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Contribution of H_2O_2 in ultrasonic systems for degradation of DR–81 dye from aqueous solutions

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ABSTRACT

In this study, the degradation of Direct Red 81 (DR–81) dye in the aqueous phase by ultrasonic/H_2O_2 (US–H_2O_2 (system was investigated. The effect of operational condition such as pH, the concentration of H_2O_2, initial dye concentration, ultrasonic time, and temperature on the degradation of DR–81 was studied. Additionally, the effect of the addition of salts on the degradation of DR–81 was investigated. It was found that degradation of DR–81 was significantly pH–dependent. The degradation of DR–81 increased with the increase in the H_2O_2 concentration, ultrasonic time, and temperature. The degradation of DR–81 decreased with the increase in the concentration of DR–81. Addition of salts increased the degradation of DR–81. DR–81 degradation followed pseudo-first-order kinetics under controlled conditions. The maximal TOC removal efficiency was 57.3% with 0.225 mM of H_2O_2 and 0.045 mM of dye. Maximum %RSE obtained for US-H_2O_2 system was 2.8%. The results showed that US-H_2O_2 system sustain ably accelerated the degradation of DR–81 in aqueous solutions.

Keywords: Degradation; DR–81 dye; Ultrasonic/H_2O_2; Mineralization; Aqueous solutions

1. Introduction

Dyes are widely used in the various industries such as textile, plastics, paper, carpet, rubber, cosmetics, and food [1]. Color production is rapidly increasing in the world [2,3]. Textile and dyeing industries use a large amount of water and are among the most water-consuming industries. Water consumption in these sectors varies between 25 to 250 m^3 per ton of products [4,5]. Dyes applied in the textile industries could be classified into several groups including anionic dyes, cationic dyes, and non–ionic dyes [6]. About 15% of the production of the total dye in the world is lost during dyeing processes and then released into the environment [7]. Dyes have directly interfered with the photosynthesis process; therefore, the growth of organisms is reduced substantially. Also, dyes may form metallic bonds and produce highly toxic compounds to fish and other aquatic organisms. Discharge of industrial wastewater containing dyes into the aquatic environment significantly reduces the light transmittance and dissolved oxygen, and increases the chemical oxygen demand (COD), which severely disturbs the marine life [8].

Many different methods, such as coagulation, flocculation, biological treatment, chemical oxidation, electrochemical technology, ion exchange, and adsorption are used to decolorize the textile waste waters [9–14]. Furthermore, a combination of processes is also used for efficient dye degradation, such as ozonation with flocculation, electro coagulation and ion exchange and chemical method followed by biodegradation process [15]. Many researchers use the ultrasonic process to the degradation of different compounds such as trichloro-

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ethylene [16], sodium dodecylbenzene sulfonate [17], naphthol blue black [18], acid blue 25 [19], trichloroethylene, *o*–chlorophenol and 1,3–dichloro–2–propanol [20], 2chloro–5methyl phenol [21], reactive black 5 [22], Congo red [23], methyl orange [24], phenol and bisphenol A [25], methylene blue and sunset yellow [26,27], and carbamazepine [28,29].

Advanced oxidation processes (AOPs), such as ultrasonic and H_2O_2, either solely or in combination, are an efficient method that oxidizes the recalcitrant dyes to harmless end-products, such as H_2O and CO_2 [30,31]. The combination of ultrasonic with H_2O_2 seems to be a promising option to enhance the production of free radicals. The concentration of H_2O_2 plays an important role in deciding the extent of enhancement achieved for the combined process [19]. Ultrasonic irradiation of the aqueous solutions can result in the growth and collapse of gas bubbles,thus producing high transient temperatures and pressures, which lead to the formation of free radicals (·OH, ·OOH) via thermal dissociation of water and oxygen. H_2O_2 is formed as a consequence of ·OH and ·OOH radicals' recombination outside the cavitations' bubble [20,32,33].

There is a concern regarding water resources contamination by dyes and its adverse effects on people's health. Since the dyes are persistent and conventional treatment processes are not able to effectively remove the dyes, it is necessary to apply a suitable technique to remove the dyes from color manufacturing industries wastewater. Until now, no study has explored the degradation of DR–81 from aqueous solutions using US–H_2O_2 system. Only in a study by Harychandran et al. [34], the ultrasonic system in combination with fenton (sonofenton) was used to remove the DR–81. Therefore, the overall objective of this research was to determine the degradation of DR–81 by US–H_2O_2 system and investigate the effect of operating parameters on the degradation.

2. Materials and methods

2.1. Chemicals and analytical methods

The experiments were conducted at different concentrations of DR–81. The characteristics of DR–81 are presented in Table 1. DR–81 was obtained from Hoechst Company, Darmstadt, Germany. NaOH (analytical grade), H_2SO_4 (analytical grade), hydrogen peroxide (H_2O_2, 30%), methanol, and carbon tetrachloride were purchased from Merck CO (Darmstadt, Germany). The stock dye (1.48 mM) was supplied by dissolving 1 g of DR–81 in 1 L of distilled water. The desired dye concentration was prepared by the dilution of stock solution by distilled water. A UV/visible spectrophotometer (DR 5000 HACH, USA) was utilized to measure the absorbance of DR–81 at the maximum absorbance wavelength (508 nm) according to the standard method (No.2120) [35]. The pH values were determined using electrode method (Metrohm, Switzerland) [36]. An electrostatic balance (HR–200 laboratory, Precision Weighing Balances, Massachusetts, USA) was used to weigh the materials used in the experiments. The total organic carbon (TOC) of the samples was determined with a TOC analyzer (Multi N/C 3000, Analytikjena, Germany).

Table 1
The basic characteristics of DR-81 dye [37]

CAS number	2610-11-9
Molecular formula	$C_{29}H_{19}N_5Na_2O_8S_2$
Molecular weight	675.60 g/mol
Color index number	28 160
EC number	220-028-9
MDL number	MFCD00003949
UV absorption	λ_{max} (nm) = 397 λ_{max} (nm) = 508 (2nd)

2.2. Sono chemical Reactor

Irradiation of the solution in a 5.75 L ultrasonic reactor was conducted with a 37 kHz frequency and input power value of 550 W. Characteristics of the ultrasonic reactor was applied in the experiments presented in Table 2.

2.3. Operating conditions

This cross-sectional study was performed in a bench-scale test. The experiments were carried out in triplicates. All over the study, control samples were run for every experiment to validate the degradation and also check for loss of volatilization. To verify the mechanism of the degradation of DR–81, 100 mg/L of carbon tetrachlorideas hydrogen radical scavenger and 1%methanol as hydroxyl radical scavenger were added to the aqueous solution. The effects of operating parameters such as pH (2–10), the concentration of H_2O_2 (0.045–0.225 mM), initial dye concentration (0.045–0.177 mM), and contact time (0–120 min) on the degradation of DR–81 were studied. The adsorption of the dye was measured before and after the ultrasonic process using the UV/visible spectrophotometer. The dye degradation was calculated through Eq. (1):

Table 2
Characteristics of ultrasonic reactor applied in the experiments

Parameters	Characteristics
Model, manufacturing company, and country	Elmasonic S 60 H (220–240 V), Elma, Germany
Frequency	37 KHz
Power consumption total	550 W
Ultrasonic effective power	150 W
Flow type	Batch
Unit outer dimensions	L = 365 mm, W = 186 mm, H = 264 mm
Tank internal dimensions	L = 300 mm, W = 151 mm, H = 150 mm
Basket internal dimensions	L = 255 mm, W = 115 mm, H = 75 mm
Weight	5.1 kg
Max filling volume tank	5.75 L
Material tank	Stainless steel

$$R (\%) = [C_0 - C/C_0]\times 100, \quad (1)$$

where C_0 and C are in the initial and final dye concentrations after adsorption, respectively.

To prepare H_2O_2 we added 300.30 µL H_2O_2 to distilled water. The solution volume by adding to distilled water reached 100 mL. That way, asolution of 29.4 mM H_2O_2 was supplied. Finally, the concentrations of 0.045, 0.09, 0.135, 0.18, and 0.225 mM H_2O_2were prepared.

Therefore, a UV-visible spectrophotometer (LAMBDA 750 UV/Vis/NIR Spectrophotometer, PerkinElmer, USA) was used to determine the concentration of DR 81 dye at 508 nm λ_{max} [4,5]. Hydrogen peroxide residual concentration was determined by absorption spectrophotometric method [38].

2.4. Reaction stoichiometric efficiency (% RSE) determination

The RSE could be defined as the amount of degraded pollutants against the amount of H_2O_2 consumed at a specific time interval. To estimate the utilization efficiency of H_2O_2, RSE was used in previous studies [39,40], which is defined the ratio of the oxidized DR–81 concentration to the consumed H_2O_2molar concentration (Δ[DR–81]/Δ[H_2O_2]). The % RSE was calculated at the various molar ratios of DR–81 to H_2O_2 (1:1, 2:1, 3:1, 4:1, and 5:1).

3. Results and discussion

3.1. Effect of pH

The effect of pH on the degradation of DR–81 using US system is illustrated in Fig. 1. The experiments were performed with 0.074 mM DR–81 concentration at pH of 2, 3, 4, 5, 7, 9, and 10 for 45 min ultrasonic time. As seen in Fig. 1, for pH of 2, 3, 4, 5, 7, 9, and 10, the degradation of DR–81 after 45 min by ultrasonic system (US system) was 31.1%, 35.6%, 25.3%, 26.7%, 27.9%, 32.8%, and 34.9%, respectively. The maximum degradation of DR–81 (35.6%) occurred at pH = 3. It was found that degradation of DR–81 was significantly pH-dependent. The degradation of DR–81 in acidic solution (pH 2to 3) was higher, especially at pH 3, and decreased from pH 3 to 4; there was almost no change in the range of pH 4 to 7. High degradation was observed in basic conditions (pH 9 to 10). Results indicated that the highly acidic and basic media were suitable for degradation of DR–81. It is possible that the change in the pH of the solution results in the change of hydrophobic property of the DR–81, which affects the ultrasonic degradation [19]. The acceleration of degradation in strongly acidic conditions is probably related to the effect of protonation of negatively charged SO^3 group in the acidic medium [19]. The increase of degradation at basic medium may be caused by the change of hydrophobic property of the dye. The same results were obtained for the degradation of reactive brilliant red by 20 kHz ultrasonic irradiation [41]. In Ferkous et al.'s study [18] that investigated the sono chemical degradation of naphthol blue black in water, that a significant degradation occurred in acidic conditions (pH = 2). Harichandran and Prasad [34] found that the optimum conditions of decolorization DR–81 by the sono fenton process were pH = 3. He et al. [24] showed that the pH of the solution had a significant influence on the sono catalytic degradation of methyl orange. The results of Meshram et al.'s study [23] showed that the ultrasonic degradation in the acidic water (pH 2–3) was higher than those achieved in the neutral phase (pH 6–8), and a higher degradation was observed in basic condition (pH 10–12).

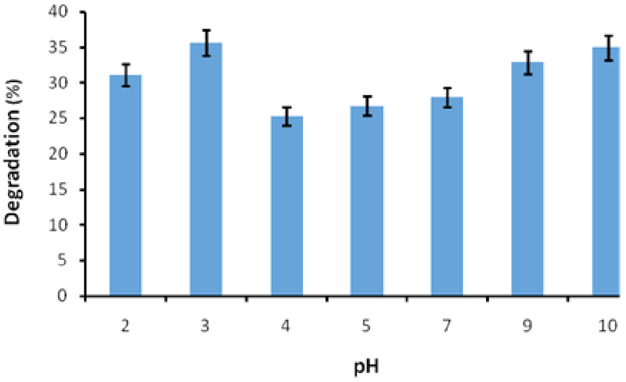

Fig. 1. Effect of pH on the degradation of DR–81 by US system (initial dye concentration: 0.074 mM; $T_{Ultrasonic}$: 45 min).

3.2. Effect of ultrasonic time

The effect of different ultrasonic times on the degradation of DR–81by US system is presented in Fig. 2. The experiments were conducted with 0.074 mM DR–81 concentration at pH 3 and at various times of 0, 15, 30, 45, 90, and 120 min. Based on the results illustrated in Fig. 2, degradation of DR–81 at reaction times of 0, 15, 30, 45, 90, and 120 min was 0%, 15.1%, 25.3%, 35.6%, 37.7%, and 38.2%, respectively. The maximum degradation of DR–81 by US system was 38.2% (t = 120 min). Reaction time is a significant factor for any chemical reaction. Results of our study showed that DR–81 degradation was directly proportional to the ultrasonic time. Nam Tran et al. [28] showed that the best performance for carbamazepine degradation (90.1% of degradation) was at a treatment time of 120 min. Also, He et al. [24] showed that ultrasonic irradiation

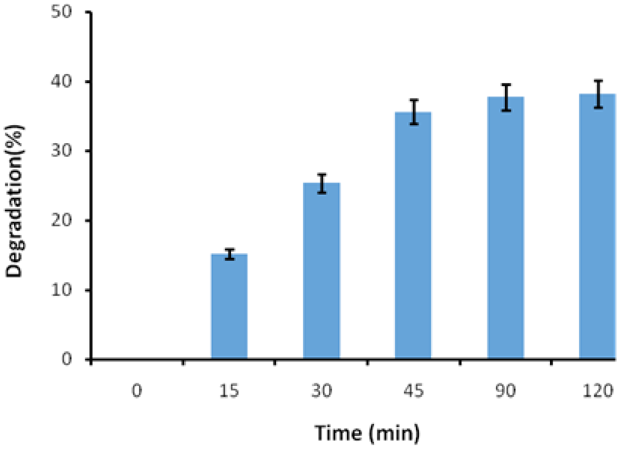

Fig. 2. Effect of ultrasonic time on the degradation of DR–81 by US system (initial dye concentration: 0.074 mM; pH: 3).

time had a significant influence on the sono catalytic degradation of methyl orange. Meanwhile, Meshram et al. [23] found that concentration Congo Red in aqueous solutions decreased with an increase in the sonication time.

3.3. Effect of H_2O_2 concentration

After the optimization of pH and ultra sonic time, the H_2O_2 optimum concentration was determined. The experiments were carried out with 0.074 mM concentration of DR–81, H_2O_2 different concentrations, pH of 3, and ultrasonic time of 120 min. The effect of H_2O_2 concentration on the degradation of DR–81 using US–H_2O_2 system is displayed in Fig. 3. Based on data presented in Fig. 3, the maximum degradation of DR–81 was observed at the H_2O_2 concentration of 0.225 mM. In addition, the degradation of DR–81 increased with the increase of concentration of H_2O_2. In Laxmi et al.'s study [21], the degradation of 2-chloro–5methyl phenol by ultrasonic irradiation coupled with H_2O_2 as an oxidant and TiO_2 as a catalyst was investigated. Their results indicated that an enhancement in the concentration of H_2O_2 from 10 to 250 mg/L led to an increase in degradation so that degradation enhanced from 4.1×10^{-3} to 6.8×10^{-3} min^{-1}. The maximum degradation rate was 6.8×10^{-3} min^{-1} which was observed at a peroxide concentration of 250 mg/L. However, addition of excess peroxide, i.e. 500 mg/L, did not increase the degradation rate. This may be due to the formation of peroxyl radicals. In Lim et al.'s study [25], the effects of H_2O_2 on the sono chemical degradation of phenol and bisphenol A were investigated. The results showed that the degradation of phenol and bisphenol A by addition of H_2O_2 enhanced from 10 to 50% and from 30 to 50%, respectively [25]. However, excessive addition of H_2O_2 may promote the $^{\circ}OOH$ formation and act as the hydroxyl radical's scavenger which reduces the oxidation efficiency [42].

3. 4. Effect of temperature

Degradation of DR–81 carried out under various temperatures ranged from 20 to 60°C for an initial dye concentration of 0.074 mM. Results of the effect of temperature on the degradation of DR–81 are presented in Fig. 4. With an increase in the temperature, the percentage of degradation was also increased. This could be explained by the hydrophilic property of the dye which is mostly degraded outside the cavitations process by the hydroxyl radicals produced by ultrasound. Thus, reactions in the bulk are facilitated by enhancing the temperature due to the higher mass transfer of various species at higher temperatures and this leads to an increase of the reaction rate of radicals with dye molecules. The increase of degradation with the enhancement in temperature is in agreement with the results of other studies [19,43].

3. 5. Effect of initial concentration of DR–81

Finally, the initial dye concentration was optimized. The effect of initial dye concentration on the degradation of DR–81 using US–H_2O_2 system is shown in Fig. 5. By fixing all the previous optimized parameters, the experiments were performed at ranges of initial DR–81 concentration 0.045, 0.059, 0.089, 0.117, and 0.177mM, pH of 3, the

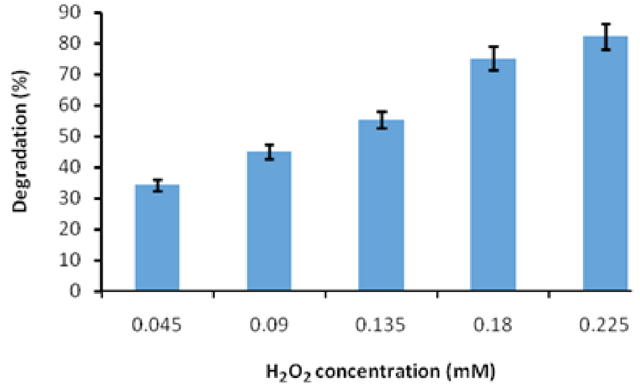

Fig. 3. Effect of H_2O_2 concentration on the degradation of DR–81 by US–H_2O_2 system (initial dye concentration: 0.074 mM; $T_{Ultrasonic}$: 120 min; pH: 3; H_2O_2 concentration: 0.045, 0.09, 0.135, 0.18, and 0.225 mM).

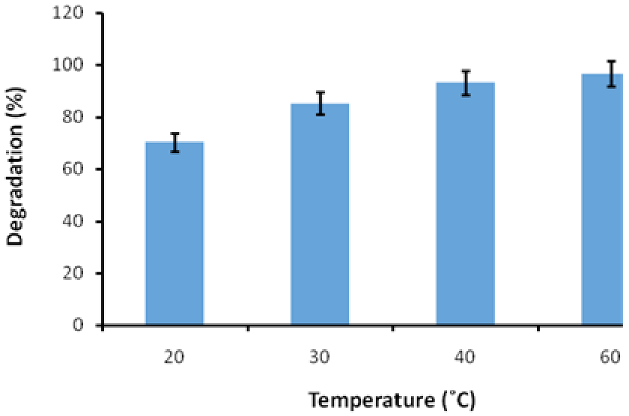

Fig. 4. Effect of temperature on the degradation of DR–81 by US–H_2O_2 system (pH: 3; $T_{Ultrasonic}$: 120 min; initial dye concentration: 0.074 mM; H_2O_2 concentration: 0.225 mM).

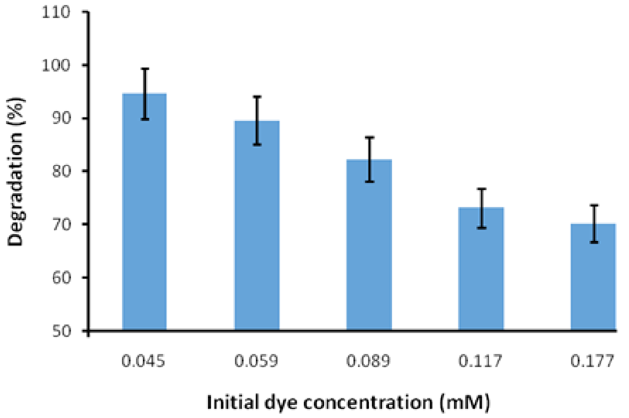

Fig. 5. Effect of initial dye concentration on the degradation of DR–81 by US–H_2O_2 system (pH: 3; $T_{Ultrasonic}$: 120 min; H_2O_2 concentration: O.225 mM; temperature: 60°C).

H_2O_2 concentration of 0.225 mM, and ultrasonic time of 45 min. Based on the results presented in Fig. 5, for DR–81 concentrations of 0.045, 0.059, 0.089, 0.117, and 0.177 mM, the degradation of DR–81 was 94.6%, 89.6%, 82.3%, 73.2% and 70.2%, respectively. The maximum degradation of DR–81 occurred at a concentration of 0.045 mM (94.6%). The degradation of DR–81 decreased with the increase of the concentration of DR–81, probably since a higher concentration enhances the number of dye molecules, not the HO· radical concentration, and the degradation decreases [44]. In Hasani et al.'s study [6], the maximum degradation of Acid Blue 292 was achieved at the concentration of 150 mg/L using poly–aluminum. In the study conducted by Song and Li [44], investigating the degradation of C.I. Direct Black 168 from the aqueous phase by fly ash/H_2O_2 combining ultrasound, the results indicated that the increase in the dye concentration from 100 to 500 mg/L decreased the degradation from 99% to 80%. In a study by Ghodbane and Hamdaoui [19], the effect of initial dye concentration varying from 10 to 150 mg/L on the degradation of acid blue 25 by high–frequency ultrasonic waves was studied. The researchers found that degradation of dye increased with the increase in the concentration. Also, they stated that this dye degradation increase might be attributed to the fact that with an increase in the solution concentration of the dye, the probability of OH radical attack on dye molecules would increase, thus leading to an increase in the degradation of dye.

3.6. Determination of the remaining DR–81 and H_2O_2 in solution

The remaining concentrations of DR–81 and H_2O_2 in the different time intervals are shown in Fig. 6. Based on the results, the remaining concentration of DR–81 in the solution reached from the initial concentration of 0.045 mM (t = 0) to the final concentration of 0.002 mM (t = 120 min).

Also, the remaining concentration of H_2O_2 in the solution reached from the initial concentration of 0.225 mM (t = 0) to the final concentration of 0.075 mM (t = 120 min).

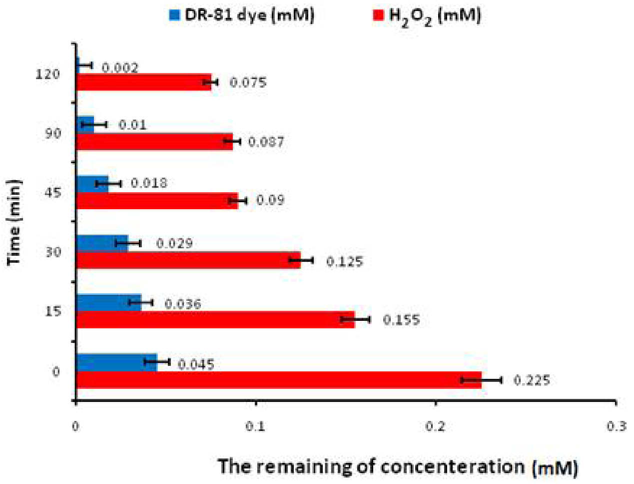

Fig. 6. The remaining concentration of DR–81 and H_2O_2 in the different time intervals by US–H_2O_2 system (pH: 3; $T_{Ultrasonic}$: 0–120 min; H_2O_2 concentration: 0.225 mM; dye concentration: 0.045 mM; temperature: 60°C).

3.7. Effect of salts

The effect of adding different salts on the degradation of DR–81 was examined for an initial dye concentration of 0.045 mM. The degradation of DR–81 in the absence and presence of 1 g/L of NH_4Cl, $NaHCO_3$, NaCl, $CaCl_2$, and $MgSO_4$ is presented in Fig. 7. Addition of salts enhanced the degradation of DR–81. Addition of salts to aqueous solution pushed AB25 molecules from the bulk aqueous phase to the bulk bubble interface [19,45,46]. The presence of salts may enhance the hydrophilicity, the surface tension and ionic strength of the aqueous phase and decrease the vapor pressure [19,45,46]. All these factors contribute to collapsing of the bubbles more violently, resulting in high degradation of DR–81.

3.8. Kinetic degradation

The degradation of DR–81 was carried out at two approaches as follows: (1) using the US, and (2) using US–H_2O_2. As illustrated in Fig. 8, after 120 min reaction in the

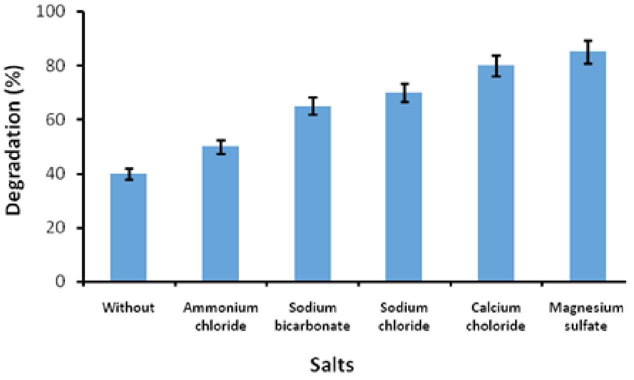

Fig. 7. Effect of salts on the degradation of DR–81 by US–H_2O_2 system (pH: 3; $T_{Ultrasonic}$: 120 min; initial dye concentration: 0.045 mM; H_2O_2 concentration: 0.225 mM; temperature: 60°C).

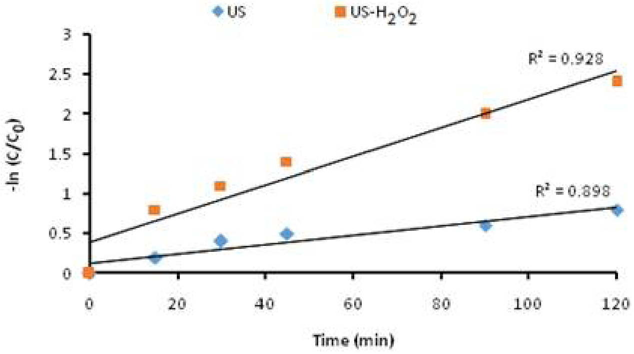

Fig. 8. The first order removal of DR–81 by US–H_2O_2 system (pH: 6.5; initial dye concentration: 0.045 mM; H_2O_2 concentration: 0.225 mM; temperature: 25±1°C; reaction time: 120 min).

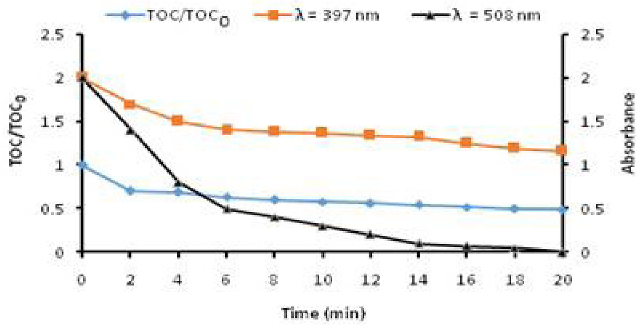

Fig. 9. Mineralization of DR–81 (pH: 6.5; initial dye concentration: 0.074 mM; H_2O_2 concentration: 2.35 mM; temperature: 25±1°C; reaction time: 20 min).

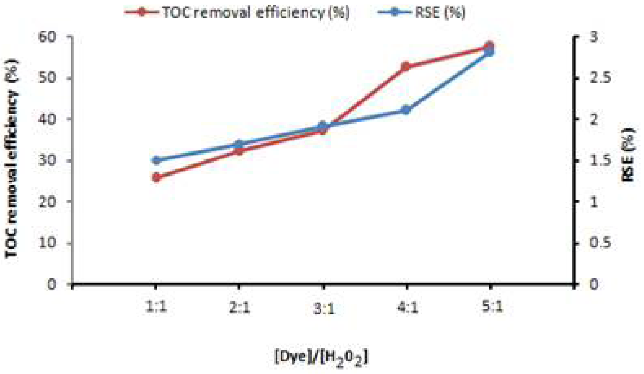

Fig. 10. TOC removal efficiency with various molar ratios of dye to H_2O_2 and % RSE for US–H_2O_2 system.

US–H_2O_2 system, the removal of DR–81 was 92.5%, while the removal efficiency of DR–81 by US system was 42.9%. The removal of DR–81 increased with the prolongation of the reaction time. The reaction was well followed by the first–order model. The correlation between ln C/C_0 and the reaction time was linear.

The kinetic expression can be presented as follows [47]:

$$\ln = C/C_0\,K_1t \tag{2}$$

where C is the DR–81 concentration at instant t, C_0 is the initial DR–81 concentration, k_1 is the first-order degradation rate constant, and t is the time of reaction.

The first-order rate constants for the degradation of DR–81 by US and US–H_2O_2 was 7.42 $\times10^{-2}$ min^{-1} (R^2 = 0.898) and 2.34$\times10^{-1}$ min^{-1} (R^2 = 0.928), respectively. The degradation efficiency of DR–81 by US–H_2O_2 system was higher than the individual effects of US system, which might be due to a synergistic effect.

3.9. Mineralization of DR–81

DR–81 has two absorbance peaks at 397 nm and 508 nm. The extent of mineralization of DR–81 can be evaluated by measuring the reduction of two absorbance peaks at 397 nm, 508 nm, and TOC removal. The reductions of two absorbance peaks were about 39.2% and 94.7%, respectively. The TOC removal of DR–81 about 46.5% was observed after 20 min for DR–81. A significant mineralization of DR–81 was observed in Fig. 9. Lin et al. [47] also reported that the TOC removal of AR14 dye about 42.3% was observed after 20 min for AR14.

Fig. 10 presents the TOC removal efficiency with different molar ratio of H_2O_2 (oxidant) to dye. According to Fig. 10, the increase of H_2O_2 increased the TOC removal efficiency. The maximal TOC removal efficiency was 57.3% with the molar ratio of 5:1, while the TOC removal efficiency decreased to 25.6% with the molar ratio of 1:1. The %RSE was calculated. The results, as seen in Fig. 10, indicated that TOC decreased with the increase of % RSE. Maximum %RSE obtained for US–H_2O_2 system was 2.8%. The % RSE in the iron-containing mineral/H_2O_2 systems varied over an order of magnitude, from 0.02% with amorphous Fe-oxide to 0.25% with FeAlSi-ox [48]. The majority of the RSE reported earlier for catalytic oxidation of organics by H_2O_2 with iron oxide fell in the range of 10^{-2}~10^{-4} (mol/mol) [49–51]. Also, Ghauch et al. [29] reported a clear opposite trend between TOC and %RSE. In Ghauch et al.'s study [39], based on the use of persulfate in UV–254 nm activated systems for complete degradation of chloramphenicol antibiotic in water, the %RSE reached up to 52%. In addition, in the study conducted by Naim and Ghauch [40], based on the use of activated persulfate systems for the degradation of ranitidine, the results showed a % RSE greater than 72% in most systems.

4. Conclusions

Based on the results of this study, degradation of DR–81 was strongly dependent on operating factors. The degradation efficiency increased with the increase in H_2O_2 concentration, ultrasonic time, and temperature, but decreased with the increase of the initial dye concentration. Addition of salts increased the degradation of DR–81. The degradation followed the first–order kinetics model. A significant mineralization of DR–81 was observed. Maximum %RSE obtained for US–H_2O_2 system was 2.8%. Our results showed that US–H_2O_2 system was an appropriate method with more than 90% efficiency for the degradation of DR–81. Our results also demonstrated the usability of the US–H_2O_2 system as a promising way in the degradation of DR–81 from aqueous solution. Therefore, using real water samples can be the next step to determine the feasibility of the US–H_2O_2 system as an alternative for the degradation of DR–81.

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