



Degradation of rhodamine B in aqueous solution by using swirling jet-induced cavitation combined with H₂O₂

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ABSTRACT

The degradation of rhodamine B in aqueous solution by using swirling jet-induced cavitation combined with H₂O₂ was investigated. It was found that there is an obvious synergetic effect between hydrodynamic cavitation and H₂O₂ for the degradation of rhodamine B. The effects of various operating parameters such as H₂O₂ dosage, medium pH, solution temperature, fluid pressure and the dye initial concentration on the degradation of rhodamine B have been studied. It was found that the removal of rhodamine B in aqueous solution was increased with increasing the addition of H₂O₂ and the fluid pressure, and the removal of rhodamine B was decreased with increasing the medium pH and dye initial concentration. It was also found that the degradation of rhodamine B is dependent on the solution temperature. The removal of rhodamine B increased with increase of temperature from 30 to 50 °C, but less degradation ratio is observed at 60 °C. The degradation kinetics of rhodamine B in aqueous solution using swirling jet-induced cavitation combined with H₂O₂ under various operational conditions was also investigated. It was found that the degradation of rhodamine B follows a pseudo-first-order kinetics.

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1. Introduction

Wastewaters from fabric and yarn dyeing are one of the major environmental pollutants. Dye components in such wastewaters can be a key source of environmental pollution. Their elimination from wastewater effluents is now the subject of considerable concern of environmental remediation and has attracted many researchers in recent years. Most textile dyes are photolytically stable and refractory towards chemical oxidation [1] and these characteristics render them resistant towards decomposition by conventional biochemical and physico-chemical methods. Generally, adsorption on activated carbon, coagulation by a chemical agent or reverse osmosis was applied to such effluents [2,3]. Nevertheless, they are non-destructive methods, and merely transfer contaminants from water to sludge.

Advanced oxidation processes (AOPs), characterized by production of •OH radicals under mild experimental conditions, have emerged as a promising technology for the decomposition of organic pollutants [4–7]. •OH radicals can attack organic pollutants via (a) addition to the aromatic ring or to double bonds, and (b)

electron or hydrogen abstraction [8]. Recently, ultrasonic cavitation has been attracting attention as a AOPs for the elimination of the hazardous chemical substances in water [9–13].

Cavitation is a term used to describe a process, which includes nucleation, growth and implosion of vapor or gas filled cavities. This process can have substantial chemical and physical effects [14]. Hydrodynamic cavitation is produced when large pressure differentials are generated within a moving liquid. Cavities are formed into a liquid when the pressure of the liquid is reduced below the vapor pressure of the liquid in current temperature. When cavities are carried to higher pressure region they implode violently and very high pressures and temperature can occur. The collapse of the cavitation bubbles also initiates some physico-chemical effects such as production of shear forces, shock waves whereas and the formation of reactive radicals. The radicals produced from hydrodynamic cavitation such as H• and HO• can react with other molecules in water to promote a variety of chemical reactions as similar to ultrasonic cavitation. However there are not many studies on the chemical effect and applications of hydrodynamic cavitation. Suslick et al. [15] have reported a conclusive experimental evidence for chemical reactions caused by hydrodynamic cavitation within a jet fluidizer in which I⁻ was oxidized to form I₃⁻. Kalumuck and Chahine [16] have studied the degradation of *p*-nitrophenol in recirculating flow loops using a variety of cavitating jet configurations and operating conditions. The result showed that jet-induced cavitation can achieve oxidation with up to two orders of magnitude greater energy efficiency

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compared to the ultrasonic means. Vichare et al. [17] have used KI decomposition as a model reaction and confirmed the occurrence as well as the intensity and thereby the efficiency of the multiple hole orifice plates in generating the cavitation. Sivakumar and Pandit [18] studied the degradation of a cationic dye rhodamine B by using multiple hole orifice plates and compared the efficiency of this technique with the cavitation generated by ultrasound. Gogate et al. [19] reported a cavitation efficiency assessment based on the oxidation reaction of potassium iodide. Recently, Chakinala et al. [20] reported a research on the treatment of industrial wastewater effluents by using hydrodynamic cavitation combined with advanced Fenton process. It was found that the novel combination of hydrodynamic cavitation with advanced Fenton process results in about 60–80% removal of TOC under optimized conditions depending on the type of industrial effluent samples. Chakinala et al. [21] also reported that the oxidation capacity of hydrodynamic cavitation can be enhanced with chloroalkanes addition. Arrojo and Benitond [22] reported a theoretical study of hydrodynamic cavitation. They found that the specific simulations of hydrodynamic bubbles reveal that time scales play a major role on the process.

The swirling jet-induced cavitation is generated by using a vortex flow. The swirling cavitation reactor setup has been described in detail elsewhere [23,24]. The flow enters a swirl chamber by means of tangential injection slots. The swirl produces a central line vortex that cavitates. As the feed pressure is increased, the filament along the jet axis attains a helicoidal shape with a vortex breakdown at the end of a rotating cork screw shape in some configurations. If the jet impacts on a nearby solid surface, the cavities so generated will collapse in the high pressure stagnation region of the jet. This jet has the additional advantage of generating a very large cavity volume and surface area. Up to now there are a few reports on the application of swirling jet-induced cavitation in the treatment of wastewater [23–25]. In our previously work [24], the chemical effect of swirling jet-induced cavitation and the energy efficiency of the process have been described, and it was found that the degradation of rhodamine B by swirling jet-induced cavitation was substantially enhanced with H_2O_2 addition. In the present work, the degradation of rhodamine B by using swirling jet-induced cavitation combined with H_2O_2 was further investigated in detail. The synergetic effect between hydrodynamic cavitation and H_2O_2 was discussed and the effects of operating conditions such as H_2O_2 dosage, medium pH, solution temperature, fluid pressure and the dye initial concentration on the degradation of rhodamine B was investigated.

2. Experiment

2.1. Regents

Rhodamine B (Analytical grade) was obtained from Shanghai Chemical Reagent Co. Ltd. (Shanghai, China). Hydrogen peroxide (30%, chemical reagent grade) was purchased from Tianjin Chemical Work and all other chemicals were of analytical reagent grade quality and were employed without further purification. Deionized and distilled water was used throughout.

2.2. Procedure

The pilot-scale experimental setup is a closed loop system designed to draw water from a holding tank of 40 L volume, then taking it into the swirling cavitation chamber and then discharging the treated solution back to a tank with a centrifugal pump (3000 rpm, 2500 W). In a typical experiment, 25 L aqueous solutions of rhodamine B with 10 mg L^{-1} initial concentration were subjected to swirling cavitation chamber with a length of 100 mm and diam-

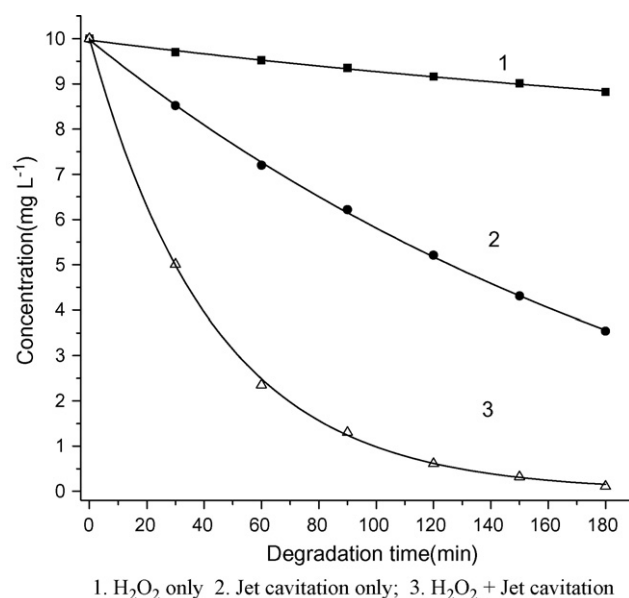


Fig. 1. Degradation vs. time dependency of rhodamine B. Operating conditions: temperature 50°C , pressure 0.6 MPa, pH 3.0, H_2O_2 100 mg L^{-1} .

eter of 10 mm (injection ports length 4.5 mm and width 0.2 mm) for degradation. The flow rate of water was fixed to $3.6 \text{ m}^3 \text{ h}^{-1}$ with 0.6 MPa inlet pressure in mainline. Prior to the degradation experiment, H_2O_2 was initially added to the desired target initial concentration. The pH of dye sonication was adjusted to 3.0 with diluted HCl. The pressure and the liquid flow in the swirling cavitation chamber can be controlled by adjusting valves. The temperature was controlled with the help of condenser in the water tank. After different experimental intervals, the remained dye was determined by measuring the absorbance at the maximum absorption wavelengths of rhodamine B (554 nm) on a UV-3000 UV-Vis spectrometer (Shimadzu, Japan).

3. Result and discussion

3.1. Synergetic effect between hydrodynamic cavitation and H_2O_2

The degradation of aqueous solutions of rhodamine B with concentration 10 mg L^{-1} was carried out at three approaches as follows: (1) using swirling jet-induced cavitation (fluid pressure 0.6 MPa), (2) using H_2O_2 (100 mg L^{-1}) and (3) using swirling jet-induced cavitation combined with H_2O_2 . Other experimental conditions involved the solution temperature 50°C and pH 3.0. The results were showed in Fig. 1. It can be seen from the results that there is an obvious synergetic effect between hydrodynamic cavitation and H_2O_2 for the degradation of rhodamine B.

As we know the cavitation in water can cause the formation of $\bullet\text{OH}$ radicals which are very strong and non-specific oxidizing species. Since $\bullet\text{OH}$ radicals are major free radical and important precursors for many products formed in degradation process, the production rate of $\bullet\text{OH}$ radicals strongly influence the oxidation efficiency of pollutants. It can be considered that the degradation of rhodamine B aqueous by swirling jet-induced cavitation is due to attack by hydroxyl radicals. It was reported that H_2O_2 can accelerate the photocatalytic degradation [26,27] and sonochemical degradation [28,29] of organic pollutants since H_2O_2 can be dissociated to form $\bullet\text{OH}$ radicals under such conditions. The result observed here suggest that the enhancement in the degradation of rhodamine B could be mainly due to contribution of additional $\bullet\text{OH}$ radicals produced from H_2O_2 .

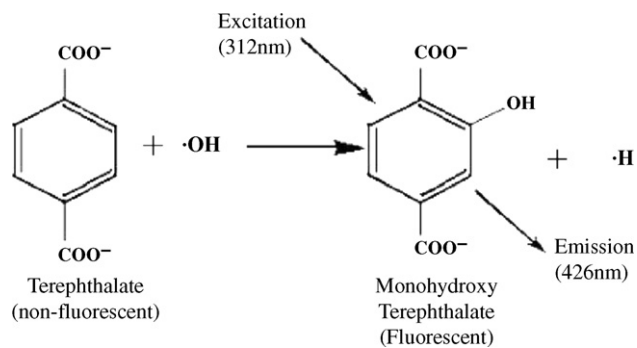


Fig. 2. The sketch of the formation of fluorescent molecule hydroxyterephthalate.

It is difficult to determine the yield of $\cdot\text{OH}$ radical because of its high reactivity, short half-life and very low concentration. Terephthalic acid dianion (TA) has proven to be an especially convenient $\cdot\text{OH}$ radical trap, which has been used to estimate relative amounts of $\cdot\text{OH}$ radicals produced by ultrasonic cavitation [30], gamma irradiation [31] and ultraviolet irradiation [32]. In the terephthalate analysis, TA reacts with $\cdot\text{OH}$ radicals to form hydroxyterephthalate (HTA) as shown in Fig. 2. This results in a fluorescent molecule HTA that is a stable and highly fluorometric isomer (excitation, $\lambda_{\text{Ex}} = 312 \text{ nm}$; emission, $\lambda_{\text{Em}} = 426 \text{ nm}$). Here we determined the relative amounts of $\cdot\text{OH}$ radicals produced by swirling jet-induced cavitation and swirling jet-induced cavitation combined with H_2O_2 respectively, by using TA as a fluorescent probe. The results were showed in Fig. 3. From the results it can be seen that TA with H_2O_2 is non-fluorescent (curve 1), but strong fluorescence spectrum was displayed when TA aqueous was treated with hydrodynamic cavitation (curve 2), and much stronger fluorescence spectrum was observed when hydrodynamic cavitation was used in combination with H_2O_2 (curve 3). This is the evidence of $\cdot\text{OH}$ radicals production in hydrodynamic cavitation, and the production of $\cdot\text{OH}$ radicals in hydrodynamic cavitation can be remarkably enhanced by H_2O_2 addition. This result suggests that the synergetic effect between hydrodynamic cavitation and H_2O_2 for the degradation of rhodamine B could be mainly due to the contribution of additional $\cdot\text{OH}$ radicals production.

3.2. Effect of the pH of medium

The pH of the medium is an important parameter for the degradation of chemical pollutants with hydrodynamic cavitation combined with H_2O_2 . The effect of the initial pH of the aqueous

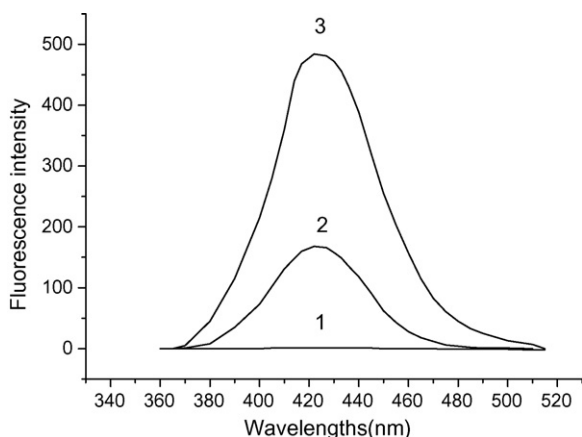


Fig. 3. The fluorescence spectrogram of hydroxyterephthalate. Operating conditions: temperature 50°C , pressure 0.6 MPa, H_2O_2 100 mg L^{-1} , pH 6.0.

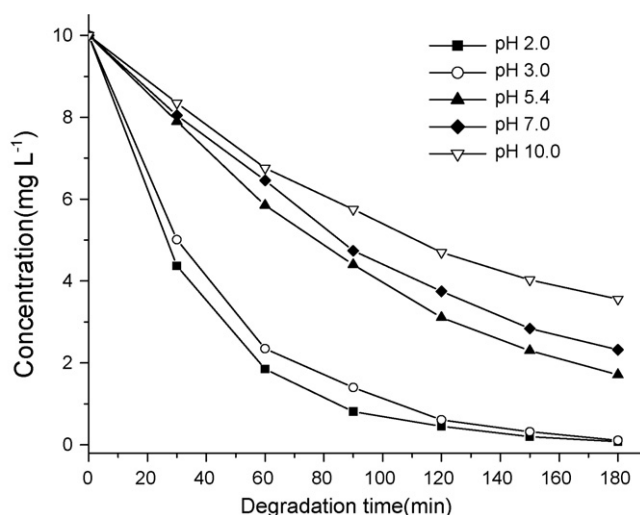


Fig. 4. Effect of medium pH on the degradation of rhodamine B. Operating conditions: temperature 50°C , pressure 0.6 MPa, H_2O_2 100 mg L^{-1} .

medium on the degradation of rhodamine B was investigated at pH range 2.0–10.0 using 10 mg L^{-1} rhodamine B aqueous solution at 50°C and the results were showed in Fig. 4. It was found that the degradation of rhodamine B is strongly dependent on medium pH. The removal of rhodamine B increased with the decrease of medium pH. When pH is 10.0, the residual concentration of rhodamine B is 3.56 mg L^{-1} and the removal ratio is only 64.4% after 180 min treatment. However, the residual concentration is only 0.08 and 0.11 mg L^{-1} at pH 2.0 and 3.0 respectively, and the removal ratio reaches 99.2% and 98.9%.

The reason may be that the oxidization potentials of $\cdot\text{OH}$ radicals and H_2O_2 is higher in acidic medium than that in basic. So the removal ratio of rhodamine B is increased with decreasing the medium pH. On the other hand, a high pH value may create more free radical scavengers (i.e., CO_3^{2-} , HCO_3^-) and results in the decrease in the concentration of $\cdot\text{OH}$ [33,34]. This may be another reason for the decrease of removal ratio of rhodamine B at higher pH value.

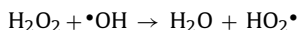
As a result, pH 3.0 was selected as the optimal operational pH value in the present research.

3.3. Effect of H_2O_2 concentration

The effect of the H_2O_2 concentration ranging from 10 to 150 mg L^{-1} on the degradation of rhodamine B was investigated using 10 mg L^{-1} rhodamine B aqueous solution at 50°C and pH 3.0. It was found that the degradation ratio of rhodamine B increased with increasing H_2O_2 concentration (Fig. 5).

The degradation ratios of rhodamine B were 61.2%, 74.5%, 90.4%, 98.9% and 99.1% with H_2O_2 initial concentration of 0, 10, 5, 100 and 150 mg L^{-1} , respectively. As mentioned above, the enhancement of H_2O_2 for the degradation of rhodamine B could be mainly due to contribution of additional $\cdot\text{OH}$ radicals produced from H_2O_2 dissociation. So more $\cdot\text{OH}$ radicals would be produced and higher degradation ratios were observed at higher H_2O_2 concentration. However, the enhancement did not increase when H_2O_2 addition was more than 100 mg L^{-1} .

As we know, H_2O_2 can also react with $\cdot\text{OH}$ radicals as following:



The rate of this reaction increase with H_2O_2 concentration and it can be competitive for $\cdot\text{OH}$ radicals. So high H_2O_2 concentration can constitute a limiting factor.

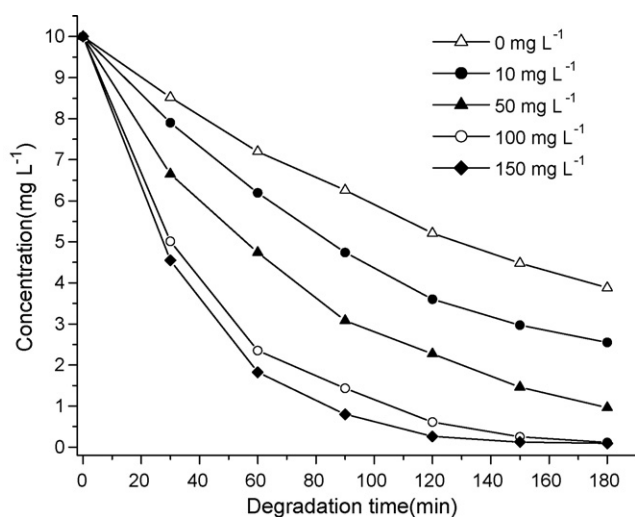


Fig. 5. Effect of H_2O_2 addition on the degradation of rhodamine B. Operating conditions: pH 3.0, temperature 50°C , pressure 0.6 MPa.

Since the degradation ratio of rhodamine B with 100 mg L^{-1} H_2O_2 addition is very close to that with 150 mg L^{-1} H_2O_2 , 100 mg L^{-1} was selected as the optimal operational H_2O_2 addition in the present research.

3.4. Effect of the solution temperature

The influence of temperature to the degradation of rhodamine B was investigated with 10 mg L^{-1} rhodamine B aqueous solution using hydrodynamic cavitation with 100 mg L^{-1} H_2O_2 addition at pressure 0.6 MPa and pH 3.0, and the result was showed in Fig. 6. It was found that the influence of temperature to the degradation of rhodamine B is complicated. The degradation ratio increased as temperature increased from 30 to 50°C , however, less degradation ratio is observed at temperature 60°C .

As we know, the forming of cavitation bubbles is related not only to gas dissolving in water, but also to water vapor. Higher temperature can increase the quantity of cavitation bubbles and result in increase of degradation. On the other hand, higher temperature could promote dissociation of H_2O_2 and more $\cdot\text{OH}$ radicals are produced. However, cavitation bubbles formed at higher temperature will fill with more water vapor. This water vapor could reduce the

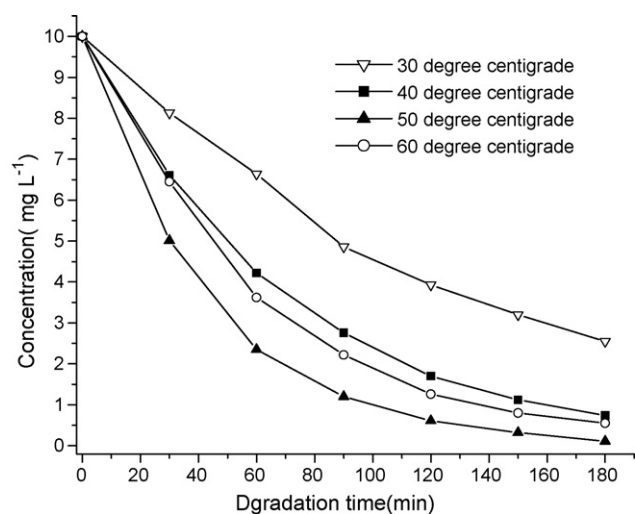


Fig. 6. Effect of temperature on the degradation of rhodamine B. Operating conditions: pH 3.0, pressure 0.6 MPa and H_2O_2 100 mg L^{-1} .

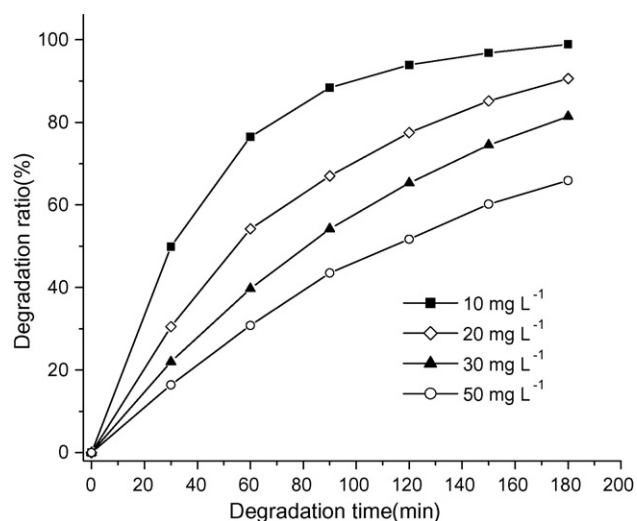


Fig. 7. Effect of initial concentration of rhodamine B on the dye degradation. Operating conditions: pH 3.0, temperature 50°C , pressure 0.6 MPa, H_2O_2 100 mg L^{-1} .

extremes of temperature and pressure generated when cavitation bubbles collapse and thus decrease the degradation efficiency of the organic compounds. As a result the highest degradation efficiency is obtained at temperature 50°C and this temperature was selected as the optimal operational temperature.

3.5. Effect of the dye initial concentration

The effect of dye initial concentrations on the degradation rate of rhodamine B was investigated by swirling jet-induced cavitation with 100 mg L^{-1} H_2O_2 addition at temperature 50°C and pH 3.0. The results were showed in Fig. 7. It was found from the results that with increasing the dye initial concentration, the degradation ratios of rhodamine B were decreased. After 180 min treatment, the degradation ratio was decreased to 63.4% from 98.9% when the initial concentration of rhodamine B increased to 50 mg L^{-1} from 10 mg L^{-1} . However, the total amount of the dye degraded was increased with the raising of the dye initial concentration.

This result is very similar to those studies on the degradation of rhodamine with hydrodynamic cavitation [25] and with sonication [35]. When the cavitation conditions and H_2O_2 addition are constant, the amount of $\cdot\text{OH}$ radicals in the system would be constant, but the total amount of dye in aqueous solution is increased with increasing the dye initial concentration. So the degradation ratio must be reduced. However, more $\cdot\text{OH}$ radicals could be caught with the dye when the dye initial concentration is increased and the total amount of the dye degraded was increased.

3.6. Effect of fluid pressure

The effects of fluid pressure on the degradation ratio of rhodamine B were also investigated. The experiment was conducted with 5.0 mg L^{-1} rhodamine B in aqueous solution at temperature 40°C and pH 5.4. The results were showed in Fig. 8. It was found that higher degradation rate of rhodamine B is observed at higher pressure. The result is very similar to those studies on the degradation of rhodamine with hydrodynamic cavitation [24]. This can be explained by the forming of cavitation. Higher pressure leads to higher injection speed to swirling cavitation chamber and then results in more strongly pressure drop in vortex center. So more cavitation bubbles can be formed and higher degradation ratio of rhodamine B was observed.

Table 1
The degradation rate constants and regression coefficients under various operational conditions.

Operation parameters					$k \times 10^{-2} (\text{min}^{-1})$	r
Concentration of rhodamine B (mg L^{-1})	pH	Addition of H_2O_2 (mg L^{-1})	Temperature ($^\circ\text{C}$)	Pressure (MPa)		
10	3.0	100	50	0.6	2.43	0.9974
20	3.0	100	50	0.6	1.27	0.9990
30	3.0	100	50	0.6	0.839	0.9985
50	3.0	100	50	0.6	0.566	0.9975
10	2.0	100	50	0.6	2.63	0.9988
10	5.4	100	50	0.6	1.00	0.9991
10	7.0	100	50	0.6	0.835	0.9980
10	10.0	100	50	0.6	0.585	0.9976
10	3.0	0	50	0.6	0.530	0.9991
10	3.0	10	50	0.6	0.785	0.9970
10	3.0	50	50	0.6	1.25	0.9992
10	3.0	150	50	0.6	2.78	0.9938
10	3.0	100	30	0.6	0.774	0.9986
10	3.0	100	40	0.6	1.46	0.9990
10	3.0	100	60	0.6	1.65	0.9983
10	3.0	100	50	0.2	0.545	0.9962
10	3.0	100	50	0.4	0.892	0.9973

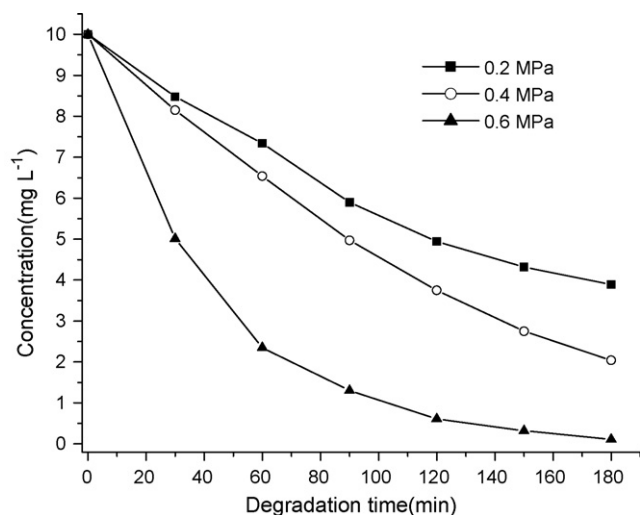


Fig. 8. Effect of fluid pressure on the degradation of rhodamine B. Operating conditions: pH 3.0, temperature 50°C , H_2O_2 100 mg L^{-1} .

3.7. Degradation kinetics

The degradation kinetics of rhodamine B in aqueous solution by using swirling jet-induced cavitation combined with H_2O_2 under various operational conditions were investigated. It was found that the concentration of rhodamine B in aqueous solutions decreased exponentially with reaction time in all experiments in present research, and the degradation rate can be expressed by the following equation:

$$C = C_0 \exp(-kt) \quad \text{or} \quad \ln \frac{C}{C_0} = -kt$$

where C is the concentration of rhodamine B at time t and C_0 is initial concentration, k is degradation rate constant, and t is degradation time. This result indicates that the degradation of rhodamine B follows a pseudo-first-order kinetics. The degradation rate constants (k) and regression coefficients (r) under various operational conditions are listed in Table 1.

4. Conclusion

The degradation of rhodamine B in aqueous solution by swirling jet-induced cavitation combined with H_2O_2 was investigated. It was

found that there is an obvious synergetic effect between hydrodynamic cavitation and H_2O_2 for the degradation of rhodamine B. The relative amounts of $\bullet\text{OH}$ radicals produced in swirling jet-induced cavitation and hydrodynamic cavitation combined with H_2O_2 were detected by using TA as a fluorescent probe, and the results showed that the production of $\bullet\text{OH}$ radicals in hydrodynamic cavitation can be remarkably enhanced by H_2O_2 addition. This result suggests that the synergetic effect between hydrodynamic cavitation and H_2O_2 for the degradation of rhodamine B could be mainly due to the contribution of additional $\bullet\text{OH}$ radicals production.

The effects of various operating parameters such as H_2O_2 dosage, medium pH, solution temperature, fluid pressure and the dye initial concentration on the degradation of rhodamine B have been studied. It was found that more addition of H_2O_2 , lower medium pH, higher fluid pressures and lower dye initial concentration are more favorable for the degradation of rhodamine B. For temperature, increase of temperature from 30 to 50°C has advantage to degradation of rhodamine B, but less degradation rate is observed at 60°C . The degradation kinetics of rhodamine B in aqueous solution using swirling jet-induced cavitation combined with H_2O_2 under various operational conditions were also investigated. It was found that the concentration of rhodamine B in aqueous solutions decreased exponentially with reaction time in all experiments. This result indicates that the degradation of rhodamine B by hydrodynamic cavitation combined with H_2O_2 follows a pseudo-first-order kinetics.

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