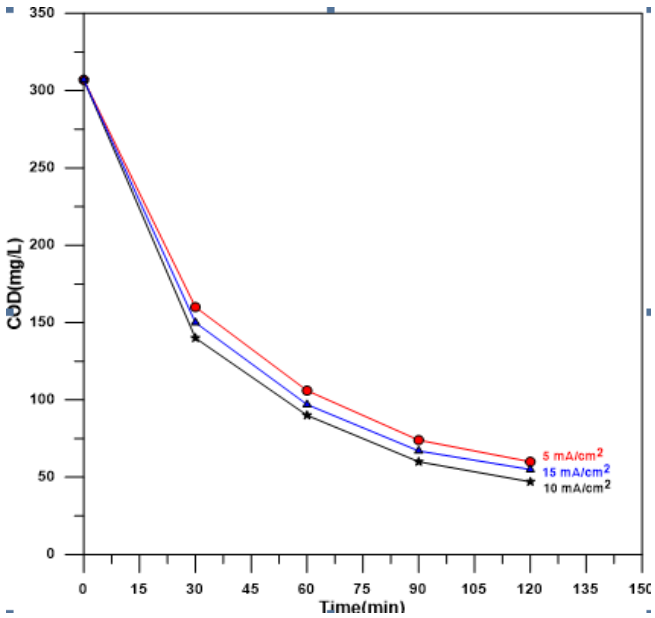

Fig. 4. Effect of the current density on the COD removal in HEF

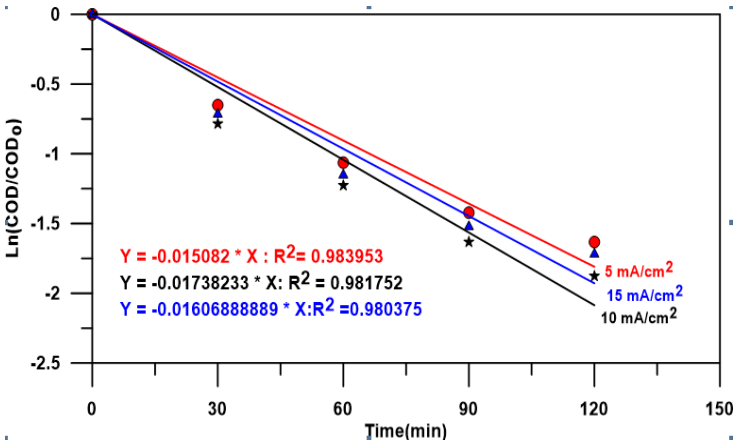

Fig. 5. Pseudo-first order kinetics for different current density at HEF. pH=6,1g/L catalyst

Table 3. Apparent rate constant at different current densities

Current density (mA/cm^2)	Final COD mg/L	RE%	SEC (KWh/ kg COD)	K_{app} (min^{-1})	R^2
5	60	80.45	5.52	0.01508	0.983953
10	47	84.5	13.38	0.01738	0.981752
15	55	82.08	24.83	0.01607	0.980375

Table 4. Apparent rate constant at different catalyst dosage

Dosage (g/L)	Final COD ppm	RE%	SEC (KWh/ kg COD)	K_{app} (min^{-1})	R^2
0.5	73	76.22	15.12	0.01350	0.979540
1.0	47	84.50	13.38	0.01738	0.981752
1.5	53	82.74	14.70	0.01558	0.980793

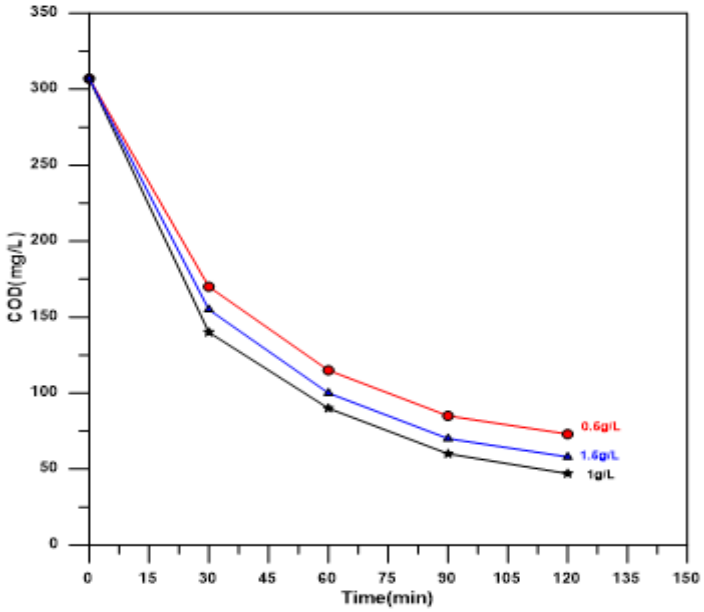

Fig. 6. Effect of catalyst dosage on the COD removal

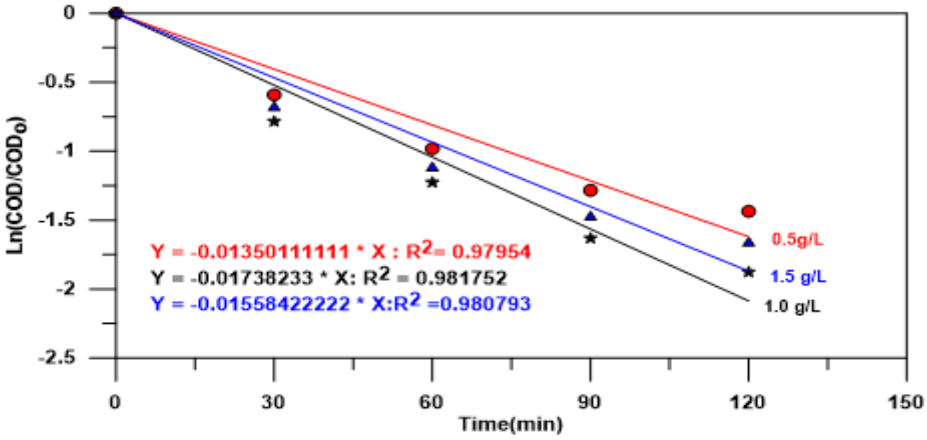

Fig. 7. Pseudo-first order kinetics for different catalyst dosage at HEF. pH=6,10 mA/cm^2

3.2.3. Effect of Ph

PH plays a vital role in controlling the activity of the catalyst, the activities of oxidants and substrates, and Fe species stability during the operation of HEF [38]. Fig. 8 shows the decrease of COD over time at different pH values for an operation time of 120 min at 10 mA/cm² and a catalyst dosage of 1.0 g/L. Fig. 9 shows the corresponding graph of ln (COD/CODo) versus time. Similarly, the decrease in COD over time was found to follow an exponential behavior, and a straight-line relationship was observed for ln (COD/CODo) with time, confirming the pseudo first-order kinetics of COD removal from PRW. The results show that increasing pH resulted in a decrease in COD removal and the rate constant, with both values decreasing significantly after pH 6. The high COD removal at pH 3 can be explained as follows: 1) the Fe and Ce loaded AC surface is prone to higher oxidation degree compared to higher pH leading to more •OH generation [37], 2) the oxidation capacity of •OH is affected by the pH of the solution and its oxidation capacity is diminished at high pH values [65], 3)the increase in pH could result in a significant reduction in rate of in-situ generation of H_2O_2 [23], and 4) at high pH level, H_2O_2 is decomposed to O_2 rather than hydroxyl radicals [66] . Furthermore, the formation of Fe-OOH increases at higher pH leading to its precipitation on the catalyst surface causing a negative effect on •OH generation [67]. Finally, with increasing pH, the resulting iron-proxy complex as an intermediate in the Fenton reaction became more stable leading to less generation of •OH [68].

Table 5 shows the K_{app} value at different pH levels, with the corresponding R^2 values exceeding 0.979. At pH 3, the K_{app} value was 0.0188538 min^{-1}, and it decreased to 0.01738 min^{-1} at pH 6, resulting in an 8.4% decrease. Furthermore, the K_{app} value decreased by 29% as the pH increased from 6 to 9. Therefore, increasing the pH above 6 reduces the reaction rate, which makes it advisable not to continue the reaction in terms of performance. The above results confirmed the ability to conduct HEF in a wide pH range to reach neutrality which cannot be

achieved using conventional homogeneous electro-Fenton technique. Table 5 shows that increasing the pH has a slightly effect on electrical energy consumption.

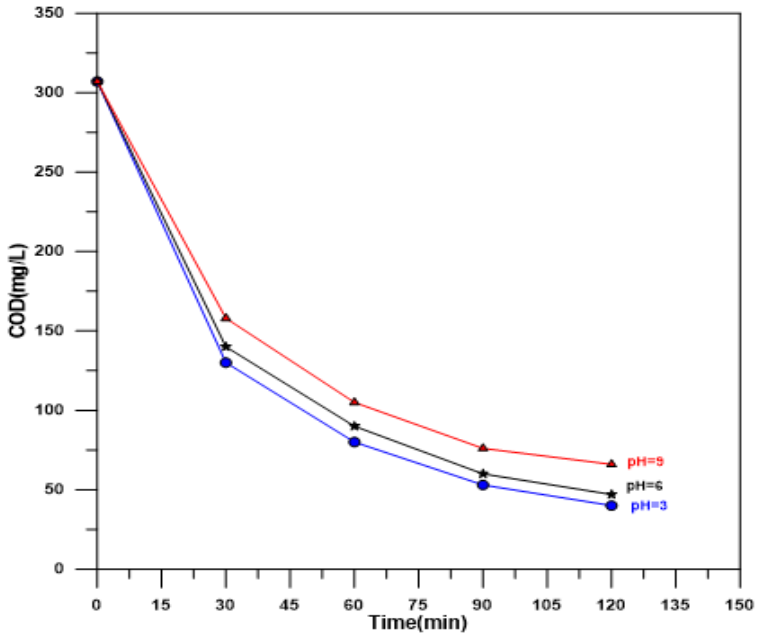

Fig. 8. Effect of pH on the COD removal in HEF

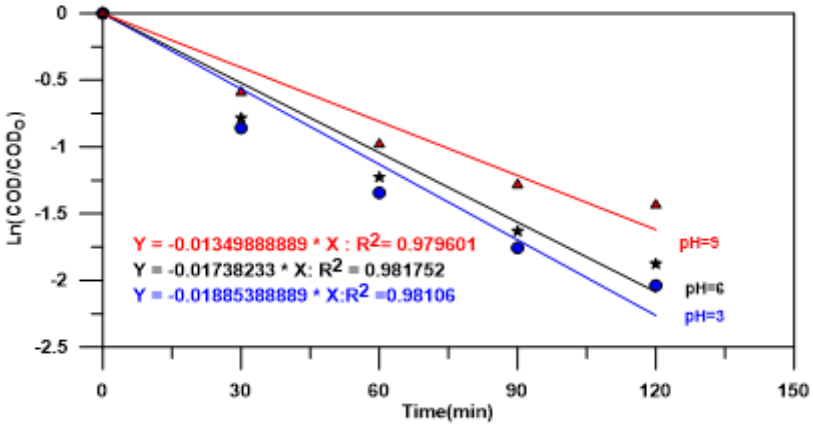

Fig. 9. Pseudo-first order kinetics for different pH at HEF. 1g/L catalyst,10 mA/cm^2

Table 5. Apparent rate constant at different pH

pH	Final COD ppm	RE%	SEC (KWh/ kg COD)	$K_{app.}$ (min^{-1})	R^2
3	40	86.97	13.06	0.0188538	0.98106
6	47	84.50	13.38	0.017382	0.98175
9	66	78.50	15.336	0.013499	0.97960

Previous work has shown different trends, but all confirmed that pH 3 had the highest removal rate and high-rate constant. Tesnim et al. [38] found that increasing pH from 3 to 9 resulted in a decrease in the pseudo-first order rate constant from 1.73 to 0.308 min^{-1}, however, the decrease was 30% with increasing pH from 3 to 5 compared to 82% with increasing pH from 3 to 9 during the treatment of wastewater containing an antipyrene via HEF using AC-NZVI. Xu et al. [23] found that the pseudo-first order rate constant decreased from 0.0112/s at pH 3 to 0.0086/s at pH 6 during the treatment of a polyacrylamide pollutant using HEF. However, they found that the removal rate at pH 5 was still high at 79.63% when compared to the homogeneous Fenton electrolyte, which showed a significant decrease in the removal rate to 45.56%, confirming that HEF has better pH adaptability.

Tan et al. [48] observed that the rate constant at pH 5 was 0.12 min^{-1}, which is much higher than at pH 7 (0.0488 min^{-1}) and at pH 9 (0.0374 min^{-1}), but slightly

lower than at pH 3 (0.1344 min^{-1}) during the treatment of rhodamine B by HEF using calcined nanopyrite. Their results are consistent with ours. Sadeghi et al. [52] also observed a similar behavior during the degradation of diclofenac by HEF using a catalyst made of magnetic single-walled carbon nanotubes. In contrast, Pormazar and Dalvand [41] found no significant effect of pH on DB80 dye removal between pH 3 and 7 in its decomposition using HEF with magnetite-loaded AC Fe_3O_4 and DB80 dye removal decreased slightly at pH 9.

4- Conclusion

The kinetics of HEF process with iron and cerium-loaded AC during petroleum refinery wastewater treatment were successfully investigated. Effects of HEF operating variables such as current density, catalyst dosage, and pH on COD removal and the reaction rate constant (k_{app}) were examined. The kinetic study confirmed that the process follows a pseudo-first-order kinetic model under different operating variables studied in the present work with R^2 higher than 0.97. It was found that the COD removal and the reaction rate constant (k_{app}) increased with increasing current density up to 10 mA/cm^2 and increasing catalyst dosage up to 1 g/L. The effect of pH was significant as pH exceeded 6, and good COD removal and a suitable k_{app} value could be maintained in the pH range between 3 and 6, confirming that the present system has better pH adaptability. High COD removal rate and k_{app} could be attained at a current density of 10 mA/cm^2, a catalyst dosage of 1.0 g/L, and a pH of 3, where 86.97% of COD was removed with recording reaction rate constant of 0.0188538 min^{-1}. The present system is a promising HEF technology for wastewater treatment, with the potential to improve the degradation efficiency and reduce energy consumption. It is also a promising process for overcoming the drawbacks of the homogeneous wastewater treatment process used in the conventional refinery wastewater treatment.

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تحلل مياه الصرف الصحي لمصافي البترول بواسطة عملية إلكترو– فنتون غير المتجانسة باستخدام الكربون المنشط المحمل بالحديد والسيريوم: دراسة حركية

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الخلاصة

تهدف هذه الدراسة إلى بحث حركية التفاعل لعملية إلكترو– فنتون غير المتجانسة (HEF) والمستخدمة في معالجة المياه العادمه الخارجه من مصافي البترول، وذلك باستخدام محفز مصنوع من الكربون المنشط المحمل بالحديد والسيريوم. تم فحص تأثير متغيرات تشغيل عملية HEF والمتمثله بكثافة التيار، جرعة المحفز، ودرجة الحموضة، على إزالة الطلب الكيميائي للأكسجين (COD) وثابت معدل التفاعل (k_{app}). أظهرت النتائج أن انخفاض الطلب الكيميائي للأكسجين مع مرور الوقت في ظل ظروف تشغيل مختلفة يتبع حركية تفاعل من الرتبة الأولى الزائفة، بمعامل تحديد (R^2) لا يقل عن ٠.٩٧. علاوة على ذلك، وُجد أن زيادة كثافة التيار تؤدي إلى زيادة معدل إزالة الطلب الكيميائي للأكسجين وثابت معدل التفاعل (k_{app}) الى حد معين بعده لايوجد أي تحسين إضافي. لوحظ سلوك مماثل فيما يتعلق بجرعة المحفز، بينما كان لزيادة درجة الحموضة تأثير سلبي. كانت أفضل الظروف التشغيل التي حققت اعلى معدل إزالة الطلب الكيميائي للأكسجين (COD) بمقدار ٨٦.٩٧٪ واكبر قيمة لثابت معدل التفاعل(k_{app}) بمقدار٠.٠١٨٨٥٣٨ دقيقة$^{-1}$ هي كثافة تيار ١٠ مللي أمبير/سم^2، وجرعة محفز ١.٠ غ/لتر، ودرجة حموضة ٣،حيث تطلب ذلك استهلاك طاقة كهربائية بمقدار ١٣.٠٦ كيلوواط ساعة/كغ من الأكسجين الكيميائي الحيوي. وقد أثبت النظام الحالي فعاليته في الحصول على معدل تفاعل عالي، ويمكن تطبيقه لمعالجة أنواع مختلفة من مياه الصرف الصحي.

الكلمات الدالة: مياه الصرف الصحي لمصفاة البترول، حركية من الدرجة الأولى الزائفة، إلكترو–فنتون غير المتجانس، عملية الأكسدة المتقدمة.