



Degradation of carbofuran in aqueous solution using persulfate/Fe²⁺/ultrasound treatment system

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ABSTRACT

Carbofuran is a methylcarbamate pesticide used in agricultural sectors and its overexposure in humans could cause coma, blurred vision and nausea. This study investigated the degradation of carbofuran in aqueous solution using persulfate/Fe²⁺/ultrasound process. To date, many researchers have been investigating the applicability of sulfate radicals in the degradation of organic contaminants, but this is the first-time sulfate radicals combined with US and Fe²⁺ (PS/Fe²⁺/US) were used to treat carbofuran in aqueous solution. Carbofuran removal was evaluated as a function of Fe²⁺ concentration, persulfate concentration, reaction time, and ultrasonic power. The result showed that 74.9 % of COD and 67.5% TOC removals were achieved within 13 min of reaction time at carbofuran concentration of 30 mg/L, Fe²⁺ concentration of 350 mg/L, and ultrasound power of 80 W. Additionally, the FTIR analysis of the treated and untreated samples revealed that the persulfate/Fe²⁺/US process successfully oxidized high-molecular-weight substances into small organic compounds. This study also showed that the Persulfate/Fe²⁺/US process had generated significantly less sludge as compared to the conventional Fenton process. In conclusion, the results confirmed the success of the persulfate/Fe²⁺/US advanced oxidation process and will probably open up a new remediation strategy to treat various recalcitrant organic pollutants.

Keywords: Sulfate radicals; Pesticides; Carbofuran; Ultrasonic oxidation; FTIR analysis

1. Introduction

The intensive use of pesticides to protect agricultural production from insects and invasive plants is necessary to minimize losses in productivity and to maintain the quality of the final product [1,2]. However, pesticides may reach the environment through various pathways including untreated wastewater, wastewater treatment plants, urban runoff and leaching from agricultural lands [3]. Their low biodegradability and high toxicity may possess adverse

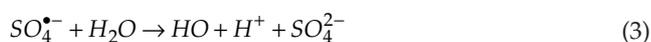
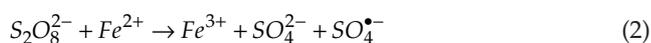
effects on public health such as mutation and hormone functions alteration [3–5]. Carbofuran is a methylcarbamate pesticide, mainly used to control rodents and beetles in paddy fields, sugarcane, soybean, vegetables and oil palm plantations [6]. However, the usage of carbofuran leads to the compound's residues in surface and groundwater sources, leading to difficulties in disposal [7]. Carbofuran has a high resistance against destruction and cannot be effectively degraded using conventional treatment methods [8].

Currently, advanced oxidation processes (AOPs) have been widely used to treat wastewater that are highly contaminated with toxic and refractory organic compounds [9–12]. AOPs such as Fenton [13], photo-Fenton [14], ozona-

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tion [15], photocatalytic [16] have been proven to be effective in reducing toxicity and ameliorating biodegradability of contaminants [7]. Most of the AOPs involve the generation of highly reactive hydroxyl radical ($\cdot\text{OH}$) [17,18]. Due to the AOPs' high efficiency, simplicity, biodegradability and treatment capability, Fenton process is highly advantageous as compared to other AOPs [18–20]. Fenton process is characterized by decomposition of hydrogen peroxide (H_2O_2), catalyzed by ferrous ion (Fe^{2+}) to produce reactive oxygen species, mainly hydroxyl radical [13]. Fenton process uses safe and cheap reagents and does not require a complex system to conduct the analyses [19]. However, there are also some significant limitations which hinder the Fenton process to be applied in the industrial wastewater treatment. One of the major problems in the application of Fenton process is the hydrogen peroxide that used for hydroxyl radical generation has a high selective reactivity to unsaturated compounds. This selective property makes the hydroxyl radical inefficient to completely mineralize the recalcitrant compounds [21]. Besides, Fenton process also requires safe disposal of solid iron containing sludge [22].

Recently, sulfate radical-based processes have attracted researchers to overcome the limitations of hydroxyl radical-based treatment systems. Sulfate radical is one of the strongest aqueous oxidizing species with a high redox potential of 2.5–3.1 V [23]. Sulfate radical is better than hydroxyl radical because it has longer half-life, faster kinetics, higher stability, and greater transport distances in the sub-surface level, the ability to work in a wide range of pH and can be activated by low-cost oxidant precursors [24]. Among others, persulphate oxidant ($\text{PS} = \text{S}_2\text{O}_8^{2-}$) is widely used as a sulfate-based oxidant and it demonstrates a great ability in degrading refractory compound by producing non-selective and high reactive sulfate radical ($\text{SO}_4^{\cdot-}$) [25]. The overall reactions involved in the production of sulfate radical by using Fe^{2+} and PS are outlined in Eqs. (1)–(3). As can be seen in Eq. (3), sulfate radical also induces the generation of hydroxyl radical by reacting with H_2O , which potentially increases the efficiency of the system by degrading the contaminants.



Besides, combining persulfate and Fe^{2+} with energy dissipating agents, such as ultrasound, ultraviolet radiation and ozone may possibly increase oxidation efficiency by decomposing water molecules (H_2O) into reactive free radicals such as HO and H^{\cdot} , resulting in the generation of high-frequency acoustic cavitations, as can be seen in Eqs. (4) and (5) [26].



To date, many researchers have been investigating the applicability of sulfate radical in the degradation of organic

contaminants, but this is the first-time sulfate radical, combined with US and Fe^{2+} ($\text{PS}/\text{Fe}^{2+}/\text{US}$) was used to treat carbofuran in aqueous solution. Performance of the treatment process was evaluated in terms of chemical oxygen demand (COD) and total organic carbon (TOC) removals. The $\text{PS}/\text{Fe}^{2+}/\text{US}$ treatment process was optimized for various process parameters, including Fe^{2+} concentration, persulphate concentration, US power and reaction time for COD and TOC removals. FTIR analysis was performed to identify degraded functional groups of compounds.

2. Material and methods

2.1. Materials and chemicals

Carbofuran ($\text{C}_{12}\text{H}_{15}\text{NO}_3$) was purchased from Merck (Malaysia) Sdn. Bhd. Potassium peroxydisulfate ($\text{K}_2\text{S}_2\text{O}_8$), ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), sodium hydroxide (NaOH) and sulphuric acid (H_2SO_4) were obtained from Sigma-Aldrich (Malaysia) Sdn. Bhd. The carbofuran solution was freshly prepared for each experiment by diluting the defined amount of carbofuran in the distilled water. All the chemicals were in the form of analytical purity and used without any further purification.

2.2. Experimental design and optimization

The statistical Design Expert Software (Version 8.0) was used to design the experiments. Response surface methodology (RSM)-central composite design (CCD), experimental model was used in this work. This model able to produce minimum experimental runs and assess the impacts of different parameters and its interaction with selected reaction responses [27]. The catalyst concentration (C_{Fe}), persulfate concentration (C_{PS}), ultrasonic power (US) and reaction time (R_t) were selected as independent variables while the COD and TOC removals were chosen as responding variables. In the present study, the range of selected parameters i.e. R_t (5–30 min), C_{PS} (30–450 mg/L), C_{Fe} (20–350 mg/L), and US (80–160 W) were determined from preliminary experiments.

2.3. Experimental procedure

The lab-scale experiments were performed in a 500 mL beaker. In each experiment, 400 mL volume of carbofuran in aqueous solution was subjected to $\text{PS}/\text{Fe}^{2+}/\text{US}$ process. A desired amount of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was then added and stirred at 310 rpm. The reaction was subsequently initiated by the addition of $\text{K}_2\text{S}_2\text{O}_8$. A sonicator equipped with titanium probe tip was used in this study. The ultrasound probe was immersed in the beaker containing wastewater and the power was adjusted to 80–160 W based on the RSM-CCD experimental design. The sample was quenched immediately by adding an excess amount of sodium nitrite to halt the oxidation after the reaction time. Then, the pH of the solution was measured and adjusted to pH 12 by adding sufficient amount of 1 M NaOH. At higher pH, Fe^{2+} will be precipitated to Fe^{3+} and inhibit further reaction of Fe^{2+} with peroxydisulfate. After settling time of 30 min, the samples were filtered using a 0.22 μm

syringe filter prior to analysis. All the experiments were repeated twice to obtain the average values. The process flow diagram is shown in Fig. 1.

2.4. Analytical methods

2.4.1. Chemical oxygen demand

Chemical oxygen demand (COD) was determined by using the fast digestion-spectrophotometric method. 2 mL of wastewater samples were added into test cells and heated at 148°C in a thermoreactor TR 420 Spectroquant for 120 min. The test cells were then cooled to room temperature and measured using a UV spectrophotometer. The COD removal is calculated using Eq. (6):

$$COD(\%) = \left(1 - \frac{COD_f}{COD_o}\right) \times 100 \quad (6)$$

2.4.2. Total organic carbon

20 mL of wastewater samples were added into the TOC cells and measured using a TOC analyzer (Shimadzu Corporation, Japan) based on the combustion/non-dispersive infrared gas analysis method. The TOC removal is calculated using Eq. (7).

$$TOC(\%) = \left(1 - \frac{TOC_f}{TOC_o}\right) \times 100 \quad (7)$$

2.4.3. Fourier transform infrared spectroscopy

For the purpose of characterization, Fourier transform infrared (FTIR) analysis was performed to study the functional groups of the treated and untreated wastewater sam-

ples. The spectrum was documented using a Perkin Elmer Spectrum One FTIR Spectrometer.

3. Results and discussion

3.1. Model analysis

Four operating parameters were chosen as independent variables to analyze the COD and TOC removals: catalyst concentration, persulfate concentration, ultrasonic power and reaction time of the treatment. The selected range of operating parameters and the level of independent variables are given in Table 1. The COD and TOC removals were chosen as response variables because they were able to provide necessary information for evaluating the analytical performances. A total of thirty experiments were performed based on the model suggested by RSM-CCD and the responses are given in Table 2.

In this study, the experimental data were fitted to a second-order polynomial equation as can be seen in Table 3. The positive and negative coefficient values showed synergistic and antagonistic outcomes for each term respectively. These equations were used to predict the percentages of COD and TOC removals with reasonably good accuracy. A positive value represented an increment of COD and TOC removals with the operating factors, while a negative value represented a reduction when the factor level increases.

Analysis of variance (ANOVA) was added to the analyses to confirm the statistical consistency (95% confidence level) of the developed quadratic model in this study. The fit of the model was evaluated by means of ANOVA, revealing the effects of the model that were statistically significant for a confidence level of 95% (p -value < 0.05). F test of ANOVA is presented in Table 4 to check the statistical significance of the quadratic model for a percentage of COD removal. The p -value represented the probability of F occurring due to

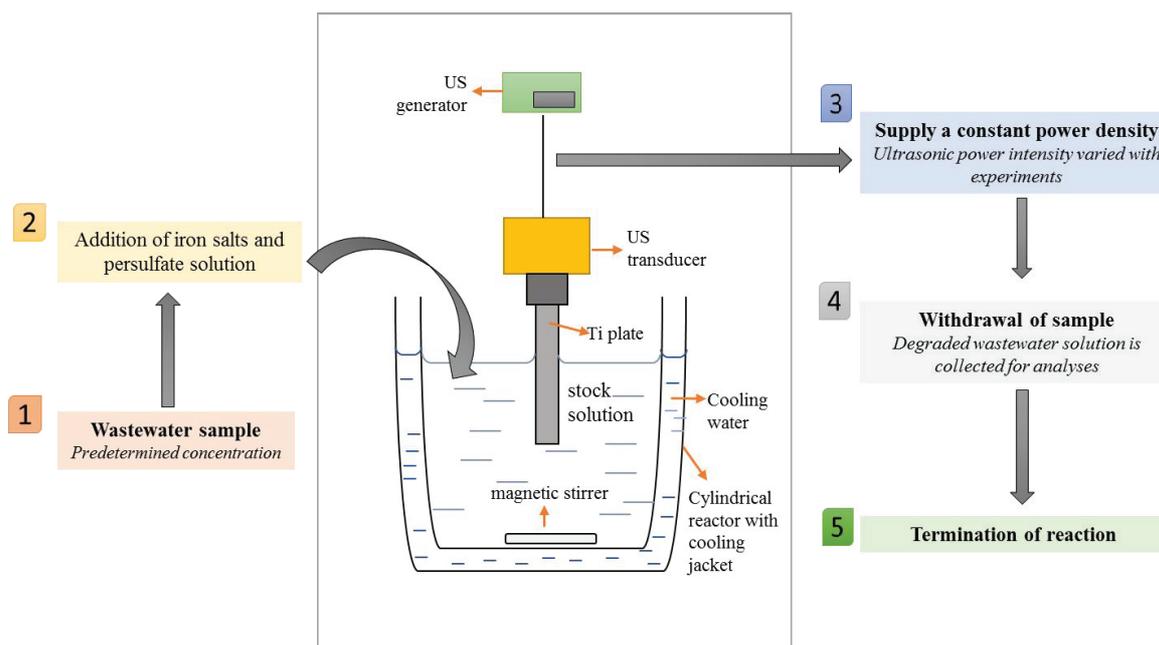


Fig. 1. Process flow diagram of persulfate/Fe²⁺/ultrasound process.

Table 1
Experimental design of pesticide containing wastewater for PS/Fe²⁺/US process

Independent variable	Units	Coded levels				
		-2	-1	0	+1	+2
Independent variable	Units	Actual levels				
Catalyst concentration (C _{Fe})	mg/l	145.0	20.0	185.0	350.0	515.0
Persulfate concentration (C _{PS})	mg/l	180.0	30.0	240.0	450.0	660.0
Ultrasonic power (US)	W	40.0	80.0	120.0	160.0	200.0
Reaction time (R _T)	min	7.5	5.0	17.5	30.0	42.5

Table 2
Experimental conditions and results for carbofuran removal in the PS/Fe²⁺/US process

Run	Independent variables				Responses (%)	
	C _{Fe}	C _{PS}	US	R _T	COD removal	TOC removal
1	350.0	450.0	160.0	5.0	33.0	20.0
2	185.0	240.0	120.0	42.5	28.0	45.0
3	20.0	450.0	160.0	30.0	56.0	24.0
4	350.0	30.0	160.0	30.0	20.0	32.0
5	-145.0	240.0	120.0	17.5	39.0	20.0
6	20.0	30.0	160.0	30.0	29.0	25.0
7	20.0	450.0	80.0	30.0	36.0	33.0
8	185.0	240.0	120.0	17.5	43.0	25.0
9	350.0	450.0	80.0	5.0	35.0	33.0
10	185.0	240.0	120.0	17.5	46.0	33.0
11	20.0	30.0	80.0	5.0	47.0	49.0
12	20.0	450.0	160.0	5.0	56.0	52.0
13	185.0	240.0	40.0	17.5	57.0	33.0
14	185.0	240.0	120.0	17.5	42.0	34.0
15	185.0	240.0	120.0	-7.5	42.0	40.0
16	185.0	-180.0	120.0	17.5	49.0	32.0
17	185.0	240.0	120.0	17.5	40.0	36.0
18	185.0	240.0	120.0	17.5	43.0	30.0
19	20.0	30.0	160.0	5.0	45.0	41.0
20	350.0	30.0	80.0	30.0	50.0	44.0
21	350.0	450.0	160.0	30.0	51.0	33.0
22	185.0	240.0	120.0	17.5	45.0	48.0
23	20.0	450.0	80.0	5.0	51.0	20.0
24	350.0	450.0	80.0	30.0	30.0	45.0
25	350.0	30.0	80.0	5.0	58.0	40.0
26	350.0	30.0	160.0	5.0	50.0	20.0
27	20.0	30.0	80.0	30.0	25.0	42.0
28	185.0	240.0	200.0	17.5	50.0	49.0
29	185.0	660.0	120.0	17.5	55.0	50.0
30	515.0	240.0	120.0	17.5	53.0	20.0

Table 3
Quadratic polynomial models derived for carbofuran removal in the PS/Fe²⁺/US process

Responses	Proposed quadratic model
COD %	77.60 + 0.07 (C _{Fe}) - 0.08 (C _{PS}) - 0.40 (US) - 0.56 (R _T) - (0.15×10 ⁻³) (C _{Fe} × C _{PS}) - (0.44×10 ⁻³) (C _{Fe} × US) + (0.84×10 ⁻³) (C _{Fe} × R _T) + (0.60×10 ⁻³) (C _{PS} × US) + (0.18×10 ⁻²) (C _{PS} × R _T) + (0.28×10 ⁻²) (US × R _T)
TOC %	73.75 + 0.04 (C _{Fe}) - 0.05 (C _{PS}) - 0.52 (US) - 0.59 (R _T) - (0.18×10 ⁻³) (C _{Fe} × C _{PS}) - (0.17×10 ⁻³) (C _{Fe} × US) + (0.14×10 ⁻²) (C _{Fe} × R _T) + (0.50×10 ⁻³) (C _{PS} × US) + (0.76×10 ⁻³) (C _{PS} × R _T) + (0.45×10 ⁻²) (US × R _T)

Table 4
ANOVA results for response surface quadratic model using PS/
Fe²⁺/US process for (a) COD and (b) TOC removals

Source	Sum of Squares	df	Mean Square	F-Value	p-value Prob > F
Model	2351.31	14	167.95	3.7481	0.0079
A (C _{Fe})	4.17	1	4.17	0.0930	0.7646
B (C _{PS})	54.00	1	54.00	1.2051	0.2896
C (US)	1.50	1	1.50	0.0335	0.8573
D (R _T)	511.47	1	511.47	11.4141	0.0041
AB	420.25	1	420.25	9.3785	0.0079
BC	400.00	1	400.00	8.9265	0.0092
BD	342.25	1	342.25	7.6378	0.0145

Source	Sum of Squares	df	Mean Square	F-Value	p-value Prob > F
Model	2150.17	14	153.58	2.9888	0.0218
A (C _{Fe})	32.67	1	32.67	0.6357	0.4377
B (C _{PS})	80.67	1	80.67	1.5698	0.2294
C (US)	0.17	1	0.17	0.0032	0.9553
D (R _T)	311.87	1	311.87	6.0690	0.0263
AB	650.25	1	650.25	12.6541	0.0029
BC	289.00	1	289.00	5.6241	0.0315

noise; where the smaller the value of p , the more significant is the corresponding parameter in the model. The values of 'Prob > F' were lesser than 0.0500 ($\alpha = 0.05$, 95% confidence level), indicating that the present quadratic models were statistically significant. The ANOVA analyses showed that both models for COD and TOC removals have P values less than 0.05. This confirmed the models were highly significant to describe the TOC and COD removal efficiencies of carbofuran. In this case, D, AB, BC, and BD were the significant model terms for COD removal; and D, AB, and BC were the significant model terms for TOC removal. In addition, "Adequate Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. Ratio values of 8.558 and 6.921 derived from COD and TOC removals indicated that there were adequate signals for all the response variables. This model can be used to navigate the design space.

3.2. Effect of parameters on COD and TOC removals

3.2.1. Effect of Fe²⁺ concentration

The ferrous ion (Fe²⁺) concentration is a critical parameter, which decides the treatment efficiency of the process [28]. The Fe²⁺ concentration was varied from 20 to 350 mg/L to study the effect of catalyst concentration on the COD and TOC removals. As can be observed in Fig. 2a, 38.0% of COD removal was achieved at Fe²⁺ concentration of 20 mg/L and persulfate concentration of 30 mg/L. COD removal increased to 50 % when the Fe²⁺ concentration increased to 350 mg/L at a constant persulfate concentration of 30 mg/L. This showed that higher Fe²⁺ concentration could improve the generation rate of sulfate radicals, which enhanced the

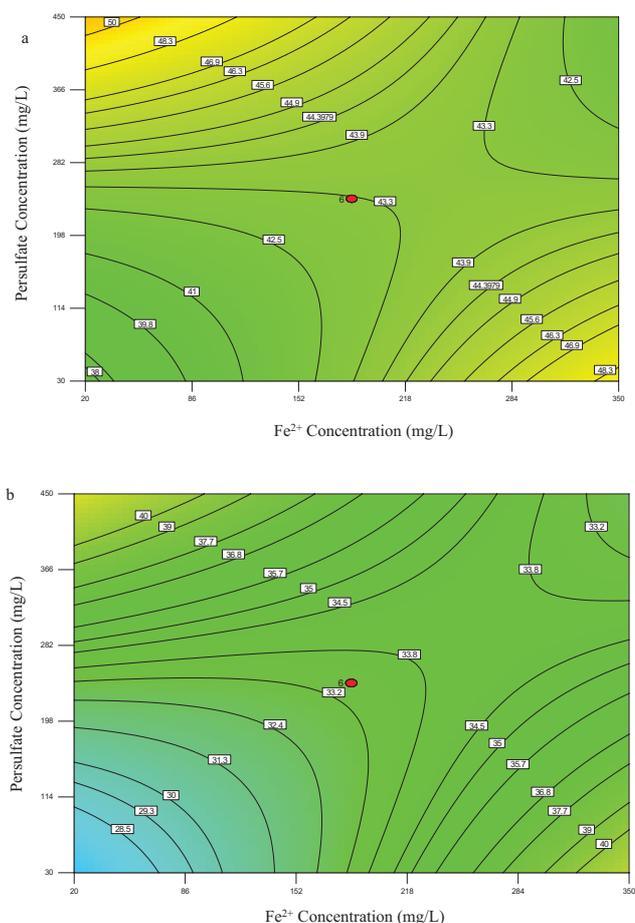
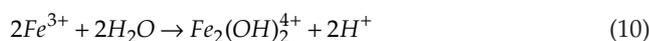


Fig. 2. 2D contour of interaction between Fe²⁺ concentration and persulfate concentration on a) COD and b) TOC removals (reaction time: 17.5 min and ultrasonic power: 120 W).

degradation rate of carbofuran. However, Fe²⁺ could also act as an effective scavenger of sulfate radicals at higher concentration. Fig. 2b shows that 30.0% of TOC removal was achieved at Fe²⁺ concentration of 20 mg/L and persulfate concentration of 30 mg/L. As Fe²⁺ concentration increased from 20 mg/L to 350 mg/L, the TOC removal increased as well. Maximum TOC removal of 40.0% was achieved when Fe²⁺ concentration was at 350 mg/L. However, when Fe²⁺ concentration is increased beyond 350 mg/L, the TOC removals were observed to be decreased to 35.0%. Ferrous ion of optimum concentration was highly favourable as the catalyst was able to improve the production rate of sulfate radical [29]. During the reaction, Fe²⁺ was used and oxidized to Fe³⁺, which will subsequently be transformed into ferric oxyhydroxides by hydrolysis process. The generation of ferric oxyhydroxides is shown in Eqs. (8), (9) and (10) [28]. Due to transient cavitation generated by the turbulent flow states of the system, the incorporation of ferrous ion was able to elevate the mass transport, which led to the increment in degradation efficiency. The ferrous ion also acted as nucleus for surface cavitation that led to the increase in cavitation intensity, where it further enhanced the degradation efficiency [30]. However, excessive concentration of ferrous ion behaved as an effectual scavenger of sulfate radical. The

excessive concentration led to the equilibrium between the production and depletion of sulfate radical. As the concentration increased, the performance of the process reduced due to the decrease in number of sulfate radical [30]. A similar result was reported by Wang et al. [31]. They found that the excessive amount of Fe^{2+} decreased the organic pollutant removal. Besides, excessive Fe^{2+} also produced much amount of Fe^{3+} , FeOH^{2+} , and $\text{Fe}(\text{OH})_2^+$, which possessed lower efficiency in initiating persulfate to generate sulfate radical compared to Fe^{2+} [28]. Therefore, it is important to optimize Fe^{2+} concentration to achieve the maximum COD and TOC removals.



3.2.2. Effect of persulfate concentration

Persulfate concentration is an initiator of the generation of sulfate radicals in PS/ Fe^{2+} /US process. Fig. 3a shows that at 45 min of reaction time, the maximum COD removal of 48.0% was achieved at the lowest persulfate concentration. As persulfate concentration increased to 450 mg/L, the COD removal decreased to 43.8%. Fig. 3b shows the increase in TOC removal when persulfate concentration increased with reaction time. At persulfate concentration of 450 mg/L, 34.5% of TOC removal was achieved. Further increment in reaction time, however, resulted in a decline in carbofuran removal. The TOC removal decreased to 24.6% when reaction time increased to 30 min. The increase in persulfate concentration caused the rise in the amount of sulfate radical and thus, enhancing the degradation rate. The excessive radical in the treatment may cause scavenging and decreased the degradation rate of the pesticide containing wastewater [29]. The scavenging effects are shown in Eqs. (11) and (12) [32]. The excessive sulfate radical and Fe^{2+} may contribute to the scavenging effects [27]. Rao and others observed a similar result in their study using Fe(II)-activated persulfate system for the degradation of carbamazepine [33]. They had reported that the excessive persulfate concentration could decrease the efficiency of the system.



3.2.3. Effect of ultrasonic power

Fig. 4a shows that the COD removal of 50.0% was achieved when the ultrasonic power increased from 80 W to 160 W at persulfate concentration of 30 mg/L. However, when the ultrasonic power increased above 160 W, the COD removal was observed to be decreased to 41.3%. Fig. 4b shows that the increase in ultrasonic power beyond 160 W decreased the TOC removal from

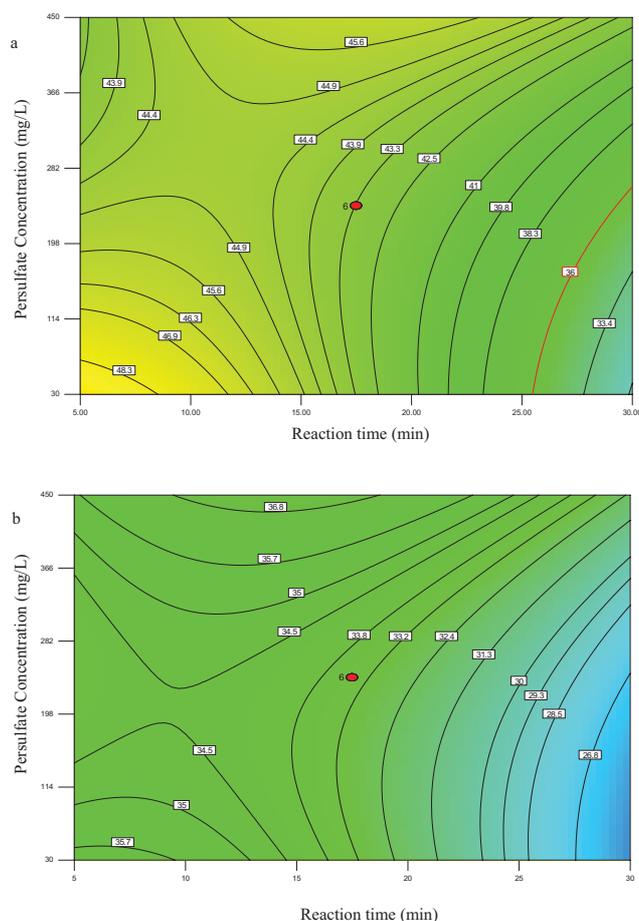


Fig. 3. 2D contour of interaction between persulfate concentration and reaction time on a) COD and TOC removals (Fe^{2+} concentration: 185mg/L and ultrasonic power: 120 W).

39% to 31.9%. The maximum TOC removal of 40.0% was achieved at ultrasonic power of 160 W. At higher ultrasound power, more sulfate radical will be produced, and the radical will increase the degradation efficiency. Additionally, higher power will generate turbulent flow in the system and eliminate the mass transfer resistances [23]. However, when the ultrasonic irradiation exceeded the limit, the excess bubbles will scatter the sound waves to the walls of the tank and back to the ultrasound probe. This may reduce the amount of sulfate radical in the treatment system [32].

3.2.4. Effect of reaction time

Fig. 5a shows a COD removal of 45.8% was achieved within 5 min at Fe^{2+} concentration of 20 mg/L. However, when reaction time increased to 30 min, the COD removal decreased to 36.6%. Fig. 5b shows that at Fe^{2+} concentration of 20 mg/L and reaction time of 5 min, the TOC removal of 35.7% was achieved. However, when the time increased to 30 min, the TOC removal decreased to 25%. A drop in the removal percentage might be caused by longer treatment time, that contributed to the formation scavenging, caused by excessive sulfate radicals and

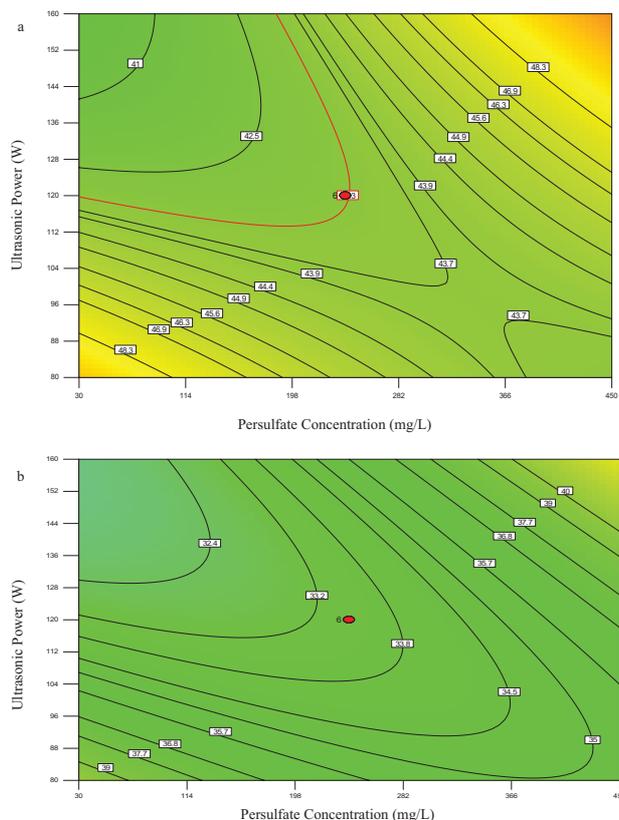


Fig. 4. 2D contour of interaction between persulfate concentration and ultrasonic power on a) COD and b) TOC removals (Fe^{2+} concentration: 185 mg/L and reaction time: 17.5 min).

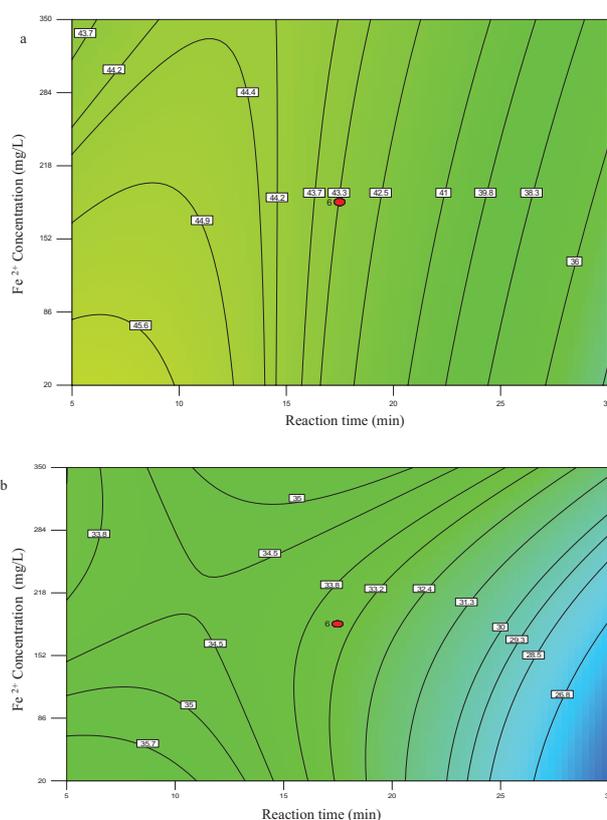


Fig. 5. 2D contour graph for a) COD removal and b) TOC removal as a function of catalyst concentration and reaction time (Ultrasonic power: 120 W, Persulfate concentration: 240 mg/l).

iron salts. The results indicated that there should be an optimized oxidation time.

3.3. Model validation and confirmation of optimized conditions

It is possible to decide specific working conditions such as maximizing the responses or keeping them in a desired range by using RSM, if multiple responses are applied. In the present study, the desired goal in terms of COD and TOC removals were defined as “maximize” to achieve the highest removal. In this study, the operating conditions such as Fe^{2+} concentration, persulfate concentration, ultrasonic power and reaction time were set to be ‘in the range’ without taking the factor of operating costs into consideration. The results are presented in Table 5. A verification experiment was carried out using the optimum conditions to confirm the adequacy of the predicted model. A good agreement between predicted value and experimental value confirmed the validity of the model for the PS/ Fe^{2+} /US process.

3.4. Surface group analysis

A comparison of the FTIR spectrum between treated and untreated carbofuran in aqueous solution are shown in Fig. 6. The PS/ Fe^{2+} /US process showed a major decrease

Table 5
Optimized condition for PS/ Fe^{2+} /US process ($C_{Carbofuran} = 50$ mg/L and $COD_i = 493$ mg/L)

C_{Fe} (mg/l)	C_{PS} (mg/l)	US (W)	R_T (min)	COD (%)		TOC (%)	
				Pred.	Exp.	Pred.	Exp.
350	30	80	13	71.1	74.9	65.2	67.5

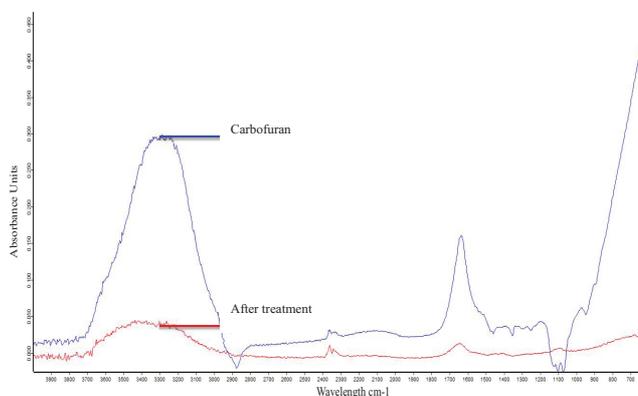


Fig. 6. FTIR spectrum of carbofuran in aqueous solution and after treated with PS/ Fe^{2+} /US process.

in the spectrum intensity at wavelengths of 3288 cm^{-1} and 1633 cm^{-1} . This observation can be associated with the degradation of hydroxyl groups and aromatic C=C bonds. In conclusion, the FTIR analysis showed that the PS/Fe²⁺/US process could oxidize high-molecular-weight substances into small organic compounds.

4. Conclusions

Based on the results obtained, it is proven that the PS/Fe²⁺/US process is an efficient method for treating carbofuran in aqueous solution. Under the optimum conditions; Fe²⁺ concentration of 350 mg/L, persulfate concentration of 30 mg/L, US power of 80 W and reaction time of 13 min, the PS/Fe²⁺/US process achieved 74.9% and 67.5% of COD and TOC removals respectively. Additionally, the FTIR analysis showed that the PS/Fe²⁺/US process could oxidize high-molecular-weight substances into small organic compounds. For future study, the by-products and their toxicity should be evaluated as it will be helpful for the degradation of carbofuran in real wastewater.

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