



Pretreatment of petroleum refinery wastewater by microwave-enhanced Fe⁰/GAC micro-electrolysis

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ABSTRACT

The pretreatment of petroleum refinery wastewater (PRW) was experimentally investigated using the Fe⁰/granular activated carbon (GAC) micro-electrolysis system in the absence or presence of microwave. Effects of reaction time, pH value, Fe⁰ and GAC dosage, Fe⁰/GAC volumetric ratio, and microwave power on the treatment efficiency of wastewater were studied. A significant synergetic effect was observed between microwave irradiation and Fe⁰/GAC. The optimum conditions were determined to be: microwave power 500 W, reaction time 15 min, iron filings dosage 30 g/L, GAC dosage 5.75 g/L, and initial pH 3. Under the optimum conditions, chemical oxygen demand (COD) removal efficiencies were 38.3 and 62.2% in the absence and presence of microwave irradiation, respectively. The biodegradability of the wastewater was greatly improved by the microwave enhanced micro-electrolysis treatment. The results of the present study demonstrated that the pretreatment process was favorable for the subsequent biological process.

Keywords: Internal micro-electrolysis; Microwave; Biodegradability; Pretreatment

1. Introduction

Petroleum refinery wastewater (PRW) is characterized by large volume, high chemical oxygen demand (COD) concentration, and complex compositions [1,2]. PRW contains high concentrations of petroleum hydrocarbons, phenol, benzene and salinity and thus, it can cause considerable environmental hazards if discharged without effective treatment [3]. Nowadays, the petroleum refinery industry is facing a huge challenge in meeting increasingly stringent environmental standards.

The conventional technologies for treating oil-containing wastewater include enhanced photo-degradation [4], combined chemical oxidation-biological treatment [5], biodegradation [6], membrane filtration [7,8], electrochemical methods [9], etc. Compared with physical and chemical processes, biological treatment is a cost-effective and environmentally friendly technique, which is more compatible with existing plant facilities and operation. However, it is difficult to obtain satisfactory treatment efficiency of PRW using bioremediation alone because of its complex nature, thus an appropriate pretreatment process is needed to avoid organic overload or toxic impact on the downstream biological treatment processes.

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As a cost-effective pretreatment method, the IME method, which is based on zero-valent iron (Fe^0) and carbon, has been extensively studied in recent years [10,11]. IME has been successfully used for the pretreatment of many wastewaters because of its particular advantages such as simple process, easy operation, low cost and high efficiency [11]. Iron chips and granular activated carbon (GAC) are commonly used as the basic materials in IME. When iron is in contact with carbon at a certain Fe^0 /GAC mass ratio in the wastewater (electrolyte solution) with a certain initial pH, numerous macroscopic galvanic cells are formed spontaneously between the particles of iron (anode) and carbon (cathode). These galvanic cells are formed on microscopic scales, but simultaneous occurrences of redox reactions on the surfaces of a large number of iron and GAC particles can generate significant electron flows. Thereupon, organic pollutants in the wastewater are reduced by [H] (i.e. atomic hydrogen with a significant reducing power), electrons or removed by adsorption, coprecipitation by ferric and ferrous hydroxides, or oxidized by some oxidative species [10,11].

Because of both its economical advantages and convenience, the MW-induced catalytic degradation process has already been applied to industrial, residential, and environmental pollution scrubbing for organic pollutants [12–14].

In this work, batch experiments were carried out to investigate the feasibility of Fe^0 /GAC IME coupled with MW process as a pretreatment of PRW. The optimal conditions for COD removal from the PRW were determined by evaluating the effects of operating conditions such as reaction time, initial pH, Fe^0 and GAC dosage, Fe^0 /GAC mass ratio and microwave power on the COD removal efficiency.

2. Materials and methods

2.1. Materials

The raw PRW employed in this work was obtained from a petroleum refinery in China. The main characteristics of the wastewater are listed in Table 1.

GAC has a particle size of 1.25–2.05 mm, which was supplied by Hongwei Activated Carbon Company (Chengde, China). The GAC was washed with distilled water and dried at 100°C. Prior to use, it was absorbed in the raw PRW for 30 min until the equilibrium was reached.

The cast iron filings were obtained from one factory of China. The particle size ranged from 1.2 to 2.5 mm. The iron filings were degreased with hexane (purity >99.5%) to remove surface oils. After being

Table 1
Characteristics of raw PRW used in this study

pH	6.8
COD (mg/L)	1,680
BOD ₅ (mg/L)	320
BOD ₅ /COD	0.19
TDS (mg/L)	86
Oil and grease (mg/L)	225
TPH (mg/L)	162
Conductivity ($\mu\text{S}/\text{cm}$)	1,120
Total phenol (mg/L)	75

dried completely by blowing at room temperature, it was re-rinsed with 0.1 M HCl. Then, it was washed with water until the eluate was neutral and dried again before use.

2.2. Experimental methods

The MW-assisted degradation experiments were conducted using a modified household microwave oven. Its maximum output was 850 W, with 2,450 MHz of MW radiation frequency. The rotating plate of the microwave oven was replaced by an aluminum plate, and a magnetic mixer was installed for mixing the reactor contents. A 250-mL long neck, flat bottom flask was used as the reactor. To facilitate sampling, a hole was pierced on the top portion of the oven and the flask was connected with a water condenser to prevent volatilization. Fig. 1 shows the schematic illustration of the experimental apparatus.

The GAC and iron filings were mixed together with a certain mass ratio and then packed in the reactor. About 100 mL of the PRW was added into the flask and then put into the microwave oven and radiated with different power for some time. The mixture was continuously agitated using a magnetic stirrer. To optimize the process parameters of the microwave-assisted Fe^0 /GAC IME system, the experiments were performed under different reaction time (5, 10, 15, 20, 25, and 30 min), influent pH (1, 2, 3, 4, 5, 6, and 7), Fe^0 dosage (5, 10, 20, 30, 40, and 50 g/L), Fe^0 /GAC volumetric ratios (1:1, 1:2, and 2:1), and MW power (100, 200, 300, 400, 500, 600, 700, and 800 W), respectively. In the experiments, pH of the influent was adjusted by adding diluted sulfuric acid (10%) or sodium hydroxide solutions (5 mol/L).

Traditional Fe^0 /GAC IME without microwave irradiation was performed at ambient temperature with the same parameters as microwave-IME reaction. All of experimental runs were performed in triplicate to diminish errors.

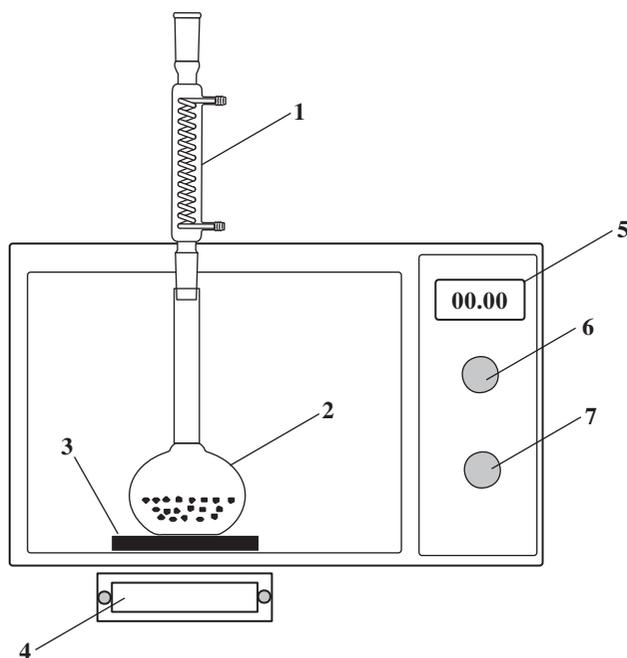


Fig. 1. Schematic illustration of the experimental apparatus. 1: condenser; 2: flask; 3: aluminum plate; 4: magnetic mixer; 5: time display; 6: time adjuster; and 7: power adjuster.

2.3. Analytical methods

The wastewater samples were determined after sedimentation and filtering through a 0.45- μm membrane filter (Millipore). The water quality index was measured according to standard methods [15]. In brief, COD was measured with potassium dichromate digestion in concentrated sulfuric acid for 2 h at 150°C, and the interference by chloride ions was eliminated by precipitating the chloride ions using HgSO_4 reagents. BOD_5 was determined by measuring oxygen concentration in the solution after five days of incubation in a thermostated incubator at 20°C in the dark. Total phenol was detected by the 4-ammonium antipyrine method. Oil and grease was determined by using an infrared spectrophotometry oil-measuring instrument (H3-OCMA-350, Japan) after dichloromethane liquid–liquid extraction. In addition, the details of petroleum hydrocarbons extraction, fractionation, and GC–MSD analysis have been described in our previous work [16].

2.4. Data analysis

All experiments, unless otherwise stated, were conducted in triplicate. The results presented here represent the average values of three independent measurements \pm standard deviations. The variance and

significant differences among various treatments were analyzed by Student's *t*-test. Data were considered to be significantly different among values if $p < 0.05$. All statistical analysis was performed with SPSS 13.0 for Windows.

3. Results and discussion

3.1. Effect of initial pH

The initial pH values investigated were 1–7, respectively. Fig. 2 shows that pH value played an important role in oxidation of organic matter by Fe^0/GAC IME, and acid condition favored the COD removal. When the pH of the wastewater was between 3 and 4, the degradation efficiencies of COD reached above 36 and 60% in the case of IME and MW/IME processes, respectively. However, if the pH range was more than 4, the COD removal dropped dramatically. Additionally, too low initial pH (<3) also led to a decrease in COD removal. Overall, COD removal was increased by approximately 25% due to the introduction of MW irradiation. Interestingly, after reaction, the pH of reaction mixture went up to about 6.8–7.0 from the initial 3 and 4. This would create favorable conditions for the subsequent biological treatment. In these processes, partly organic matter in the reaction system was removed because of the flocculation effect of $\text{Fe}(\text{OH})_3$ at high pHs.

Activated carbon is believed to be an especially efficient microwave absorbent because of its high porosity and the resulting high surface area [12]. Activated carbon can act as direct catalyst in the

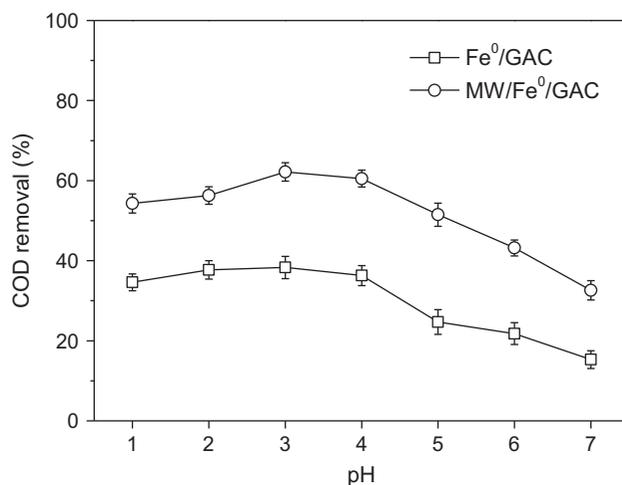
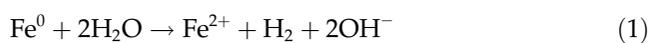


Fig. 2. Effect of initial PRW pH on the COD removal during IME process. Experimental conditions: PRW volume 100 mL; MW power 500 W; Fe^0 dosage 30 g/L; GAC dosage 5.75 g/L; reaction time 15 min. Fe^0 and GAC have the same volume.

catalytic wet air oxidation of organic components in the absence of any active metal [12,13]. Activated carbon particles can strongly absorb MW energy and then form many “hot spots” on their surface [14]. The temperature of “hot spot” on the activated carbon surface under microwave irradiation in water can reach 1,200°C or more [14]. Thus, the organics around the “hot spots” would be thermally decomposed.

In general, low pH is deemed capable of enhancing the reduction of IME [17]. Since corrosion reaction of Fe^0 is affected greatly by the hydrogen ion concentration, the influent pH is an important parameter influencing the kinetics in the pretreatment process by the Fe^0/GAC IME system [17]. Different reactions are involved for Fe^0 in the solution with different initial pHs, which are presented as following equations:



According to Nernst Equation, the reaction rate of $\text{Fe}^0 \rightarrow \text{Fe}^{2+}$ increases with an increment in the H^+ concentration, and thus, the COD removal can be influenced markedly by the influent pH of the Fe^0/GAC IME system. However, if the influent pH is too low, iron would be excessively corroded and its utilization efficiency becomes low. Moreover, the increment in pH results in the precipitation of both ferrous ion and ferric ion, blocking the electron transfer between the Fe^0 and the wastewater [18]. This accounts for the deactivation of the iron surface and the termination of IME.

Thereupon, an initial pH of 3 was selected for further experiments based on the COD removal efficiency.

3.2. Effect of Fe^0/GAC volumetric ratio

Electrochemical reaction between the iron filings (Fe^0) and GAC particles in the PRW is an electrochemical corrosion process. When the iron filings and GAC particles were immersed in the PRW (electrolyte solution), many macroscopic galvanic cells were formed between the iron filings and the carbon particles. In the Fe^0/GAC IME system, the iron particles were used as an anodic metal in $\text{Fe}^0\text{-H}_2\text{O}$ systems, providing electrons for the destruction of organics; meanwhile, GAC particles acted as cathodes to form numerous macroscopic galvanic cells by the touch with iron particles and greatly enhance the current efficiency of the Fe^0/GAC IME system [17]. Thus, the degradation efficiency of PRW would be influenced by the number of cathodes (GAC).

Fig. 3 shows the COD removal of PRW by the Fe^0/GAC IME system with or without MW irradiation at different Fe^0/GAC volumetric ratios (v/v). The results showed that the removal efficiency of COD was the highest with Fe^0/GAC ratio of 1:1 (v/v). The iron and GAC particles had similar particle size, and thus, the surface area ratio of the two particles was almost in accord with their volumetric ratio. The same volume of iron and GAC particles in the Fe^0/GAC IME system means the similar number of anode and cathode, which is beneficial for the formation of macroscopic galvanic cells. The less GAC cannot exert the best internal electrolysis efficiency, while the more GAC may inhabit much more space and cumbered the reaction. Therefore, the volumetric ratio of 1:1 (v/v) for Fe^0/GAC was selected for the following experiments under otherwise identical experimental condition. Liu et al. [17] obtained the same result that the optimum Fe^0/GAC ratio was 1:1 in treating azo dye Acid Orange 7 by ultrasound-assisted internal electrolysis.

3.3. Effect of iron (GAC) dosage

The impact of iron dosage on COD removal was investigated by altering the masses of iron filings and GAC with a fixed Fe^0/GAC ratio (1:1, v/v). As shown in Fig. 4, the removal efficiency of COD increased with increasing the iron filings (also GAC) dosage and reached a maximum when the dosage in iron filings reached to 30 g/L, and finally ended in a decreasing tendency with the further increase of iron filings mass. In general, the number of microscopic galvanic cells is positively proportional to the dosage of iron and GAC

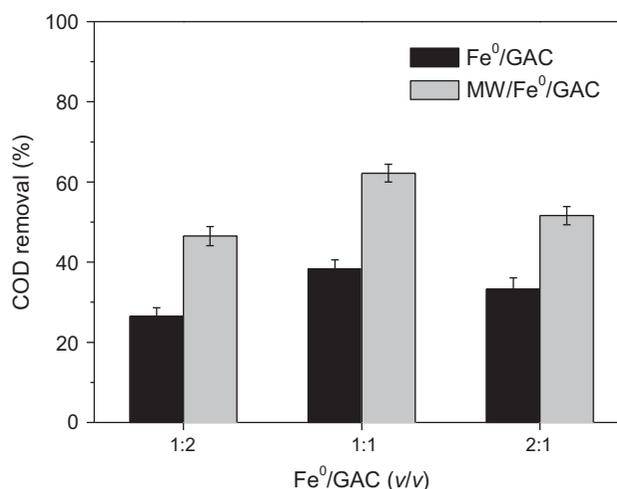


Fig. 3. Effect of Fe^0/GAC volumetric ratio on the COD removal during IME process. Experimental conditions: PRW volume 100 mL; initial pH 3; MW power 500 W; GAC dosage 5.75 g/L; reaction time 15 min.

particles. Thereupon, it is reasonable that the COD removal was enhanced with the increase in iron-filings dosage. However, when excessive iron and GAC particles existed in the treated wastewater, the agglomeration of the excess particles would result in decreased contacting area among iron filings, GAC particles, and wastewater. Additionally, the excess iron would react with H^+ present in the treated solution, leading to weakened reaction from IME [19]. Moreover, GAC utilization efficiency would decrease remarkably if its dosage is too high [20]. Therefore, it

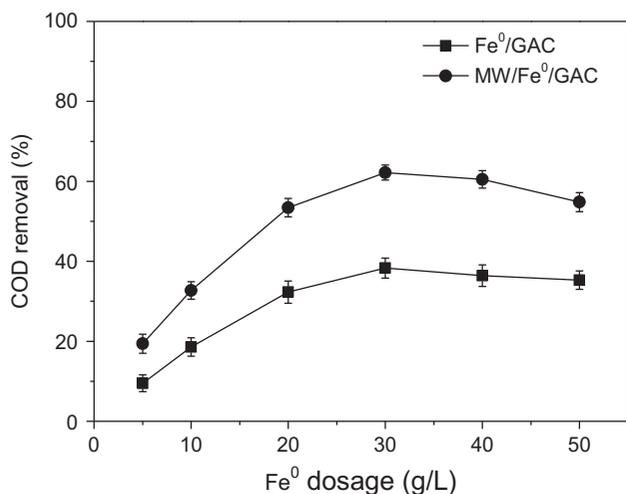


Fig. 4. Effect of iron dosage on the COD removal during IME process. Experimental conditions: PRW volume 100 mL; initial pH 3; MW power 500 W; Fe⁰/GAC 1:1 (v/v); reaction time 15 min.

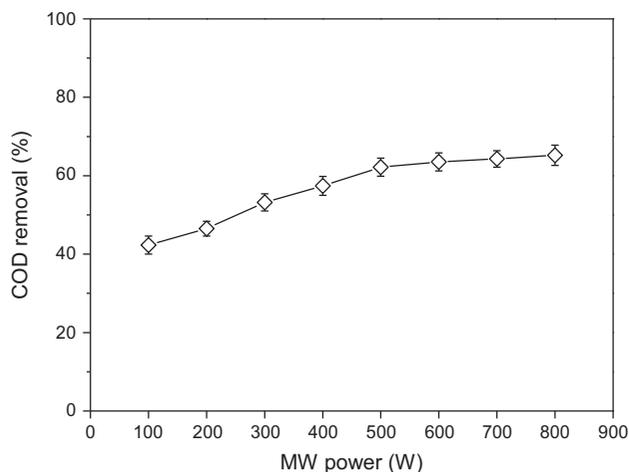


Fig. 5. Effect of MW power on the COD removal during MW-assisted IME process. Experimental conditions: PRW volume 100 mL; initial pH 3; Fe⁰ dosage 30 g/L; GAC dosage 5.75 g/L; reaction time 15 min.

is desirable to carry out the treatment with a 30 g/L dosage of iron filings to achieve maximum degradation efficiencies.

3.4. Effect of MW power

The influence of MW power on degradation efficiency of PRW was investigated. As shown in Fig. 5, COD removal increased with increasing the MW power up to 500 W, and a further increment in MW power resulted in little enhancement of treatment efficiency. In addition, we found that the volume of reaction solution expanded dramatically under MW power of 800 W because of strong exotherm, which was not favorable for the degradation reactions. Although GAC is a good MW absorbent, too high MW power could result in declining utilization efficiency because of the limited amount of GAC. From an economic efficiency viewpoint, a microwave power of 500 W was therefore applied afterwards.

3.5. Effect of reaction time

The effect of reaction time on the IME process was investigated when the initial pH was 3, the iron filings loading was 30 g/L, GAC dosage was 5.75 g/L, and MW power was 500 W, respectively. As can be seen from Fig. 6, the COD was quickly removed from the PRW during the first stage of treatment. After 15 min of treatment, the COD removal efficiencies were 38.3 and 62.2% for the Fe⁰/GAC and MW/Fe⁰/GAC processes, respectively. After 15 min, the increase in COD removal in the Fe⁰/GAC treatment became insignificant. This is

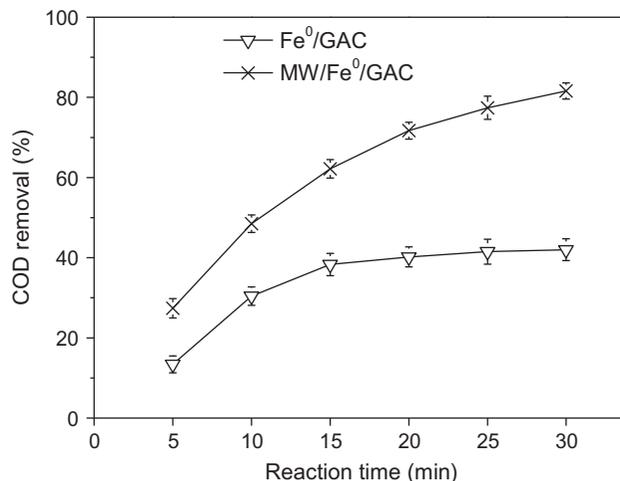


Fig. 6. Effect of reaction time on the COD removal during IME process. Experimental conditions: PRW volume 100 mL; initial pH 3; Fe⁰ dosage 30 g/L; GAC dosage 5.75 g/L; MW power 500 W.

Table 2

Comparison of MW/Fe⁰/GAC with other treatments. Experimental conditions: PRW volume 100 mL; initial pH 3; Fe⁰ dosage 30 g/L; GAC dosage 5.75 g/L; MW power 500 W; treatment time 15 min

Process	Removal efficiency (%)			
	COD	TPH	Total phenol	Oil and grease
MW/Fe ⁰ /GAC	62.2	78.4	92.2	71.6
Fe ⁰ /GAC	38.3	47.1	64.6	41.3
MW/Fe ⁰	8.6	10.5	ND	9.3
MW/GAC	35.8	36.2	55.3	32.4
Fe ⁰	7.2	9.3	ND	8.5
GAC	ND	ND	ND	ND
MW	ND	ND	ND	ND

ND: no significant loss ($p < 0.05$).

in agreement with the results of Yang [21] who treated polyester wastewater using Fe/C IME. As the IME reactions proceeded, more and more ferrous and ferric hydroxides were generated, leading to the precipitation of hydroxides on the Fe⁰ surface. This would block the electron transfer between iron and wastewater, thus resulting in deactivation of the iron surface and termination of IME [18].

For the MW-assisted Fe⁰/GAC process, COD removal efficiency increased continuously after 15 min of treatment, which was ascribed to the MW-induced decomposition effect. However, this growth tendency had slowed down compared with that of initial 15-min period. Extending MW radiation time could increase the removal efficiency of COD; however, long-time MW treatment may result in high consumption of electricity and treatment costs. Therefore, 15 min was chosen as the optimum radiation time.

Based on the above results, the optimum treatment condition of MW/Fe⁰/GAC system for the pretreatment of the PRW was: MW power 500 W, radiation time 15 min, iron filings dosage 30 g/L, GAC dosage 5.75 g/L, and initial pH 3. Under the optimum conditions, COD in the PRW decreased from the initial 1,680 mg/L to 635 mg/L, and BOD₅/COD was enhanced from 0.19 to 0.48. Thus, this pretreatment was beneficial for the following bioremediation process.

3.6. Comparison of treatment efficiency among various processes

With the optimum reaction conditions, the result of MW/Fe⁰/GAC system was compared with those of Fe⁰/GAC, MW/Fe⁰, MW/GAC, Fe⁰, GAC, and MW treatments. As shown in Table 2, the removal efficiencies of MW/Fe⁰/GAC system were significantly

higher than those of others. There was no obvious change of various indexes occurring in the single iron filings or GAC processes. MW alone did not cause significant volatilization losses of pollutants due to the utilization of water condenser. Iron filings resulted in small removal of pollutants by flocculation of ferrous hydroxide.

3.7. Degradation of *n*-alkanes

The *n*-alkanes were detected from C₈ to C₄₀ (Table 3). As shown, degradation efficiency of individual *n*-alkanes was obviously higher in the MW/Fe⁰/GAC process than in the Fe⁰/GAC process. This is consistent with the results of COD and TPH removal in both treatments. Additionally, it was observed that the removal efficiency of hydrocarbon components was higher than that of COD, indicating the presence of hydrocarbon intermediates during reactions.

Table 3

Degradation efficiency of individual hydrocarbons during the Fe⁰/GAC and MW/Fe⁰/GAC treatments. Experimental conditions: PRW volume 100 mL; initial pH 3; Fe⁰ dosage 30 g/L; GAC dosage 5.75 g/L; MW power 500 W; treatment time 15 min

Hydrocarbon	Degradation efficiency (%)		
	Fe ⁰ /GAC	MW/Fe ⁰ /GAC	
<i>n</i> -alkanes	C _{8–12}	48.6	76.3
	C _{13–16}	52.6	79.2
	C _{17–20}	49.3	75.4
	C _{21–24}	47.2	78.6
	C _{25–28}	45.1	73.2
	C _{29–32}	43.5	68.4
	C _{33–40}	43.9	65.3

4. Conclusions

The pretreatment of PRW by Fe⁰/GAC system in the absence and presence of MW irradiation was studied. The COD removal efficiencies using MWFe⁰/GAC were much higher in the presence of MW irradiation than those in the absence of MW. The highest COD removal efficiency was obtained when the Fe⁰/GAC ratio was set at 1:1 (*v/v*). The degradation performance was strongly pH-dependent, and the optimum pH value was found to be 3 for the pretreatment of PRW in the examined pH values. The biodegradability of the wastewater was markedly improved, revealing the great potential of MW/Fe⁰/GAC in engineering application as a pretreatment of PRW.

Abbreviations

PRW	—	petroleum refinery wastewater
COD	—	chemical oxygen demand
BOD ₅	—	five-day biochemical oxygen demand
TDS	—	total dissolved solids
TPHs	—	total petroleum hydrocarbons
PAHs	—	polycyclic aromatic hydrocarbons
GAC	—	granular activated carbon
IME	—	interior micro-electrolysis
MW	—	microwave
GC/MSD	—	gas chromatograph/mass selective detector

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