



Oxidation study of an azo dye, naphthol blue black, by Fenton and sono-Fenton processes

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ABSTRACT

This work is focused on the combination of classic and modified (ultrasound-assisted) of the Fenton process, for the treatment of water contaminated by an azo dye. The experiments presented herein were carried out on naphthol blue black (NBB), and the obtained results demonstrate a remarkable elimination attaining 99%. The influence of different operating parameters, such as the concentrations of H₂O₂ and Fe²⁺, the pH and ultrasound (US) effect, on the dye degradation, was studied as well. The conditions for maximum decolorization efficiency were found to be a pH of 3 and initial concentrations of NBB, Fe²⁺ and H₂O₂ of 4.05 × 10⁻² mM, 0.05 mM and 2 × 10⁻³ mM, respectively, after 60 min of reaction time with a stirring rate of 300 rpm. It was found that under these conditions, a decolorization of NBB of 99% is achieved. In addition, the combination of low-frequency ultrasound (40 kHz and 30 W) with the Fenton process (US/H₂O₂/Fe²⁺) gave a better decolorization efficiency compared to H₂O₂/Fe²⁺ alone. Where the decolorization efficiency increased from 63% to 95% at 5 min of reaction time. This result may be caused by ultrasonic irradiation, which greatly increases the rate of H₂O₂ production. In view of these results, the sono-Fenton process can be used for the oxidation of NBB to reduce reaction time and chemical costs.

Keywords: Fenton; Decolorization; Naphthol blue black; Ultrasound; Treatment of water

1. Introduction

Dyes are widely used in industry, printing, food, cosmetics, clinical industry and textile dyeing owing to their chemical stability, ease of synthesis and versatility. Water pollution from colored effluents is one of the major environmental concerns in today's world. Dyes give water an undesirable color that is harmful to aquatic life; which leads to the blocking of the re-oxygenation capacity of the water and

prevents sunlight from entering deep waters, disrupting respiration and photosynthesis process [1–3].

Azo dyes are the most important class of commercial dyes. In addition, the azo dyes consist of one or more azo bonds (–N=N–) in the structure, which are the most difficult parts that can be removed and separated, giving mutagenic or carcinogenic intermediates [4,5].

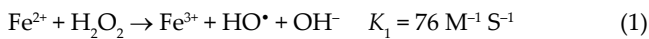
Their biological treatment is therefore of major scientific interest. Many techniques have been proposed for its

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removal from wastewater such as physical adsorption, electrochemical oxidation, chemical oxidation, chemical coagulation/precipitation, and biological anaerobic/aerobic decomposition [6–9]. These methods have proven ineffectiveness in meeting the standards required for these releases [10,11].

The hydroxyl radical (HO^\bullet) is a very reactive and non-selective species that can initiate oxidative degradation reactions of organic compounds in aqueous solution. HO^\bullet radicals can be produced by various oxidation systems, advanced oxidation processes (AOPs), such as the Fenton process [12], ozonation, electro-Fenton [13] or electrochemical oxidation, photo-Fenton [14], photocatalysis and ultrasound [15]. They represent one of the most promising options for the removal of persistent compounds present in wastewater effluents [16].

Fenton oxidation is one of the most favored AOPs carried at an acidic pH [17], in which ferrous ions react with hydrogen peroxide (H_2O_2) to generate hydroxyl radicals (Eq. (1)). The main reactions that occur during the Fenton process are presented in Eqs. (2)–(6) [18,19].



In recent years, the Fenton process has received considerable attention as an effective pretreatment method for less biodegradable wastewater because it is cost-effective, easy to treat, reacts well with organic contaminants and does not produce toxic compounds during oxidation. Moreover, it has a high chemical oxygen demand reduction efficiency [20,21].

Nevertheless, many researchers have worked on the coupling strategy between sonochemistry and various AOPs such as the Fenton process; they reported that this combination is more efficient than the independent application of each technique for water treatment [22,23]. The ultrasonic irradiation seems to improve the generation of hydroxyl

radicals by ultrasonic sound and minimize the amount of chemicals conventionally used in the Fenton process [24].

The objective of the present work is to study the decolorization of an azo dye, naphthol blue black (NBB), by the Fenton process. The effects of experimental parameters such as initial concentrations of NBB, Fe^{2+} and H_2O_2 , temperature, pH, ultrasound irradiation and the addition of salt (NaCl) on the degradation process were examined. Finally, the degradation of NBB present in various natural matrices such as natural mineral water and seawater was also investigated.

2. Materials and methods

2.1. Chemicals

The different chemical products used in this study are shown in Table 1. They were employed without preliminary purification.

2.2. Experimental procedure

The oxidation of the dye (NBB) is carried out in a 250 mL volume batch reactor. The volume of the dye solution used in experiments is fixed at 100 mL. The pH was adjusted using H_2SO_4 (0.1 N) or NaOH (0.1 N). A mass of catalyst and then a volume of hydrogen peroxide is added to the medium. The treatment is carried out at ambient temperature and under a constant stirring speed of 300 rpm. Samples were taken from the reactor using a pipette at fixed time intervals and were immediately analyzed.

A low-frequency ultrasonic reactor (40 kHz and 30 W) was used to study the combination of ultrasound with the Fenton process. A schematic of the device used for the sono-Fenton process is illustrated in Fig. 1. This reactor consists of a copper cooling system combined with a thermostated bath to maintain the temperature of the solution constant. The temperature is monitored by means of a digital thermometer.

2.3. Analytical methods

The determination of the concentration of NBB is performed using a spectrophotometric technique in the UV-visible region (SECOMAM-UviLine 9400). Spectronic quartz cell (1 cm path length) was used for holding samples. The maximum absorbance wavelength (λ_{max}) from the spectra was recorded at 618 nm.

The calibration curve established from dilute solutions of NBB on the basis of the wavelength mentioned indicates

Table 1
Chemicals products used in the treatment of NBB by Fenton and sono-Fenton processes

Chemical	Formula and purity (%)	Molar weight (g mol ⁻¹)	Supplier	
Naphthol blue black	$\text{C}_{22}\text{H}_{14}\text{N}_6\text{Na}_2\text{O}_9\text{S}_2$; 99%	616.49	Sigma-Aldrich	France
Iron sulfate	$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$; 99%	278.01	Chiminova	Danemark
Hydrogen peroxide	H_2O_2 ; 50%	34	Merck	France
Sulfuric acid	H_2SO_4 ; 96%–98%	98.08	VWR Chemicals	United States
Sodium hydroxide	NaOH; 99%	40	VWR Chemicals	United States
Sodium chloride	NaCl; 99.5%	58.44	Sigma-Aldrich	France

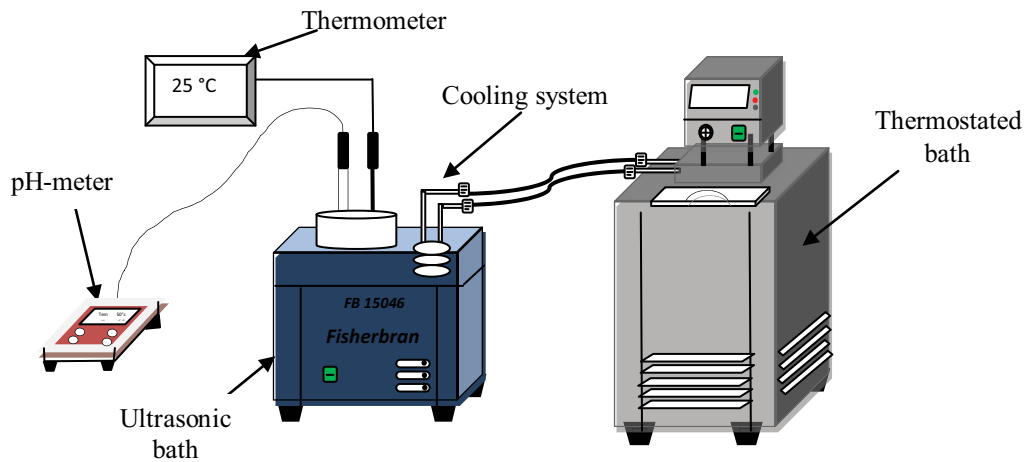


Fig. 1. The experimental setup used for sono-Fenton decolorization of NBB.

linearity and the possibility of analyzing this dye by this method (Fig. 2).

Eq. (7) was used to determine the degradation efficiency of NBB.

$$\text{Decolorization efficiency (\%)} = \left(1 - \frac{C_t}{C_0}\right) \times 100\% \quad (7)$$

where C_0 is the initial concentration of NBB at $t = 0$ while C_t is the concentration NBB at an instant t .

3. Results and discussion

3.1. Effect of initial pH

The pH of the dye solution is a very important parameter in the Fenton process. It has a direct effect on the oxidation mechanism of the dye because a change in the

pH of the solution causes a change in the Fe^{2+} concentration and therefore the production rate of the radicals HO^\bullet responsible for the oxidation of the dye. Therefore, it will be necessary to determine precisely the value to which the pH of the water must be fixed for all experiments.

The effect of pH on the decolorization of NBB by the Fenton process was studied at various values ranging from 1 to 9, at fixed concentrations of 2×10^{-3} mM of H_2O_2 and 0.05 mM of Fe^{2+} . It is important to note that the maximum absorption wavelength remained constant (618 nm) in the selected pH range. The obtained results are shown in Fig. 3. It is clear from this figure that the decolorization of NBB was maximum (about 99%) at pH 3 as compared to 14%, 50%, 96%, 10% and 4% at pH 1, 2, 4, 7 and 9, respectively, for a reaction time of 60 min. This result is in accordance with what was observed in the study of the oxidation of this dye by the $\text{Fe(III)P}_2\text{W}_{12}\text{Mo}_3/\text{H}_2\text{O}_2$ system, where the reaction was promoted at pH 3 [25]. This result

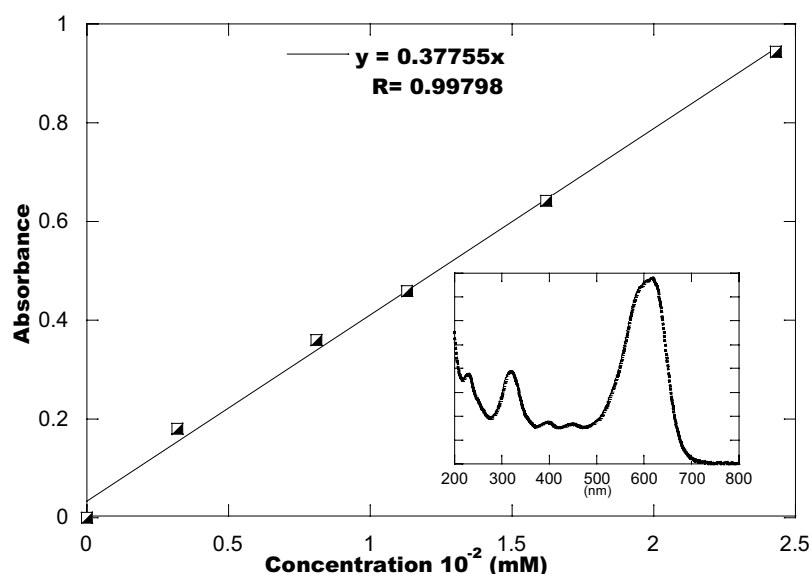


Fig. 2. Calibration curve of NBB at $\lambda_{\text{max}} = 618$ nm.

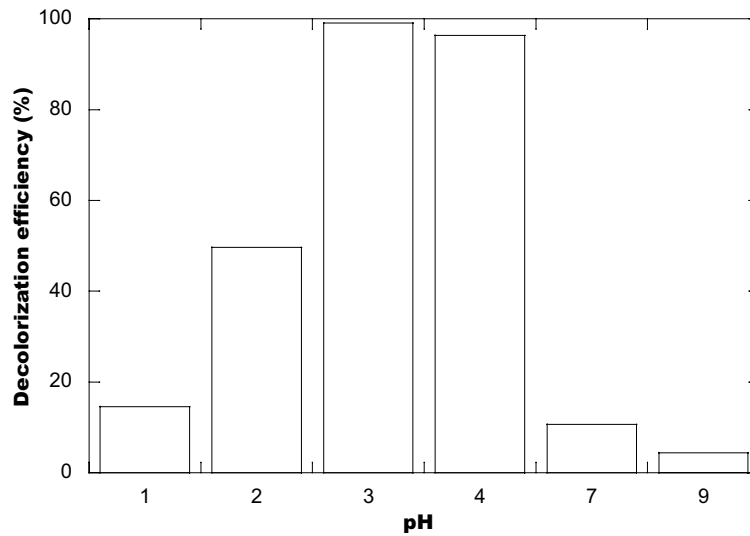


Fig. 3. Effect of initial pH on the decolorization of NBB by Fenton process. Experimental conditions: $[\text{NBB}]_0 = 4.05 \times 10^{-2}$ mM; $[\text{H}_2\text{O}_2]_0 = 2 \times 10^{-3}$ mM; $[\text{Fe}^{2+}]_0 = 0.05$ mM; $T = 25^\circ\text{C}$; pH 1–9.

can be explained by the stability of the catalyst at this pH. Some other studies showed that the degradation of dyes in aqueous solution by Fenton process was maximum in the pH range 3–4 [26]. Moreover, and knowing that in basic solutions, iron precipitates as iron hydroxide [27], some other studies proved that iron hydroxide has no influence on the oxidation of organic dyes [28].

3.2. Effect of iron concentration

In this study, the initial Fe^{2+} concentration was varied from 0.05 to 0.3 mM using fixed concentrations of H_2O_2 (2×10^{-3} mM) and NBB (4.05×10^{-2} mM) at pH 3. Fig. 4

reports the effect of Fe^{2+} concentrations on the decolorization of NBB by the Fenton process. From this figure, it is clearly shown that the decolorization efficiency attains 82%, 96%, 97% and 98% in 10 min of reaction time when the concentration of Fe^{2+} was 0.05, 0.1, 0.2 and 0.3 mM, respectively. It is obvious from the results that with the increase of Fe^{2+} concentration, the degradation rate also increases. The enhancement in decolorization efficiency is due to the fact that with an increase of Fe^{2+} , more hydroxyl HO^\bullet radicals are produced according to Eq. (1) [29].

In the present work, we select a catalytic concentration of 0.05 mM of Fe^{2+} for subsequent experiments. In literature, there was no optimal ratio of $[\text{H}_2\text{O}_2]/$

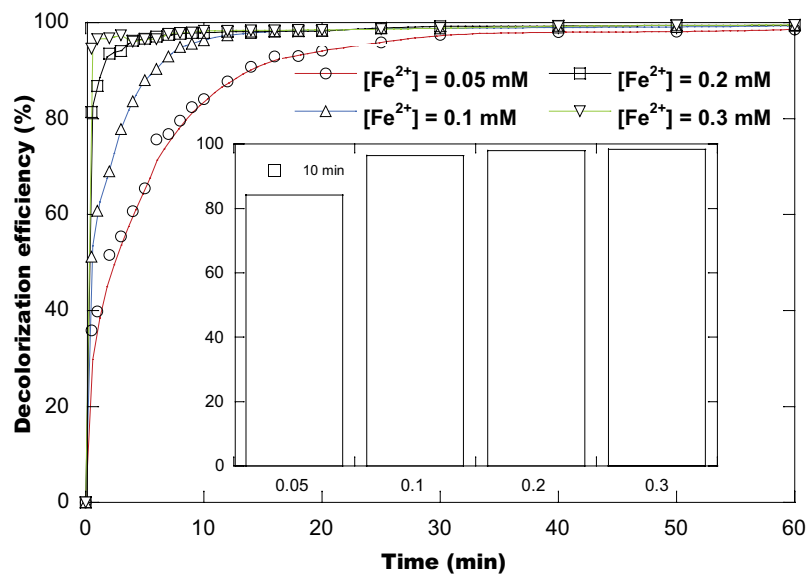


Fig. 4. Effect of initial Fe^{2+} concentration on the decolorization of NBB by Fenton process. Experimental conditions: $[\text{NBB}]_0 = 4.05 \times 10^{-2}$ mM; pH 3; $T = 25^\circ\text{C}$; $[\text{H}_2\text{O}_2]_0 = 2 \times 10^{-3}$ mM; $[\text{Fe}^{2+}]_0 = 0.05\text{--}0.3$ mM.

Fe^{2+}] for the degradation of pollutants by the Fenton process.

3.3. Effect of H_2O_2 concentration

The oxidation in the Fenton process is carried out by HO^\bullet radicals, which are directly produced from the decomposition of hydrogen peroxide [26,29]. In order to determine the concentration of H_2O_2 leading to maximum decolorization efficiency, a series of experiments are performed with the same concentrations of dye (4.05×10^{-2} mM NBB) and Fe^{2+} (0.05 mM), and different concentrations of H_2O_2 (10^{-3} ; 1.5×10^{-3} ; 2×10^{-3} and 3×10^{-3} mM). The obtained results are presented in Fig. 5. It is obvious from this figure that the percentage of dye degradation increases with the increase of H_2O_2 concentration at constant initial concentrations of dye and Fe^{2+} .

The increase in the initial concentration of H_2O_2 causes augmentation in the oxidizing power of the Fenton process as a result of the high production of hydroxyl radicals. The degradation rate of NBB increased from 95% to 99% when the initial concentration of H_2O_2 increased from 10^{-3} to 2×10^{-3} mM. At 3×10^{-3} mM of H_2O_2 , the percentage of degradation was reduced to 96%. The decrease in degradation may be due to the reaction of reactive hydroxyl radicals and H_2O_2 to form less reactive $^\bullet\text{OOH}$ radicals or scavenging of hydroxyl radicals by H_2O_2 [30]. It may be due to the capture of radicals HO^\bullet according to Eqs. (2)–(4).

3.4. Effect of initial NBB concentration

Dye concentrations in industrial effluents vary with the seasons and modes. It is necessary to study the oxidation kinetics of dyestuffs in a fairly wide range of dye concentrations to better understand their effect. The study of the effect of dye concentration on decolorization kinetics was achieved by preparing four solutions of NBB

(1.62 – 8.11×10^{-2} mM) with the same initial concentration of H_2O_2 and Fe^{2+} . Fig. 6 shows the percentage of NBB degradation at different dye concentrations (1.62 – 8.11×10^{-2} mM) during 60 min of reaction time. It is clear from this figure that increasing the initial dye concentration from 1.62×10^{-2} to 4.86×10^{-2} mM results in an increase in the NBB decolorization percentage from 68% to 98%, respectively, during the first 10 min. At low substrate concentrations, the removal efficiency is better than at a higher concentration (8.11×10^{-2} mM). Prior, it should be known that when the initial concentration of the dye increases, the concentration of hydroxyl radicals remains constant for all dye molecules and therefore, the efficiency of the degradation decreases [31,32].

3.5. Effect of temperature

The effect of the temperature on the Fenton process was studied in the temperature range of 25°C – 55°C . The obtained results are presented in Fig. 7 show that temperature has a positive effect on the decolorization efficiency.

The degradation efficiency for first 5 min increased from 63.3% to 99.3%, 99.8% and 99.9% for as rinsing temperature from 25°C to 35°C , 45°C and 55°C , respectively. It can also be noted that the decolorization time for high temperatures is much shorter compared to low temperatures. The temperature has a positive effect on the degradation efficiency of NBB. Indeed, it is known that the increase in temperature increases the number of collisions between molecules and consequently leads to an increase in oxidation speed [33]. In addition, the increase in the percentage of decolorization can also be explained by the improved generation of HO^\bullet radical and their impact on NBB particles. This result is in good agreement with what was observed in the study of the degradation of acid yellow 17 by the Fenton oxidation process [12]. A similar evolution was also observed with the degradation of the acid dye fuchsin by $\text{HP}_{25}\text{W}_{15}\text{Mo}_3\text{Co}_{2.5}\text{O}_{62} \cdot 20\text{H}_2\text{O}/\text{H}_2\text{O}_2$ [34].

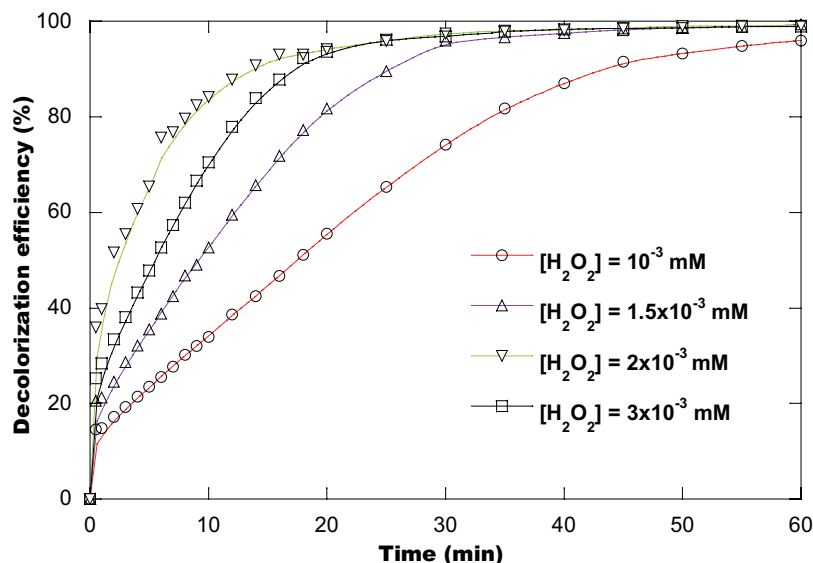


Fig. 5. Effect of initial H_2O_2 concentration on the decolorization of NBB by Fenton process. Experimental conditions: $[\text{NBB}]_0 = 4.05 \times 10^{-2}$ mM; pH 3; $[\text{Fe}^{2+}]_0 = 0.05$ mM; $T = 25^\circ\text{C}$; $[\text{H}_2\text{O}_2]_0 = 1$ – 3×10^{-3} mM.

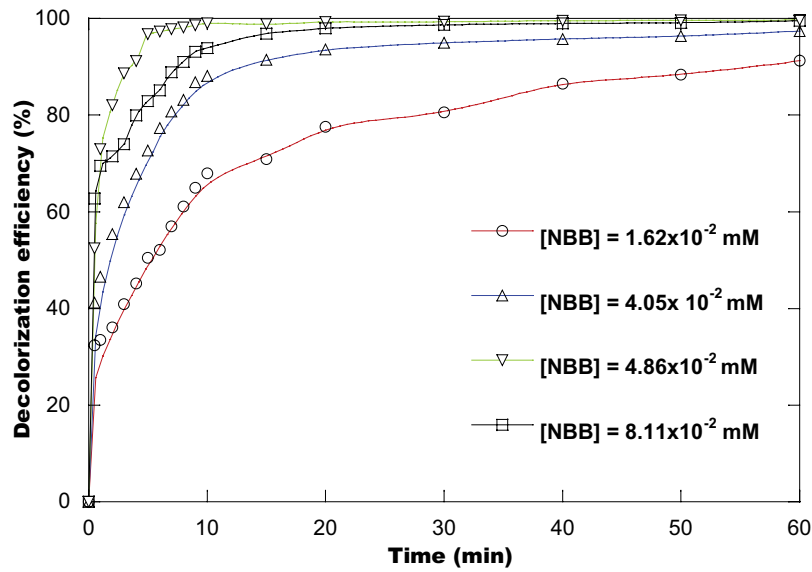


Fig. 6. Effect of initial NBB concentration on its decolorization by Fenton process. Experimental conditions: $[Fe^{2+}]_0 = 0.05 \text{ mM}$; $[H_2O_2]_0 = 2 \times 10^{-3} \text{ mM}$; pH 3; $T = 25^\circ\text{C}$; $[NBB]_0 = 1.62\text{--}8.11 \times 10^{-2} \text{ mM}$.

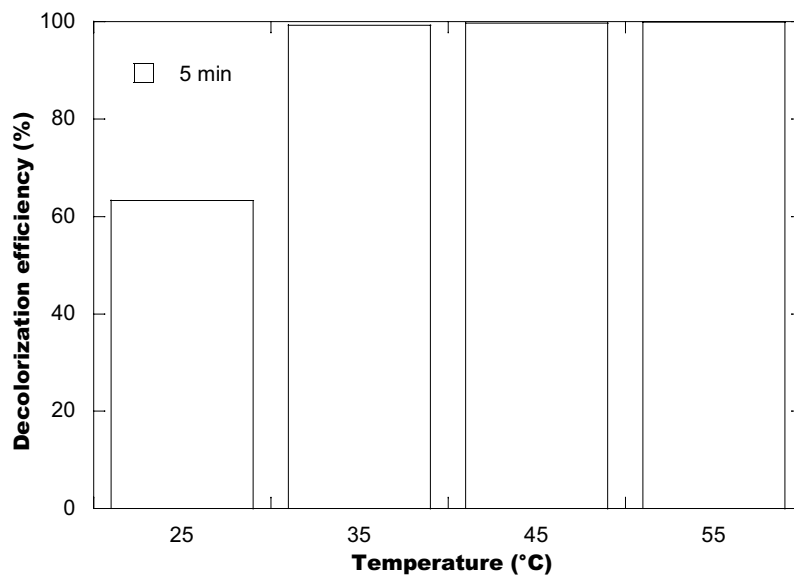


Fig. 7. Decolorization efficiency of NBB during Fenton process as a function of different temperatures. Experimental conditions: $[Fe^{2+}]_0 = 0.05 \text{ mM}$; $[H_2O_2]_0 = 2 \times 10^{-3} \text{ mM}$; pH 3; $[NBB]_0 = 4.05 \times 10^{-2} \text{ mM}$; $T = 25^\circ\text{C}\text{--}55^\circ\text{C}$.

3.6. Effect of chloride ions

In the textile industry, a huge amount of salts is used in coloring processes. These salts may affect the degradation process. In the present study, the effect of chloride ions on the degradation of NBB by the Fenton process was investigated. The experiments were carried out at pH 3 and for 60 min of reaction time. From Fig. 8 it can be seen that the decolorization efficiency decreased from 99.8% to 60.51% after 60 min of reaction time when the Cl^- concentration increases from 0 to 17.1 mM, respectively. The decrease in the rate of dye decolorization is due to the Cl^- ions, which

form complexes with an iron according to the following reactions (8)–(10):



In addition, the decrease in decolorization efficiency can be explained by the presence of Cl^- ions that capture the radicals HO^\bullet , according to the following reaction [34].

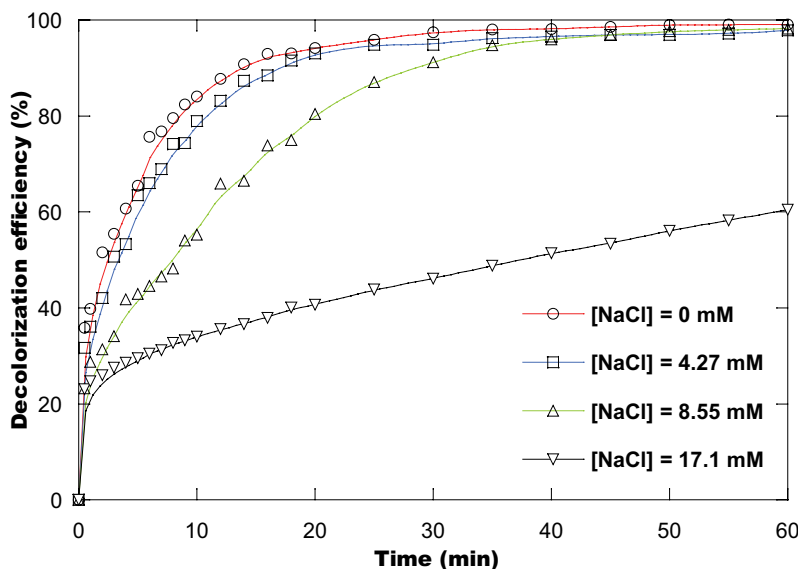
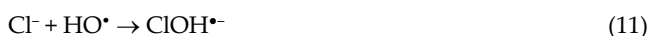


Fig. 8. Effect chloride (Cl^-) ion on the decolorization of NBB by Fenton process. Experimental conditions: $[\text{Fe}^{2+}] = 0.05 \text{ mM}$; $[\text{H}_2\text{O}_2]_0 = 2 \times 10^{-3} \text{ mM}$; $\text{pH } 3$; $[\text{NBB}]_0 = 4.05 \times 10^{-2} \text{ mM}$; $[\text{NaCl}]_0 = 0\text{--}17.1 \text{ mM}$; $T = 25^\circ\text{C}$.



Similar drop has been reported by the addition of sodium chloride in UV/ H_2O_2 [36] and UV/ TiO_2 processes [37]. The aggregation stability of dye molecules in water is influenced in particular by the concentration of dyes and salts. It has been concluded that in the presence of the last one, the electronic repulsion between two molecules of ionic dyes is low compared to the affinity of the hydrophobic association. The addition of sodium chloride may reduce the electrostatic repulsion forces between the anions of the dye, which will lead to an increased tendency for aggregation of the dye species [38].

3.7. Effect of complex matrices

It is very interesting to evaluate the effect of inorganic compounds and organic species present in natural mineral waters and seawater on the degradation of pollutants by the Fenton process. To examine the applicability of this technique to water treatment, the dye ($4.05 \times 10^{-2} \text{ mM}$) was dissolved in natural mineral water and seawater. The main characteristics of this natural mineral water are: $\text{pH } 7$, $\text{K}^+ = 1 \text{ mg L}^{-1}$, $\text{Ca}^{2+} = 30 \text{ mg L}^{-1}$, $\text{Mg}^{2+} = 9.1 \text{ mg L}^{-1}$, $\text{Na}^+ = 11 \text{ mg L}^{-1}$, $\text{Cl}^- = 28.4 \text{ mg L}^{-1}$, $\text{SO}_4^{2-} = 11 \text{ mg L}^{-1}$; $\text{HCO}_3^- = 60 \text{ mg L}^{-1}$, total dissolved solids (TDS) = 152 mg L^{-1} .

The obtained results (Fig. 9) show that the percentage of NBB decolorization in distilled water and natural mineral water is higher than that obtained in seawater. The degradation of NBB in distilled water and natural mineral water was not affected during treatment and the destruction of the dye was complete, opposite to what was observed in seawater. It seems that the excess of salt in the seawater is an obstacle to the removal of the dye by the Fenton process because the presence of chloride ions in a solution containing ferrous ions leads to the formation of complexes

and captures hydroxyl radicals [39]. A similar trend was observed with the degradation of acid red 66 and direct blue 71 from aqueous solution by the Fenton process [40].

3.8. Comparison between Fenton and sono-Fenton processes

The degradation of NBB was carried out under different experimental conditions, which are ultrasound alone (US), Fenton alone ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$) and a combination of ultrasound and Fenton process (US/ $\text{H}_2\text{O}_2/\text{Fe}^{2+}$). The obtained results are shown in Fig. 10. At 40 kHz of ultrasonic irradiation as the only process, NBB is almost unaffected, only 2% degradation of the dye was achieved after 1 h of sonication alone under 30 W of ultrasonic power. The reason for this low rate of sonochemical degradation can be explained by the low production of the HO^\bullet radicals produced under low-frequency ultrasound. In general, the research literature reports that low-frequency ultrasound generally causes physical effects, whereas high-frequency ultrasound can produce active compounds such as free hydroxyl radicals that can oxidize organic substances [41,42].

The coupling of the Fenton process with low-frequency ultrasound (US/ $\text{H}_2\text{O}_2/\text{Fe}^{2+}$) improves significantly the degradation of the dye, obtaining its total destruction after only 10 min of treatment. The reaction time is considerably reduced compared to the Fenton process alone. This can be due to the ultrasonic irradiation, which considerably increases the rate of H_2O_2 production. This enhancement is explained by the increased mass transfer and the additional generation of hydroxyl radicals that can react with the substrate [43–45].

4. Conclusion

This work was focused on the decolorization of an azo dye, namely NBB, by Fenton and sono-Fenton treatments.

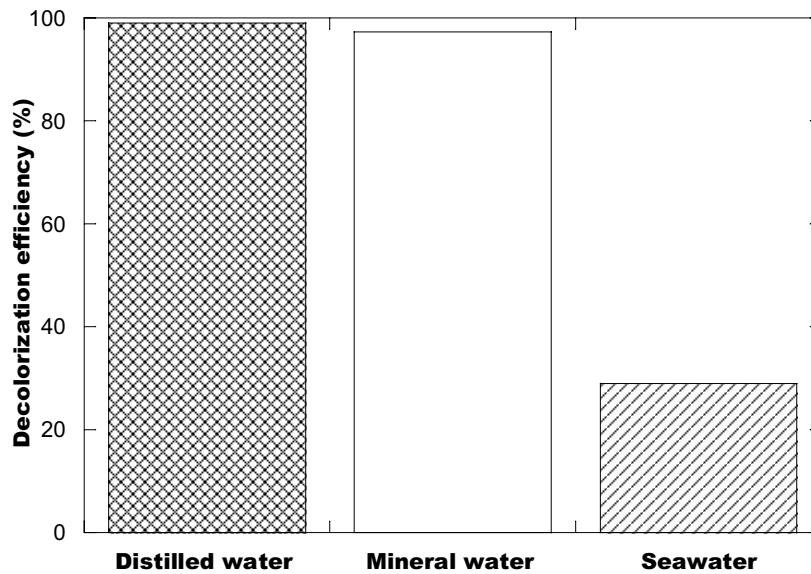


Fig. 9. Decolorization efficiency of NBB during Fenton oxidation treatment in distilled water, natural mineral water and seawater. Experimental conditions: $[Fe^{2+}] = 0.05 \text{ mM}$; $[H_2O_2]_0 = 2 \times 10^{-3} \text{ mM}$; pH 3; $[NBB]_0 = 4.05 \times 10^{-2} \text{ mM}$; $T = 25^\circ\text{C}$.

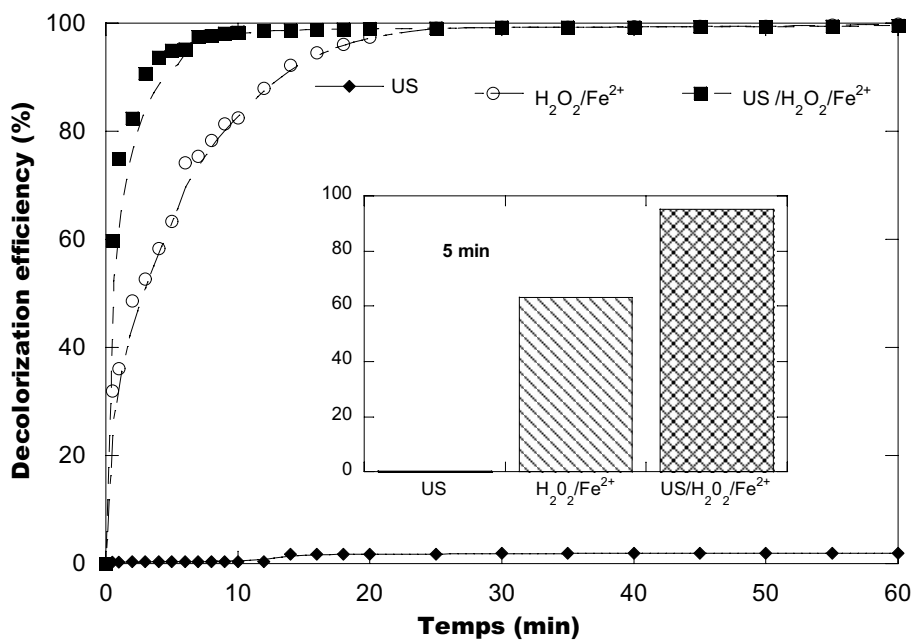


Fig. 10. Decolorization of NBB by ultrasound, Fenton process and sono-Fenton process. Experimental conditions: $[Fe^{2+}] = 0.05 \text{ mM}$; $[H_2O_2]_0 = 2 \times 10^{-3} \text{ mM}$; pH 3; $[NBB]_0 = 4.05 \times 10^{-2} \text{ mM}$; $T = 25^\circ\text{C}$.

The experimental results show that the degradation efficiency of the dye depends on many parameters. In order to get the optimal kinetics as well as the higher efficiency, we studied the influence of the various parameters. The pH is a very important parameter; we have noticed that the favorable pH for the decolorization of NBB by the Fenton process is 3, with an efficiency of 99% after 60 min. The effect of Fe^{2+} concentration on dye degradation was examined. It results from this study that the best decolorization efficiency

(99%) was obtained for 0.05 mM of Fe^{2+} and 2×10^{-3} mM of H_2O_2 after a contact time of 60 min. The degradation of the dye increases with an increasing temperature between 25°C and 55°C . The initial concentration of the dye was studied. The results show that the reaction is favored at low initial dye concentration. A decrease in the decolorization efficiency is observed due to the effect of the chloride ion, which captures the HO^\bullet radicals. The combination of US/ Fe^{2+}/H_2O_2 gave a better decolorization efficiency compared

to $\text{Fe}^{2+}/\text{H}_2\text{O}_2$. In regard to these results, the sono-Fenton process is the best for NBB oxidation since the reaction time has been reduced as well as the chemical reagents.

In this study, some results have been obtained, some mechanisms have been explained, but the complexity of the subject leaves the field open for further work in certain areas: comparing the sono-Fenton process by photo-Fenton and electro-Fenton proses.

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