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Sonochemical degradation of basic fuchsin in water

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ABSTRACT

Basic fuchsin (BF) is an important dye used in textile and in biological and chemical assays, suspected of being carcinogenic. The environmental and occupational issues related to it are very important. This study aimed at investigating the sonochemical degradation of BF in water using high-frequency ultrasound waves (600 kHz). The effects of several operating parameters, namely initial substrate concentration, ultrasonic power, and liquid temperature (25–45°C), on the efficiency of ultrasonic process were investigated. Additionally, experiments were conducted in the presence of salts (NaCl and Na_2SO_4) as well as tert-butyl alcohol as a radical scavenger. The viability of ultrasound to degrade BF in natural water was also examined. The obtained results showed that ultrasound (600 kHz) is an efficient technique for removing BF. It was found that the initial degradation rate of BF increased with increasing initial dye concentration in the range 1–15 mg L^{-1} and liquid temperature in the interval 25–35°C. The initial degradation rate increased by factor of 2.8 when the delivered power increased from 30 to 90 W. Excepting the salting out effect, the presence of salts, even at high concentration, has practically no significant effect on the efficiency of BF removal, making ultrasonic treatment as a promising technique for removing organic pollutants from industrial and natural waters. The presence of tert-butyl alcohol in a mass ratio of 1:1 (BF:alcohol) drastically inhibited the degradation rate of BF, making sure that hydroxyl radical was the main species involved in the degradation of this compound.

Keywords: Basic fuchsin (BF); Sonochemical degradation; Ultrasonic waves; Hydroxyl radicals (·OH); Natural matrix

1. Introduction

One of the main sources with severe pollution problems worldwide is the textile industry and its dye-containing wastewaters (i.e. 10,000 different textile dyes with an estimated annual production of 7×10^5 tons are commercially available worldwide; 30% of these dyes are used in excess of 1,000 tons per year, and 90% of the textile products are used at the level of 100 tons per year or less) [1]. 10–25% of textile dyes are lost during the dyeing process, and 2–20% are directly discharged as aqueous effluents in different environmental components [1]. The discharge of very small amounts of dyes (less than 1 ppm for some

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dyes) is esthetically displeasing, impedes light penetration, affects gas solubility damaging the quality of the receiving streams and may be toxic to treatment processes, to chain organisms, and to aquatic life [2]. The majority of these compounds consumed at industrial scale are azo derivatives, although anthraquinone, indigoid, tryphenylmethane, xanthenes, sulfur or phthalocyanine derivatives are frequently utilized [2]. Triphenylmethane dyes are aromatic xenobiotic compounds that are used extensively in many industrial processes, such as textile dyeing, paper printing, and food and cosmetic manufacture [3]. They are known to be highly toxic to mammalian cells and mutagenic and carcinogenic to humans [4–6]. Based on their potential risk for human health and environment, the removal of these dyes from water is necessary to offer a cleaner environment as a requirement for human health and has attracted the most wanted attention of environmentalists, technologists, and entrepreneurs.

A variety of physical, chemical, and biological techniques are presently available for the treatment of wastewater discharged from various industries. Among them, much attention has recently been focused on the so-called advanced oxidation processes (AOPs). In these processes, various techniques (e.g. photolysis, photocatalysis, Fenton reaction, UV/H_2O_2) are applied to produce reactive species, principally hydroxyl radicals ($^{\cdot}OH$), which are able to induce the degradation and mineralization of water-dissolved organic pollutants [7]. A new way of generating $^{\cdot}OH$ radials is the application of ultrasound in the frequency range of 20 kHz–1 MHz, in which important chemical effects can be observed [8,9]. Application of ultrasound to aqueous solutions induces the formation of vapor- and gas-filled microbubbles that grow, undergo a series compression–expansion cycles, and then adiabatically collapse causing temperatures of about 5,000 K and pressures in excess of 1,000 atm therein [10,11]. These extreme conditions induce, inside the bubbles, the pyrolysis of water vapor, oxygen molecules, and volatile organic substrates that can be present in the gas phase [9]. Pyrolysis of water vapor yields hydroxyl radicals ($^{\cdot}OH$) and hydrogen atoms ($H^{\cdot}$) and with other gasses present in the bubble, other active species such as O atoms and $HO_2^{\cdot}$ radicals may create from H_2O and O_2 dissociation and their associate reactions [12,13]. Hydroxyl radicals in particular are very reactive and can transform organic compounds [14,15]. Reactions with free radicals can take place in the gas phase, at the gas–liquid interface and in the bulk solution after transfer of gaseous content into the liquid phase [16]. Accordingly, the sonochemical degradation of an organic compound can occur in gas-phase pyrolysis and oxidation for volatile substrates and by reaction with $^{\cdot}OH$ radicals at the gas–liquid interface and in the aqueous phase for nonvolatile substrates [17]. In the absence of any solutes in the liquid medium, these primary active species of sonolysis mostly recombine at the bubble–solution interface (reactions (1) and (2)) to form hydrogen peroxide (H_2O_2) that is released in the medium [18]:

$$2^{\cdot}OH \rightarrow H_2O_2 \quad k_1 = 5.5 \times 10^9\ M^{-1}\ s^{-1} \tag{1}$$

$$2^{\cdot}OOH \rightarrow H_2O_2 + O_2 \quad k_2 = 8.3 \times 10^5\ M^{-1}\ s^{-1} \tag{2}$$

Due to their high reactivity and their short lifetime, the total number of primary active species produced by the acoustic bubbles cannot be directly measured. It is generally accepted that the yield of H_2O_2 can be considered as an indicator for quantifying the overall chemical yield of ultrasound in aqueous medium [19,20].

In the present work, the dye under consideration is basic fuchsin (BF), which is a cationic dye of the tryphenylmethane class. In addition to its use in textile, BF is also used in the analysis of SO_2 and formaldehyde present in the air and water [21], as a pH indicator and also as biological stain [22]. In biological assays, BF has been used for staining of bacteria, antibody, or other organisms [22]. BF is toxic [23] and has been shown to cause cancer in animals and mutagenic effects in laboratory studies [24]. Its occupational and environmental issues are therefore of concern and especially the treatment and final disposition of BF wastes. Kosanic and Trickovic [25] present the process of photo-assisted degradation of BF in aqueous solutions through treatment with visible light. The authors report that illumination of the BF solution in the presence of oxygen or argon leads to the decrease in dye absorption. Martins et al. [23] showed that ozonation is an efficient technique for degrading BF. However, the same authors affirmed the non-effectiveness of ultrasound at 20 kHz toward the degradation of BF.

This study aimed at demonstrating the effectiveness of ultrasonic action at 600 kHz for the treatment of water contaminated with BF. The effects of several parameters such as initial dye concentration, ultrasonic power and liquid temperature on the sonochemical elimination of BF were evaluated. Additionally, the potential of ultrasonic waves to degrade BF in the presence of salts and mineral and organic scavengers was investigated.

2. Materials and methods

2.1. Reagents

Basic fuchsin (abbreviation: BF; CAS number: 569-61-9; molecular formula: $C_{20}H_{20}ClN_3$, molecular weight: 337.84 g mol^{-1}) was supplied by Sigma-Aldrich (≥98%) and used without any purification. The molecular structure of BF was shown in Fig. 1.

Potassium iodide (Riedel-de Haën), sodium sulfate (Acros Organics), sodium chloride (Sigma-Aldrich), sodium hydroxide (Sigma-Aldrich), sulfuric acid (Sigma-Aldrich), Mohr's salt $Fe(NH_4)_2(SO_4)_2{\cdot}6H_2O$ (Panreac), and tert-butyl alcohol (Prolabo) were used as received (analytical grade).

2.2. Ultrasonic reactor

Sonolysis experiments were conducted in cylindrical water-jacketed glass reactor (Fig. 2). The ultrasonic waves of 600 kHz were emitted from the bottom of the reactor through a piezoelectric disc fixed on stainless steel plate. The emitting system was connected to a high-frequency generator operating at variable supplied electric power. The temperature of the solution was monitored using a thermocouple immersed in the reacting medium. Acoustic energy dissipated in the solution was estimated using a standard calorimetric method [26]. The range of supplied electrical power used in this study was from 30 to 90 W. The estimated acoustic powers are 10.5, 13.5, 21.5, and 28 W for, respectively, 30, 50, 70, and 90 W of electrical powers.

2.3. Procedures

All BF solutions were prepared with distilled water. Experiments of BF degradation were carried out under different conditions using constant solution volume of 300 mL. The pH of the solution was adjusted using NaOH or H_2SO_4. Aqueous samples were taken periodically from the solution and the concentrations of the dye were determined using a UV–vis spectrophotometer (Lightwave II) at 543 nm. The temperature of the sonicated solution was kept constant by circulating cooling water through a jacket surrounding the cell. In the tests to investigate the effects of dissolved gasses, a gas cylinder was used for bubbling air or N_2 at least 15 min prior to start and until completion of experiments. In all the other experiments, the reactor was open to the air without any saturation.

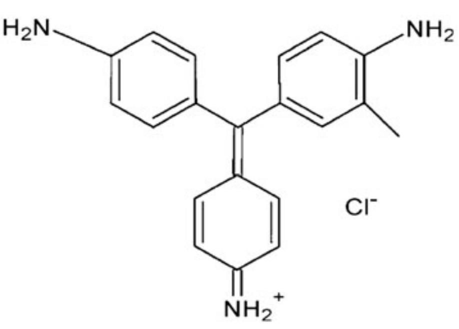

Fig. 1. Molecular structure of BF.

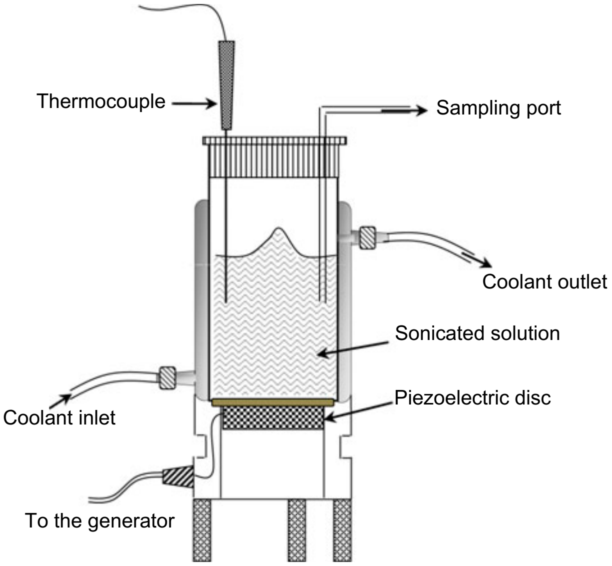

Fig. 2. Scheme of the ultrasonic reactor.

Potassium iodide solution was sonicated in the above-described reactor. The absorbance was recorded with a UV–vis spectrophotometer at the maximum wavelength of the formed triiodide (I_3^-) (352 nm; the molar absorptivity $\epsilon = 26{,}000$ L mol^{-1} cm^{-1}).

Fricke solution was prepared by dissolving $FeSO_4(NH_4)_2SO_4{\cdot}6H_2O$ (10^{-3} M), H_2SO_4 (0.4 M) and NaCl (10^{-3} M) in water. The obtained solution was sonicated and the absorbance of Fe^{3+} formed was measured at 304 nm (the molar absorptivity $\epsilon = 2{,}197$ L mol^{-1} cm^{-1}) by using a UV–vis spectrophotometer.

Hydrogen peroxide concentrations formed during water sonolysis were analytically determined using the iodometric method [18]. The iodide ion (I^-) reacts with H_2O_2 to form the triiodide ion (I_3^-) that absorbs strongly at 352 nm ($\epsilon = 26{,}000$ L mol^{-1} cm^{-1}). Sample aliquots taken from the reactor were added in the quartz cell of the spectrophotometer containing potassium iodide (0.1 M) and ammonium heptamolybdate (0.01 M). The mixed solutions were allowed to stand for 5 min before absorbance was measured. In the dye solution, the accumulated concentration of H_2O_2 was measured using the same procedure as the resulted absorption band of I_3^- ($\lambda_{max} = 352$) is more distinct from those of the dye (two bands with $\lambda_{max} = 287$ and 543 nm), which allow an easy monitoring of the peak intensity at 352 nm without any interference with the dye absorption bands.

3. Results and discussion

3.1. Characterization of the sonochemical reactor

The direct quantification of the species inducing sonochemistry, i.e. radicals such as $^{\bullet}OH$, $H^{\bullet}$, and $HO_2^{\bullet}$, is still not possible. However, there are several chemical dosimetries, which might be used to monitor the generation of hydroxyl radicals: Fricke dosimetry [27], iodide dosimetry [28], and dosimeters based on organic substances such as terephthalate [29] or salicylic acid [30]. Sonochemical production of hydrogen peroxide, nitric or nitrous acid in water is also used as a chemical dosimeter [20,31]. All these chemical methods are based on oxidation reactions occurring in an aqueous solution. In the present work, three dosimetries, namely iodide dosimetry, Fricke dosimetry, and the production of H_2O_2 in water, were used for characterizing, in term of radicals' production, the 600-kHz ultrasound reactor used for the degradation experiments. A brief description of these methods is given below:

(1) When aqueous solution of potassium iodide (KI) is irradiated, oxidation occurs and I^- ions are oxidized to I_3^- by means of $^{\bullet}OH$ radicals generated acoustically. The main reactions occurring in this method are shown below (reactions (3)–(6)) [20]. The amount of the generated I_3^- ions was quantified spectrophotometrically at 352 nm:

$$^{\bullet}OH + I^- \rightarrow I + OH^- \tag{3}$$

$$I + I^- \rightarrow I_2^- \tag{4}$$

$$2I_2^- \rightarrow I_2 + 2I^- \tag{5}$$

$$I_2 + I^- \rightarrow I_3^- \tag{6}$$

(2) When ultrasonic waves are passed through Fricke solution, Fe^{2+} ions in the solution are oxidized to Fe^{3+} ions as follows [20]:

$$^{\bullet}OH + Fe^{2+} + H^+ \rightarrow Fe^{3+} + H_2O \tag{7}$$

$$H_2O_2 + Fe^{2+} + H^+ \rightarrow Fe^{3+} + H_2O + {}^{\bullet}OH \tag{8}$$

$$HOO^{\bullet} + Fe^{2+} + H^+ \rightarrow Fe^{3+} + H_2O_2 \tag{9}$$

The amount of Fe^{3+} ions can be then quantified using UV spectrophotometer at 304 nm.

(3) As indicated earlier, if there are not any solutes in the liquid medium, the primary radicals recombine at the bubble–solution interface to form hydrogen peroxide (H_2O_2) according to reactions (1) and (2). As $k_1 >> k_2$, the yield of H_2O_2 can be used to estimate the amount of $^{\bullet}OH$ radicals released by the bubbles at determined sonochemical conditions [20].

Fig. 3 shows the evolution of I_3^-, Fe^{3+}, and H_2O_2 concentrations during the sonication at 600 kHz and 90 W of, respectively, KI solution (0.1 M), Fricke solution and pure water. The three species concentrations evolve linearly with time, following a zero-order kinetic law. This linear increase in the sonochemical activity is generally expected as the sonication time corresponds to a linear increase in the power input. Thus, the three dosimetries demonstrated the viability

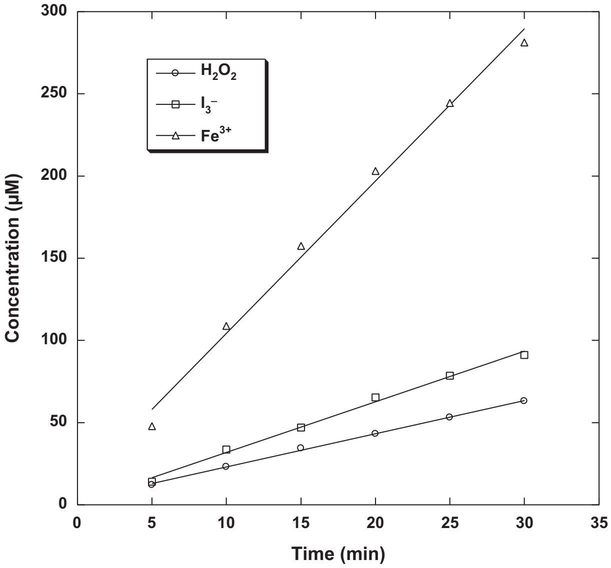

Fig. 3. Evolution of the species H_2O_2, I_3^-, and Fe^{3+} concentrations during the sonication of KI (0.1 M), Fricke solution and pure water, respectively (conditions: volume: 300 mL, temperature: 25˚C, frequency: 600 kHz, ultrasonic power: 90 W).

of the 600-kHz reactor to produce chemical oxidants, particularly ·OH. The obtained production rates of I_3^-, Fe^{3+}, and H_2O_2 with respect to the supplied ultrasonic power are presented in Fig. 4. An increase in the chemical reactivity of ultrasound is observed with an increase in ultrasonic power from 30 to 90 W. Therefore, the production of the oxidants is strongly sensitive to the variation in operational condition, i.e. supplied power.

3.2. Degradation of BF

3.2.1. Characteristics of BF sonolysis

The degradation of the dye during the reaction period may be assessed by absorption abetments at two bands: the visible, which is responsible for the chromophoric group of the dye and the UV at 287 nm, which represents the absorption of the aromatic derivatives rings. However, the absorption decay at 287 nm did not follow a common kinetic pattern as it did in the visible band to avoid confliction between intermediates (formed during oxidative degradation of the dye) absorption in the UV region.

Fig. 5 displays the evolution of the UV–vis spectra during the sonolysis at 600 kHz and 90 W of ultrasonic power of 10 mg L^{-1} BF solution at several time intervals. In absence of ultrasound, the absorption spectrum presents two main bands: the first is in the visible region (responsible for the color), with a maximum absorption at 543 nm, and the second is in the UV region, with a maximum absorption at 287 nm. Following a period of sonolysis, these characteristic absorption bands decreased in intensity and undergo a change in their spectral shape, indicating the degradation of the dye and the formation of absorbing products in the UV region. At this level, it should be mentioned that BF is a water-soluble compound (solubility: 2,650 mg L^{-1} at 25°C [32]) which suggests a negligible volatility in water (vapor pressure: 7.49×10^{-10} mmHg at 25°C [32], Henry's law constant: 2.28×10^{-15} atm m^3 mol^{-1} [32]); consequently, it cannot enter the bubbles to be incinerated by the high core temperature but must be eliminated through reaction with hydroxyl radicals (·OH) at the outside of the collapsing bubbles. Fig. 5 shows that after 60 min of sonication, the color decay was 81%, whereas the absorbance decay at 287 nm was only 37%, signifying the priority of ·OH attack to the chromophoric group of the dye and hence, the rapidly increase in aromatic intermediates that likely absorb in UV region, which is clearly reflected by the appearance of a new absorption peak at 206 nm. When the reaction progressed (>60 min), the absorption peaks at 543 and 287 nm completely disappeared and the intensity of the 206 nm peak increased with increasing irradiation

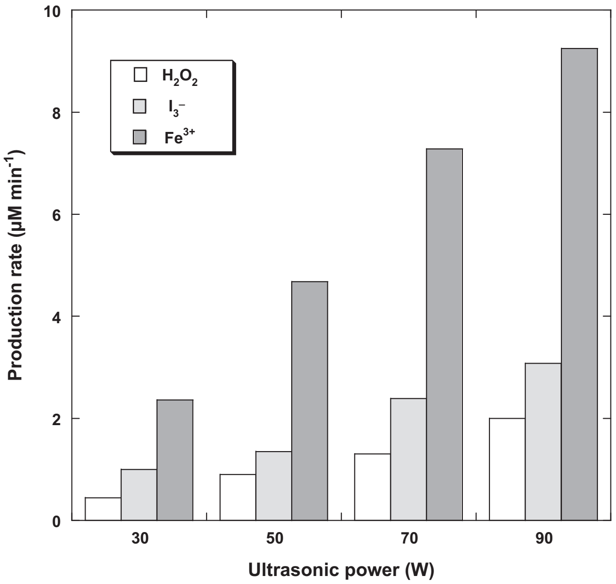

Fig. 4. Dependence of the rate of H_2O_2, I_3^-, and Fe^{3+} formation on ultrasonic power during the sonication of 0.1 M KI, Fricke solution and pure water, respectively (conditions: volume: 300 mL, temperature: 25°C, frequency: 600 kHz, ultrasonic power: 90 W).

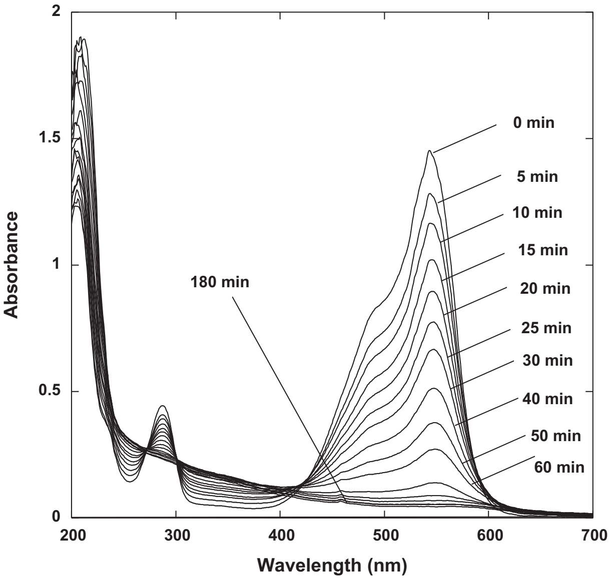

Fig. 5. Time evolution of the UV–vis spectra of BF solution upon ultrasonic action (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25°C, pH 5.9, frequency: 600 kHz, ultrasonic power: 90 W).

time, indicating the increase in the concentration of the BF byproducts as well as nitrous and nitric acids that absorb light at ~200 nm. Particularly, nitrous and nitric species have been detected and quantified in several works [33–35]. At 300 kHz and 80 W, Torres-Palma et al. [34] found that both nitrite and nitrate ions concentrations in water increased with sonication time, reaching a maximum (approximately 300 μM for both nitrite and nitrate) after 90 and 150 min, respectively. At 447 kHz, Mead et al. [35] reported that HNO_2 and HNO_3 are formed in sonicated aerated water with initial rates of 22 and 6 μM min^{-1}, respectively. In our case (600 kHz, 90 W), HNO_2 and HNO_3 were not quantified but the decrease in the solution pH from an initial value of 5.9 to 2.8 after 160 min of sonication reflected the formation of these acidic species.

In order to verify the priority of ·OH radical attack rather than pyrolysis in the bubble for BF sonodegradation, the evolution of the H_2O_2 concentration during sonication was monitored in the absence and presence of BF. Fig. 6 shows the kinetics of BF (10 mg L^{-1}) degradation and the H_2O_2 evolution in the absence and presence of BF during sonolysis at 600 kHz and 90 W. While the BF showed an exponential decay (BF was completely destroyed after 160 min), the H_2O_2, produced primarily from hydroxyl radical recombination, evolved in linear manner with a formation rate of 2 μM min^{-1} in the absence of BF. In the presence of BF, a lower formation rate was observed (1.66 μM min^{-1}), indicating that a fraction of ·OH reacts with the pollutant molecules. This observation confirmed that ·OH radical attack at the outside of the cavitation bubble is the mean pathway for the sonolytic degradation of BF.

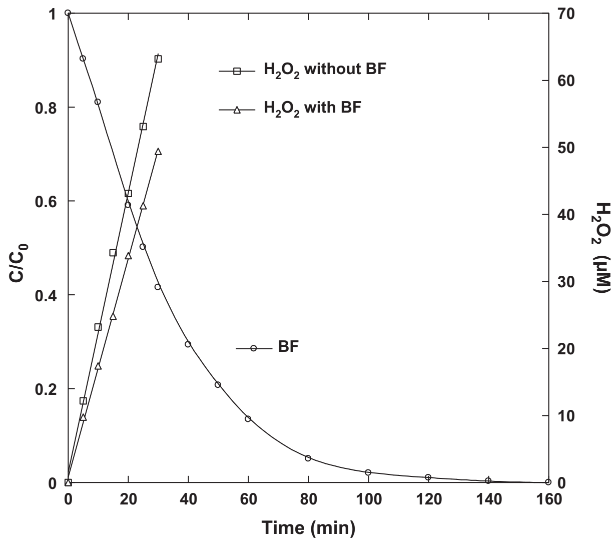

Fig. 6. BF degradation and hydrogen peroxide production for aerated water (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25°C, pH 5.9, frequency: 600 kHz, ultrasonic power: 90 W).

3.2.2. Effect of initial BF concentration

BF solutions of different initial concentrations (1, 2, 5, 10, and 15 mg L^{-1}) were submitted to sonolysis at 600 kHz and 90 W. Fig. 7 depicts the results of the degradation kinetics of BF for the five initial concentrations. The results show that ultrasonic action is efficient toward the degradation of BF, particularly when the dye is present at low concentration. In all cases, the concentration of BF decreases exponentially with time, suggesting apparent first-order kinetics law. The removal percentage of BF (calculated after 25 min of treatment) decreases progressively with increasing the initial dye concentration but, however, the removed amount of the dye increased with the rise of its initial concentration (Fig. 7, the insert). These findings are in line with that reported in the literature [36–38].

On the other hand, several authors [34,39–42] have recently claimed that the sonolytic degradation of organic pollutants does not obey first-order kinetics. To confirm this hypothesis, the initial BF degradation rates (calculated from Fig. 7 as $\Delta C/\Delta t$ after few minutes of sonication) were evaluated and displayed as function of initial BF concentration in Fig. 8. It was remarked that the higher the substrate concentration, the higher the initial decomposition rate. However, a linear relationship was not observed as expected for a first-order kinetic and thus, effectively, the degradation of BF cannot be associated with a first-order kinetic law. Consequently, the degradation kinetics depends on the concentrations of both the dye and ·OH radical.

In order to clarify the reactivity zone of BF sonooxidation, our results were analyzed using two heterogeneous kinetic models based on a Langmuir-type mechanism. The first model (Eq. (10)), proposed by Okitsu et al. [43], is based on the fact that organic molecules adsorb and desorb from the liquid interface layer surrounding the cavitation bubble, assuming a pseudo-steady state and so the degradation occurred mainly at the liquid–bubble interface. The second model (Eq. (11)), proposed by Serpone et al. [44], assumed that the overall rate of solute decomposition r is the sum of the rates in the bulk solution and the interfacial layer:

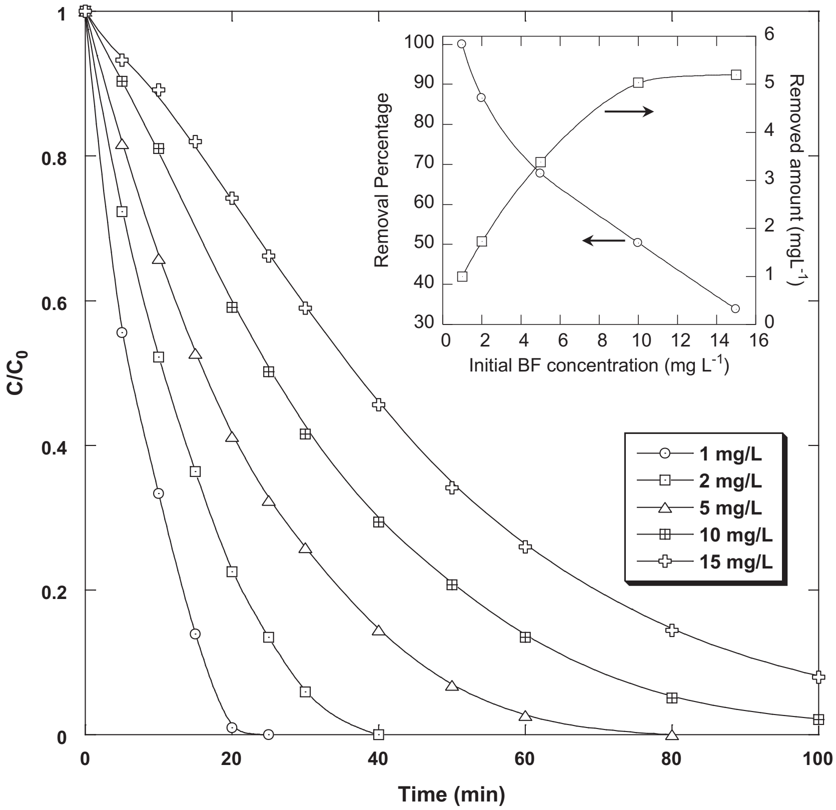

Fig. 7. Kinetics of BF degradation for various initial concentrations (conditions: volume: 300 mL, initial BF concentration: 1–15 mg L^{-1}, temperature: 25°C, pH 5.9, frequency: 600 kHz, ultrasonic power: 90 W). The inserted figure represents the removal efficiency, and the removed amount calculated after 25 min of ultrasonic treatment, as function of initial BF concentration.

$$r = \frac{kKC_0}{1 + KC_0} \tag{10}$$

$$r = K_b + \frac{kKC_0}{1 + KC_0} \tag{11}$$

where K_b is a constant representing the rate of decomposition in the bulk liquid (mg L^{-1} min^{-1}), r is the initial degradation rate (mg L^{-1} min^{-1}), k is the pseudo-rate constant (mg L^{-1}), K is the equilibrium constant (L mg^{-1}), and C_0 (mg L^{-1}) is BF initial concentration.

The models parameters were determined by nonlinear curve fitting method using KaleidaGraph© software. This method was found to be the more appropriate technique to determine the model parameters [45]. The obtained values of the two models parameters are listed in Table 1 and the corresponding theoretical curves were superimposed on the experimental data points (Fig. 8). It was clearly seen that the two models perfectly described the sonolytic destruction of BF. This indicates that the degradation of the dye mostly takes place at the bubble/solution interface by hydroxyl radical attack, whereas some radical reactions also occurred in the bulk of the solution as the Serpone's model showed the more perfected fitting of the experimental data at low dye concentration (1 mg L^{-1}). On the basis of this conclusion, the effect of initial BF concentration on the initial degradation rate can be explained as follow: at low dye concentration, a high fraction of $^{\cdot}OH$ generated would recombine to yield H_2O_2 (Fig. 8) and the oxidation reaction takes place far from the surface of the collapsing bubble where the concentration of $^{\cdot}OH$ radical is relatively high [46]. This conducted to lower degradation rates of BF (Fig. 8). In contrast, at high BF concentrations (>1 mg L^{-1}), the degradation is likely occurred at the interface of the collapsing bubble. As a result, high portion of $^{\cdot}OH$ radical will be scavenged by the dye molecules, resulting in high degradation rate and lower production rate of H_2O_2 as observed in Fig. 8. It should be noted that the higher and lower levels of H_2O_2 at, respectively, lower and higher substrate concentration are largely reported in the literature [39,42,45–47].

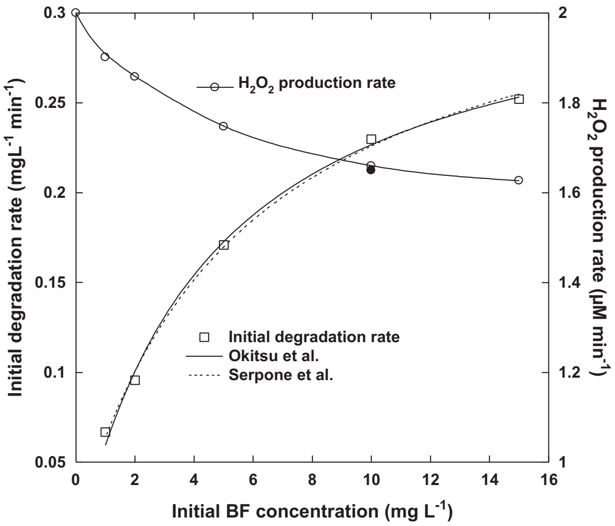

Fig. 8. Effect of initial BF concentration on the initial degradation rate and the production rate of H_2O_2. The predicted curves of the initial degradation rate vs. initial BF concentration are superimposed on the experimental data points (conditions: volume: 300 mL, initial BF concentration: 1–15 mg L^{-1}, temperature: 25˚C, pH 5.9, frequency: 600 kHz, acoustic power: 90 W). The production rate of H_2O_2 in BF solution, $(r_{H_2O_2})_s$, was evaluated by the equation [36]: $(r_{H_2O_2})_s = (r_{H_2O_2})_w - 0.5 r_{BF}$, where $(r_{H_2O_2})_w$ is the production rate of H_2O_2 in pure water and r_{BF} is the initial degradation rate of the BF. $(r_{H_2O_2})_w$ was given at 90 W form Fig. 4 as 2 μM min^{-1}. The dark dote (●) near the curve of H_2O_2 represents the experimental value of the production rate of H_2O_2 during the sonication of 10 mg L^{-1} BF (the error between experimental and predicted rates of H_2O_2 at 10 mg L^{-1} BF is ~1%, showing an excellent agreement between them).

3.2.3. Effect of power

The applied ultrasonic power is an important parameter that determines the cost effectiveness of the sonolytic process. In order to study the effect of power on the degradation of BF, sonolytic experiments were carried out using 30, 50, 70, and 90 W for an initial BF concentration of 10 mg L^{-1}. The selected range of power is in an interval between the cavitation threshold, below which no reaction is observed, and a maximum intensity above which the sonochemical effects are attenuated [48]. A significant enhancement in the BF degradation rate was observed when the ultrasonic power was increased as shown in Fig. 9. For example, the removal efficiency increased from 40.5% at 30 W to 75 and 97% at 70 and 90 W, respectively. Correspondingly, the initial degradation rate increased notably with the ultrasonic power increase (Fig. 9, the inset), which is in good agreement with previous studies on the sonochemical degradation of several organic pollutants [39–42,47]. The initial BF degradation rate increased by factor of 2, 2.7, and 3.2 when the ultrasonic power increased from 30 W to, respectively, 50, 70, and 90 W.

The reason for the observed enhancement in the rate at higher ultrasonic power may be explained as follow. The acoustic bubble during collapse can be considered as a closed microreactor within which high-temperature chemical reactions occur. In this situation, the extent of free radicals, principally $^{\bullet}OH$, production in the bubble will be controlled by means of three factors: the bubble temperature, the amount of water vapor trapped in the bubble, and the collapse time. An increase in power results in an increase in acoustic amplitude that favors more violent collapse. The acoustic amplitude, P_A, is given by the following equation [9]:

$$P_A = (2 I_a \rho_L c)^{1/2} \tag{12}$$

where I_a is acoustic intensity (power per unit area), ρ_L is the density of the liquid, and c is the speed of sound in the liquid (1,500 m s^{-1} at 25˚C). With an increase in acoustic amplitude, the expansion ratio of the cavity R_{max}/R_0 as well as the compression ratio R_{max}/R_{min} increase (R_{max}, and R_{min} are, respectively, the maximum and the minimum bubble radius reached during bubble oscillation, whereas R_0 is the initial bubble radius) [49]. Using the adiabatic collapse approach [49], R_{max}/R_{min} and R_{max}/R_0 are correlated with the collapse time (τ_c), the maximum bubble temperature (T_{max}) and the amount of water vapor trapped at the collapse (y_{H_2O}) as [9,49]:

Table 1
Parameters of the Okitsu et al. and Serpone et al. models obtained using the non-linear curve fitting method

Model	K_b (mg L^{-1} min^{-1})	k (mg L^{-1} min^{-1})	K (L mg^{-1})
Okitsu et al.	–	0.3301	0.2193
Serpone et al.	0.0136	0.3317	0.1776

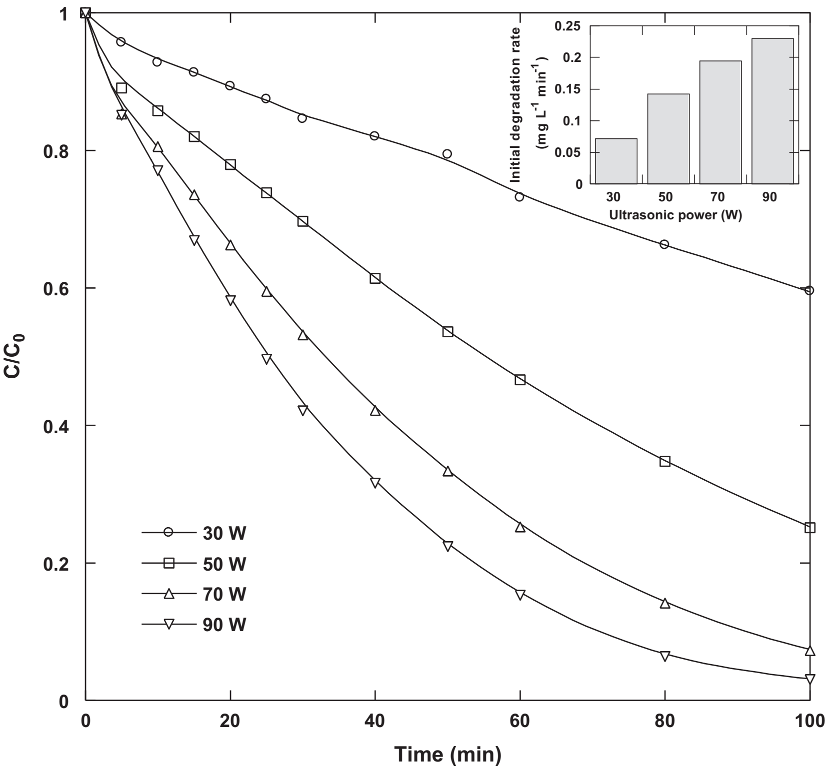

Fig. 9. Kinetics of the sonochemical degradation of BF at various ultrasonic powers (conditions: volume: 300 mL, initial dye concentration: 10 mg L^{-1}, temperature: 25°C, pH 5.9, frequency: 600 kHz).

$$\tau_c = 0.915 R_0 \left(\frac{R_{\max}}{R_0}\right)\left(\frac{\rho_L}{P_h}\right)^{1/2} \tag{13}$$

$$T_{\max} = T_\infty \left(\frac{R_{\max}}{R_{\min}}\right)^{3(\gamma-1)} \tag{14}$$

$$y_{H_2O} = P_v \left[P_v + P_{g0}\left(\frac{R_{\max}}{R_0}\right)^3\right]^{-1} \tag{15}$$

where T_∞ is the bulk liquid temperature, γ is the ratio of specific heats capacities (c_p/c_v) of the gas/vapor mixture, P_v is the vapor pressure, P_{g0} is the gas pressure in the bubble at its ambient state, and P_h is the hydrostatic pressure (1 atm).

Thus, higher bubble temperatures will be reached at higher compression ratios (Eq. (14)). Similarly, the higher the expansion ratio, the higher will be the amount of water vapor trapped in the bubble at the collapse (Eq. (15)). The increase in both the amount of the trapped water and the collapse temperature with increasing acoustic amplitude promotes the formation of free radicals, since they result essentially from the dissociation of the water vapor molecules inside the bubble. In addition, as the collapse time (τ_c) is proportional to the expansion ratio of the bubble (Eq. (13)), chemical reactions in the bubble at higher power have enough time to evolve and then convert reactant molecules to free radical and atoms. Consequently, an increase in acoustic amplitude will thus result in greater sonochemical effects inside a single collapsing bubble.

On the other hand, the number of the collapsing bubble may also increase at higher ultrasonic power [19,50]. Therefore, the observed enhancement in the rate of BF degradation at higher powers, interpreted earlier with respect to the single-bubble event, may also be due to the increased number of active cavitation bubbles and hence the generation of more hydroxyl radicals in the medium, resulting in higher BF degradation rates.

3.2.4. Effect of saturation with N_2

The nature of the dissolved gas is an important factor that affects the internal bubble chemistry and thus controls the extent of hydroxyl radical generation. In our case, nitrogen was continuously dissolved before and until complete degradation of BF solution (10 mg L^{-1}) at 600 kHz and 90 W. The results of

Fig. 10 illustrate the effect of N_2 saturation on the degradation of the dye. The presence of N_2 decelerated the BF removal. The initial degradation rate was reduced by more than 40%. Similar results were found previously for naphthol blue black [36] and ibuprofen [42].

The nature of the dissolved gas influence sonolysis through the single bubble yield and the number of active bubbles [51]. A gas with greater polytropic index γ ($\gamma = c_p/c_v$) and lower thermal conductivity produces higher temperature upon collapse resulting in higher sonochemical reactivity inside the collapsing bubble. On the other hand, gas with higher solubility can create more nucleation sites, and improve the cavitation events by increasing the number of bubbles. In our case, air and N_2 have the same polytropic ratio ($\gamma_{Ar} = 1.4$) and thermal conductivity ($\lambda = 0.026$ W m^{-2} K^{-1} [52]) and hence the implosion for the two cases (air and N_2 saturations) will generate the same bubble temperature (Eq. (14)). Therefore, the beneficial effect of air compared to N_2 was mainly attributed to the nature of the reactants trapped in the bubble at the collapse ($O_2/N_2/H_2O$ in case of air and N_2/H_2O in the case of N_2) and the amount of the dissolved gas in liquid phase. In our previous report [51], we carried out numerical simulations of chemical reactions inside a collapsing air and N_2 bubbles. We found that the production rate of hydroxyl radical is strongly sensitive to the amount of N_2 trapped in the bubble at the collapse, and the higher the concentration of N_2, the lower was the production rate of ·OH radical from the collapsing bubble. The reason for this trend was associated to the consumption of ·OH radical through the reaction $NO + {}^{\cdot}OH + M \leftrightarrow HNO_2 + M$. Thus, because NO was found to be formed mainly through the reactions $N_2 + O \rightleftharpoons NO + N$ and $NO_2 + M \rightleftharpoons O + NO + M$, the higher the concentration of N_2 in the bubble, the higher will be the concentration of NO and this accelerates the consumption rate of ·OH radical through the reaction $NO + {}^{\cdot}OH + M \rightleftharpoons HNO_2 + M$. Therefore, basing on single-bubble chemistry results, it is well justified that between air and N_2, the degradation rate of BF follows the order air > N_2 as the production rate of ·OH radical is higher in the case of air bubble than N_2 bubble. In addition to all of this, there is the fact that air will generate a greater number of bubbles in the BF solution as its solubility is relatively higher than that of N_2 ($x_{air} = 1.524 \times 10^{-5}$, $x_{N2} = 1.276 \times 10^{-5}$ [53]).

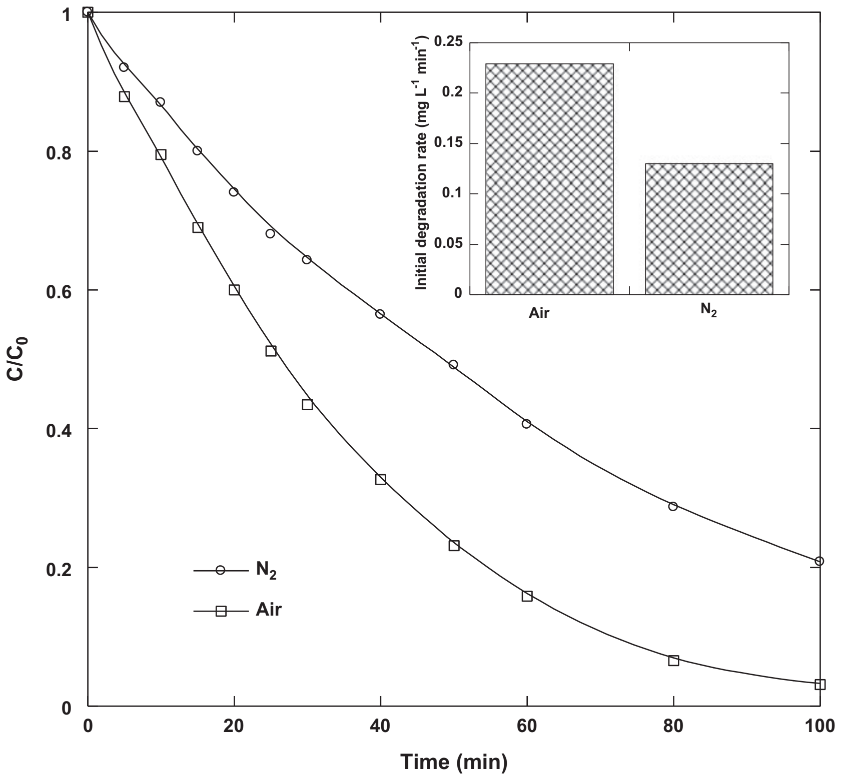

Fig. 10. Effect of the nature of dissolved gas on the sonochemical degradation of BF (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25˚C, pH 5.9, frequency: 600 kHz, ultrasonic power: 90 W).

3.2.5. Effect of liquid temperature

To assess the effect of liquid temperature on the sonochemical removal of BF (10 mg L^{-1}), experiments (600 kHz, 90 W) were performed at three liquid temperatures: 25, 35, and 45˚C. The obtained results (Fig. 11) showed that the rise in temperature from 25 to 35˚C enhances the degradation rate of the dye by factor of 1.3. However, further increase in temperature to 45˚C shows no further enhancement in BF removal. It seems that there exists an optimum temperature (~35˚C) for the degradation of BF. This result is in line with that of Jiang et al. [46] that found that the degradation rate of 4-chlorophenol as well as the production of H_2O_2 at 500 kHz increase with increasing in solution temperature up to 40˚C and decrease afterward. A similar trend was also reported by Entezari and Kruus [54] at 900 kHz where an optimum liquid temperature is observed for the sono-oxidation of KI in aqueous solution.

The change in the liquid temperature affects both the single-bubble sonochemistry and the number of bubbles. An interesting discussion about the dependence of these two parameters to the liquid temperature has been made elsewhere [55]. Briefly, as the bulk liquid temperature increases, the liquid vapor pressure increases and consequently more water vapor is trapped in the bubble at the collapse. Vapor dissociation inside a bubble at the collapse by the high temperature cools the bubble considerably due to the endothermal heat of the dissociation ($H_2O \rightarrow {}^{\cdot}OH + {}^{\cdot}H$, $\Delta H = 508.82$ kJ mol^{-1} [56]), which makes the maximum bubble temperature much lower for high liquid temperature (Eq. (14)). As consequence, the production of radicals, i.e. ·OH, for a single bubble will be lower at lower liquid temperature. On the other hand, higher liquid temperature produces a greater number of bubbles, which enhances the production rate of ·OH radical. The competition between these two factors, the single bubble yield and the number of bubbles, yields the observed optimum temperature for the degradation of BF. From the results of Fig. 11, it can be concluded that the effect of rising liquid temperature in the interval 25–35˚C is controlled by the number of bubbles, whereas the strong competition takes place above 35˚C, resulting in a fixed production rate of radicals in the BF solution as can be seen in the insert of Fig. 11.

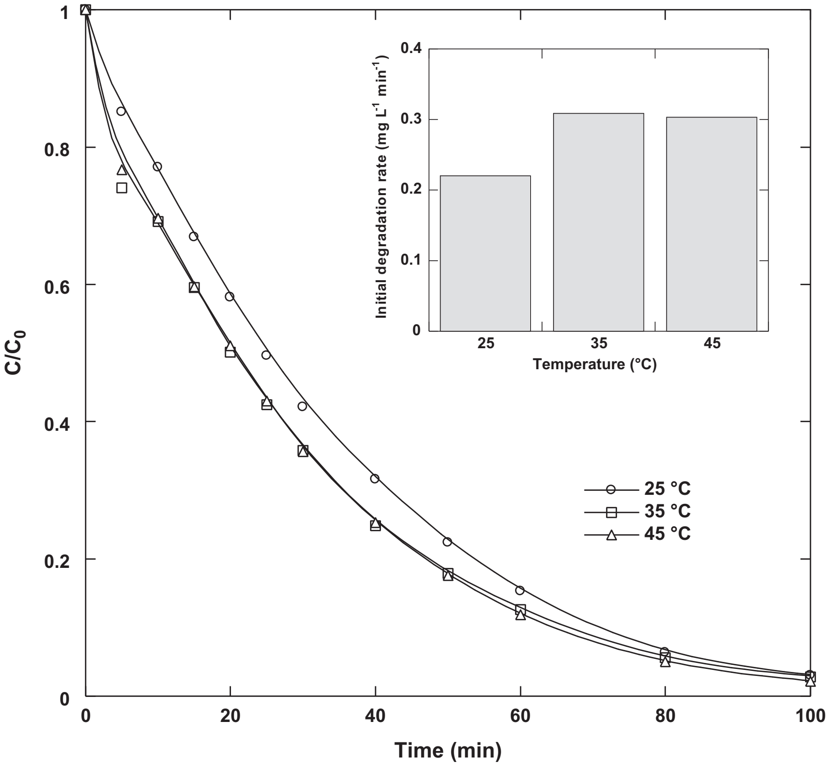

Fig. 11. Effect of liquid temperature on the sonochemical degradation of BF (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25 and 45˚C, pH ~5.9, frequency: 600 kHz, ultrasonic power: 90 W).

3.2.6. Effect of dissolved salts and natural matrix

It was recently reported that sonochemical treatment of organic pollutants may be enhanced (intensified in some cases [57–59]) in the presence of some mineral anions usually present in water [39,41,47,57–61]. To verify this suggestion toward the degradation of BF, experiments (600 kHz, 90 W and 10 mg L^{-1} BF) were conducted in solutions containing NaCl and Na_2SO_4 in a concentration range of 0.1–10 g L^{-1}. The obtained results are shown in Figs. 12 and 13 for NaCl and Na_2SO_4, respectively. The presence of NaCl and Na_2SO_4, even at high concentration, showed an enhancement in the rate of BF elimination. However, the salt loading was not shown a significant effect.

The fact that sonochemical removal of substrate, at least, was not affected by the presence of salts, this making ultrasonic treatment a promising technique for removing organic pollutant form industrial and natural waters which usually containing salts. To verify this hypothesis, sonolytic experiments (600 kHz, 90 W and 10 mg L^{-1} BF) have been conducted in real natural water. The natural water characteristics are pH 7.2, Ca^{2+} = 81 mg L^{-1}, Mg^{2+} = 24 mg L^{-1}, Na^+ = 15.8 mg L^{-1}, Cl^- = 72 mg L^{-1}, SO_4^{2-} = 53 mg L^{-1}, HCO_3^- = 265 mg L^{-1}, and TOC = 0 mg L^{-1}. The obtained results are depicted in Fig. 14. As can be seen, the high quantity of salts present in the real water does not reduce the efficiency of the ultrasonic process. Contrarily, BF removal is more efficient in the natural water than in pure water. Additionally, unlike the other AOPs when the negative effect of HCO_3^-, a well-known $^\cdot$OH scavenger ($k_{\mathrm{HCO_3^-}-{}^\cdot\mathrm{OH}} = 8.5 \times 10^6\ \mathrm{M^{-1}\ s^{-1}}$) is well established (i.e. photocalysis [62], Fenton [63], UV/H_2O_2 [64], and UV/O_3 [65]), the presence of 265 mg L^{-1} HCO_3^- did not affect the degradation of the dye. It has been recently reported that the effect of HCO_3^- on the sonochemical degradation of organic pollutants is notably only at low substrate concentration [57–59].

The positive effect of salts on the sonochemical degradation of BF was attributed to the salting out effect on both the solute and the dissolved gas. On one hand, salts present during sonication of aqueous solutions may increase the ionic strength of the aqueous phase. This phenomenon (commonly called "salting-out" effect) pushes organic pollutants toward the bubble–bulk interface where the concentration of oxidants is high and, therefore, leads to a higher degradation rate [60]. On the other hand, Brotchie

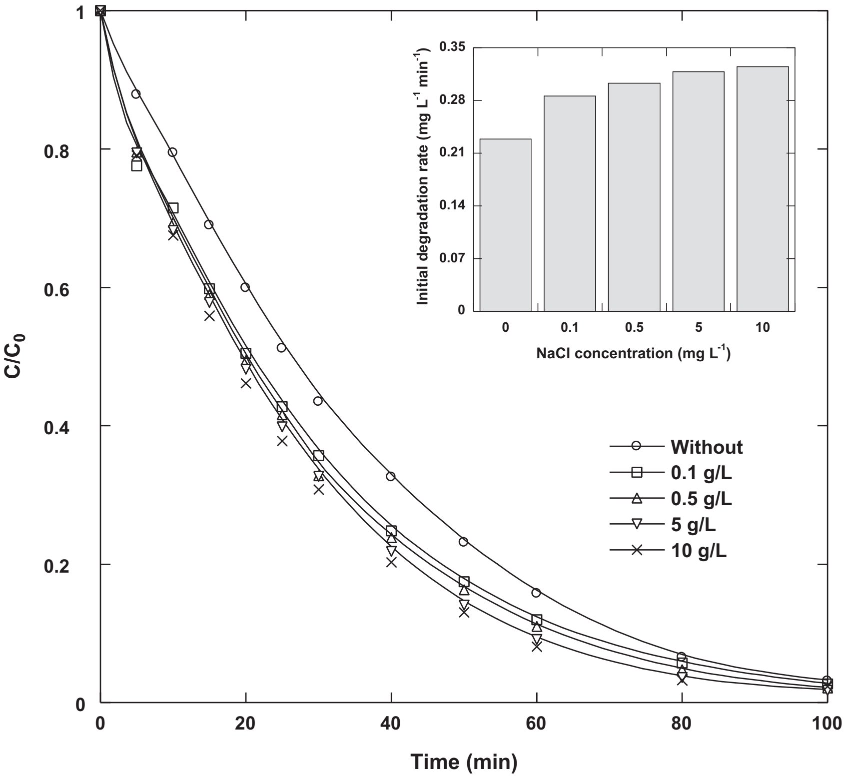

Fig. 12. Effect of NaCl addition on the sonochemical degradation of BF (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25°C, pH ~5.9, frequency: 600 kHz, ultrasonic power: 90 W).

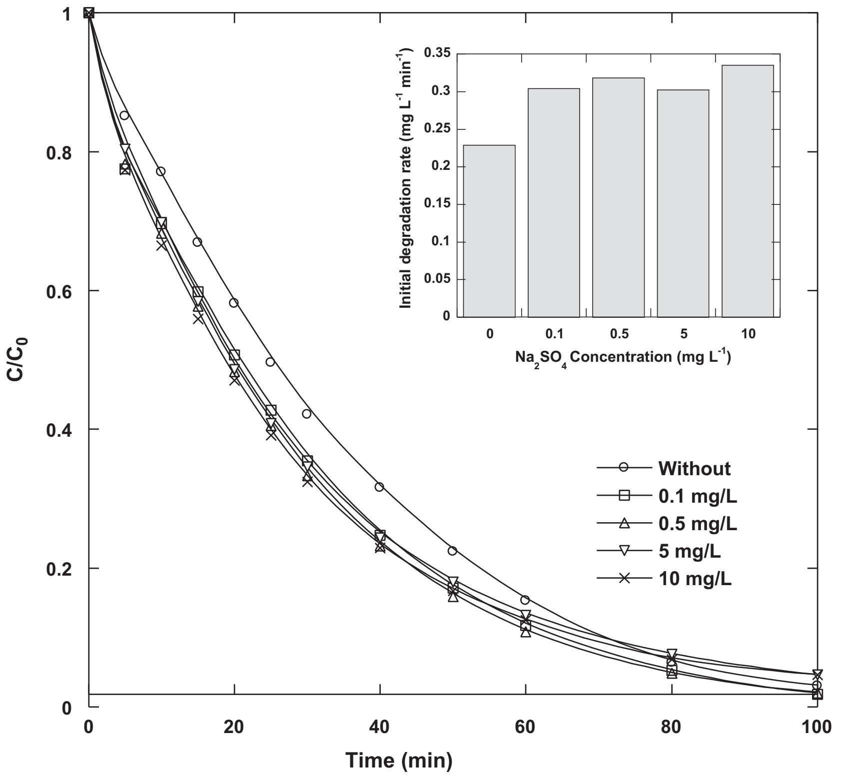

Fig. 13. Effect of Na_2SO_4 addition on the sonochemical degradation of BF (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25°C, pH ~5.9, frequency: 600 kHz, ultrasonic power: 90 W).

et al. [66] showed that the effects of salts could be completely attributed to their "salting-out" on the dissolved gas (*not on the substrate molecules*). They found that bubble coalescence is strongly proportional to the gas concentration in the sonicated liquid and that the presence of salts reduces the bubble coalescence, which resulted in smaller bubbles of higher sonochemical and sonoluminescing activities. This conclusion was supported by Wall et al. [67] which showed experimentally that of all physicochemical proprieties of water that are affected by the addition of salt (activity, ionic strength, viscosity, surface tension and gas solubility), gas solubility appeared to be the only parameter that links the general behavior of an increase in sonoluminescence intensity with increasing salt concentration in high-frequency ultrasonic cavitation field in water.

3.2.7. Effect of tert-butyl alcohol addition

Tert-butyl alcohol is a well-known hydroxyl radical scavenger ($k_{(CH_3)_3COH-{}^{\cdot}OH} = 6 \times 10^8$ M^{-1} s^{-1}). It is a volatile substrate that can easily penetrated in the bubbles and pyrolyzed via a free-radical-induced mechanism [68]. The tert-butyl alcohol is able to scavenge $^{\cdot}OH$ radicals in the gas phase of the bubble (reaction (16) [68]) and prevent their accumulation at the bubble interface:

$$(CH_3)_3COH + {}^{\cdot}OH \rightarrow (CH_3)_2CH_2COH^{\cdot} + H_2O \qquad (16)$$

Therefore, if $^{\cdot}OH$ radical is the main participant in the sonolytic oxidation of BF, the addition of tert-butyl alcohol to the reaction mixture should suppress the BF degradation. Sonolysis experiments (600 kHz, 90 W, and 10 mg L^{-1} BF) were performed in the presence of 1 and 10 mg L^{-1} of alcohol and the results are presented in Fig. 15. The presence of tert-butyl alcohol quenches, but not completely, the degradation of the dye. After adding 1 and 10 mg L^{-1} of alcohol, the efficiency of BF removal decreased to 10 and 3%, respectively. Thus, it can be concluded that $^{\cdot}OH$ radical plays the major role in the oxidation of BF. It can be also observed from Fig. 15 that the degradation of the dye in the presence of tert-butyl alcohol follows two steps: a slightly decrease during the first few minutes (~10 min), followed by a plateau ($C/C_0 = 0.9$ and 0.97 for 1 and 10 mg L^{-1} of alcohol, respectively). Thus, it

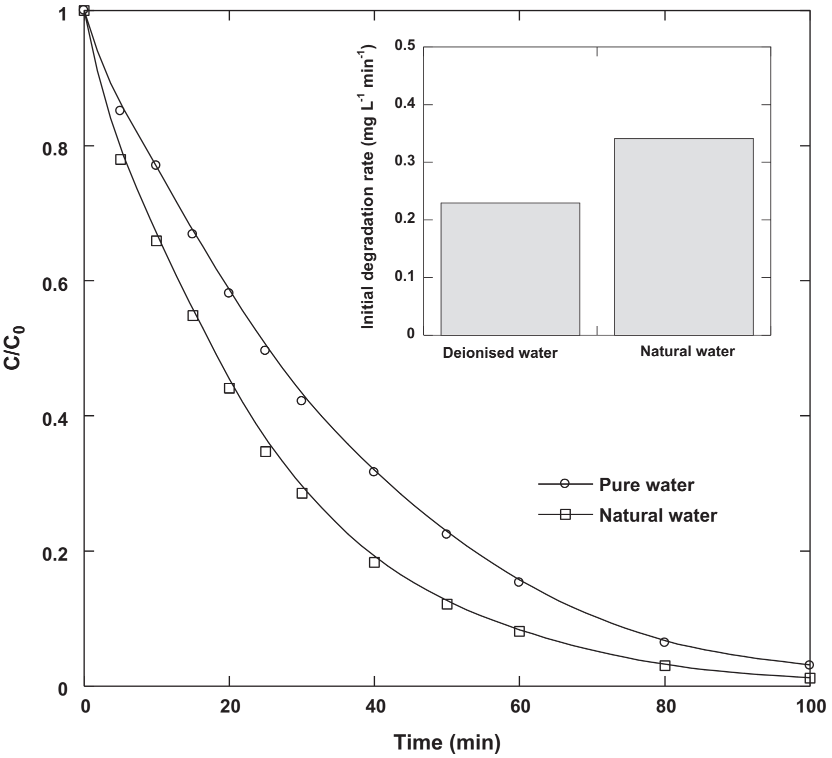

Fig. 14. Kinetics of the sonochemical degradation of BF in natural and pure waters (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25˚C, natural pH, frequency: 600 kHz, ultrasonic power: 90 W).

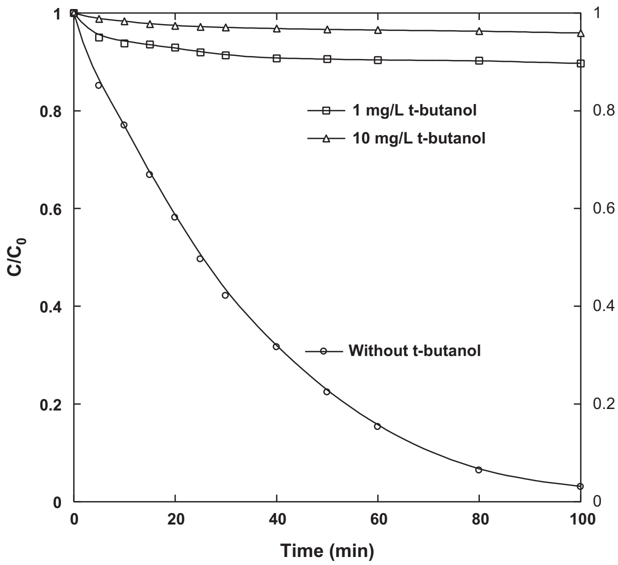

Fig. 15. Effect of tert-butanol addition on the sonolytic degradation of BF (conditions: volume: 300 mL, initial BF concentration: 10 mg L^{-1}, temperature: 25˚C, natural pH, frequency: 600 kHz, ultrasonic power: 90 W).

is difficult to calculate an order of reaction for BF degradation in the presence of alcohol.

4. Conclusion

The results presented in this work prove the potential of ultrasound as an AOP for treating water contaminated with BF. The dye oxidation occurred mainly through hydroxyl radical attack at the surface of the collapsing cavitation bubbles. The degradation rate of the dye was strongly affected by the operational conditions. It increased with increasing initial substrate concentration and ultrasonic power. The liquid temperature rise enhances the degradation rate of the dye, but an optimum of 35˚C was observed. The degradation of the dye was more efficient under air than N_2 saturation atmosphere. The presence of salts, even at high concentration, did not alter the degradation of the dye, making sonochemistry as one of the promising techniques for the treatment of industrial and natural waters. The presence of tert-butyl alcohol as a hydroxyl radical scavenger inhibited at high extent the degradation of BF, making sure that hydroxyl radical was the main species involved in the degradation of this compound. Finally, even if the present study furnished some interesting results concerning the sonolytic oxidation of BF, further efforts should be carried out to assess the oxidative scheme of dye.

Acknowledgments

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