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Photodegradation Kinetics and Color Removal of 2-(4-hydroxyphenylazo) Benzoic Acid by Advanced Oxidation Processes

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Authors' contributions

This work was carried out in collaboration between both authors. Author YK designed the study, wrote the protocol and wrote the first draft of the manuscript. She also managed the analyses of the study and the literature searches. Authors YK and EB performed the statistical analyses. Both authors read and approved the final manuscript.

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ABSTRACT

Degradation kinetics and color removal of 2-(4-hydroxyphenylazo)benzoic acid azo dye by the hydrogen peroxide/ultraviolet irradiation and sodium hypochlorite/ultraviolet irradiation processes were carried out in Spectroline CM-10A Model ENF-260C/FE photoreactor which has ultraviolet lamps irradiating at 254 and 365 nm wavelengths. All experimental studies were performed at room temperature in 0.04 M Britton Robinson buffer (pH 2-12). For this purpose, the degradation kinetics of 2-(4-hydroxyphenylazo)benzoic acid was investigated depending on pH, initial dye concentration and hydrogen peroxide concentration. Optimum pH and hydrogen peroxide concentration were determined as pH 10 and 3.57×10^{-2} M, respectively, for 3.5×10^{-5} M 2-(4-hydroxyphenylazo)benzoic acid. Optimum pH value in 1.55×10^{-3} M sodium hypochlorite medium has been found as pH 8 for 3.5×10^{-5} M azo dye. These methods used for degradation of dye are compared, it is concluded that sodium hypochlorite/ultraviolet irradiation and sodium hypochlorite methods are more effective than hydrogen peroxide/ultraviolet irradiation and hydrogen peroxide methods because of color removal of 83% and 64%, respectively.

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1. INTRODUCTION

Aromatic azo compounds which are characterized by one or more azo groups form a large and significant class of organic compounds due to their extensive applications as coloring agents in industrial processes involving textile dyeing, plastics, drugs, cosmetics and foods [1-4]. Large quantities of the azo dye effluents and other toxic chemical compounds are produced as environmental contaminants in the dyeing process of textile industries, by discharging of them into the rivers. Sensitive analytical methods are essential for the determination of these carcinogenic pollutants [5-8].

Textile wastewater generally contains a significant amount of dissolved dyes, the majority of which are composed of azo dyes. Advanced oxidation methods offer a promising technology in the removal of dyestuff impurities in wastewater due to their high color removal efficiency [9-11]. In many cases, effluents of these industries involve little or no biodegradable organic compounds. Therefore, the pollutant loads in terms of biological oxygen demand (BOD) may be negligible and chemical oxygen demand (COD) would be higher than BOD [12]. For medicinal wastewater treatment, several methods were proposed, including anaerobic-aerobic or membrane bioreactors, electrochemical oxidation, biochemical combined method and advanced oxidation processes (AOPs) [12]. Decomposition and destruction methods for converting phenolic compounds into safe materials, include the use of oxidizing agents such as ozone, hydrogen peroxide, potassium permanganate and sulfur dioxide. Biological methods, aeration, advanced oxidation and physico-chemical processes have been also found to be efficient methods for the degradation of many organic pollutants from wastewater [13-15].

Aromatic amines yielded as a result of degradation of azo dyes and a majority of the dyes can also absorb light in the ultraviolet (UV) ranges. Consequently, it is possible to monitor the degradation of these organic compounds by measuring the UV absorbance. Each dye has a characteristic absorbance spectrum, a feature that is very beneficial for model systems that is only one dye compound is included [16].

UV-light has been found to be very effective in the degradation of dyes [17]. Hydrogen peroxide/ultraviolet irradiation (H_2O_2/UV) method is able to destroy totally the chromophoric structure of azo dyes depends on the basic structure and the nature of auxiliary groups attached to the aromatic nuclei of the dyes [17-19].

Azo dyes were degraded by sodium hypochlorite (NaOCl) successfully, due to sodium hypochlorite has a vigorous oxidant capacity. It has been reported in the literature that the amount of NaOCl is a very important factor to achieve optimal degradation of the Reactive Brilliant Red (K-2BP) dye. Color removal was investigated using different amounts of NaOCl under the same conditions. As the amount of NaOCl used increased, color removal and chemical oxygen demand (COD) percentages increased for K-2BP dye. This is explained by the fact that the increase in NaOCl content will increase the frequency of collisions between K-2BP and oxidants [20].

In this study, color removal and degradation kinetics of 2-(4-hydroxyphenylazo)benzoic acid (HABA) azo dye have been investigated by using H_2O_2, H_2O_2/UV, NaOCl and NaOCl/UV methods. This dye has not been investigated by these methods up to now. The molecular structure of the azo dye is given in Scheme 1.

2. MATERIALS AND METHODS

2-(4-hydroxyphenylazo)benzoic acid (≥98%) was purchased from Sigma-Aldrich and used without further purification. Phosphoric acid, acetic acid, boric acid and sodium hydroxide used for buffer preparation and H_2O_2 (30-35% w/w Merck) used for oxidant were of purity p.a. UV spectral analysis was performed using Shimadzu UV-2600 spectrophotometer.

Degradation studies with UV light were carried out in Spectroline CM-10A Model ENF-260C/FE photoreactor which has UV lamps irradiating at 254 and 365 nm wavelengths. UV lamp capable of irradiation at 365 nm (6 W) was used for degradation studies. All experimental studies were performed at room temperature in 0.04 M Britton Robinson buffer (pH 2-12).

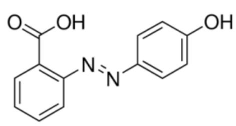
A 10^{-3} M stock solution of HABA was prepared by dissolving appropriate amount of HABA in ethanol.

3. RESULTS AND DISCUSSION

3.1 Effect of pH

In order to investigate the effect of pH on the degradation kinetics of HABA dye, experimental studies were carried out at different pH values (pH 3, 5, 7, 8, 10 and 11) in 0.04 M BR buffer and $3.57x10^{-2}$ M H_2O_2 media. At certain intervals, 5 mL samples were taken from 100 mL solution of $3.5x10^{-5}$ M HABA and absorbance values were measured. C/C_o changes of HABA as a function of time (t) are given in Fig. 1. As can be seen in Fig. 1, C/C_o values of HABA have not changed in the presence of H_2O_2 in six hours.

H_2O_2 exhibited a color removal of 12% after 6.2 hour at a pH of 10. The desired amount of color removal was not obtained from the dye in six hours in the presence of H_2O_2 in studied pH values. Since the photodegradation rate of $3.5x10^{-5}$ M HABA at pH 10 was higher than other pHs (pH 3, 5, 7, 8 and 11.0), it was preferred as the optimum pH in BR buffer and $3.57x10^{-2}$ M H_2O_2 media.



Scheme 1. Molecular structure of the 2-(4-hydroxyphenylazo)benzoic acid

3.2 Effect of Initial Dye Concentration

In order to evaluate the influence of initial dye concentration on the azo dye degradation rate, different concentrations of HABA from $0.5x10^{-5}$ to $3.5x10^{-5}$ M were prepared at fixed $3.57x10^{-2}$ M H_2O_2 concentration in 0.04 M BR buffer (pH 10) medium. As the initial dye concentration increases, the degradation efficiency decreases as well. The higher the initial dye concentration, the lower penetration of UV radiation into the solution, and as a result, hydroxyl radical concentration diminishes [17].

In this present work, $3.5x10^{-5}$ M concentration of HABA was selected at fixed hydrogen peroxide concentration in accordance with the literature [21].

3.3 Degradation of HABA in H_2O_2 and H_2O_2/UV Media

The degradation process of the dye was initiated by a 100 mL solution containing $3.5x10^{-5}$ M HABA and $3.57x10^{-2}$ M H_2O_2 in 0.04 M BR buffer (pH 10) medium. At this time, the solution was continuously stirred homogeneously using a magnetic stirrer. At certain intervals, 5 mL samples were taken from 100 mL solution of HABA and absorbance values were measured.

Under the same experimental conditions, 100 mL of solution was homogeneously mixed and irradiated. Then 5 mL samples were taken from this solution at certain time intervals and absorbance values were measured. Absorption spectra of degradation of the $3.5x10^{-5}$ M HABA in 0.04 M BR buffer (pH 10) and H_2O_2/UV media were given in Fig. 2. When UV-Vis spectrum of the degradation of $3.5x10^{-5}$ M HABA in H_2O_2/UV medium was examined, $\pi \rightarrow \pi^*$ electronic transition of –N=N– azo group was observed at 399 nm, whereas at 262 nm, $\pi \rightarrow \pi^*$ transition was observed due to phenyl groups [22].

In H_2O_2 method, approximately 12% color removal was achieved after 373 minutes. There was not much change in the absorbance of HABA for 344 minutes. When the absorbance change of the aromatic ring was examined, it was observed that its absorbance remained approximately constant.

In the presence of $3.57x10^{-2}$ M H_2O_2, the HABA solution was irradiated with continuous stirring for two hours. In the H_2O_2/UV method, 16% color removal was achieved after the first 76 minutes (Fig. 3).

When H_2O_2 and H_2O_2/UV methods were compared each other, it can be said that color removal was not achieved as desired amounts in two methods due to the stability of HABA. However, it is seen that H_2O_2/UV method is more effective than the H_2O_2 method because of providing 16% color removal in a shorter time (Fig. 3).

The exponential decrease of (C/C_o) values of HABA with time can be described by Equation (1).

$$\ln(\frac{C}{C_o}) = -kt \qquad (1)$$

where C_o is the initial dye concentration, C is the concentration at any time t and k is the rate constant. The logarithmic plot of normalized dye concentration as a function of time gives straight lines (Fig. 4). The degradation rate constants have been determined by using Equation (1). The linear variations of $\ln(C_o/C)$ against time for

HABA solutions in 0.04 M BR buffer (pH 10) for different concentrations of H_2O_2 were given in Fig. 4. Equations and k (min^{-1}) values of ln (C_o/C)-t lines of HABA were also given in Table 1. Similar results were observed in previous studies [8,21,23].

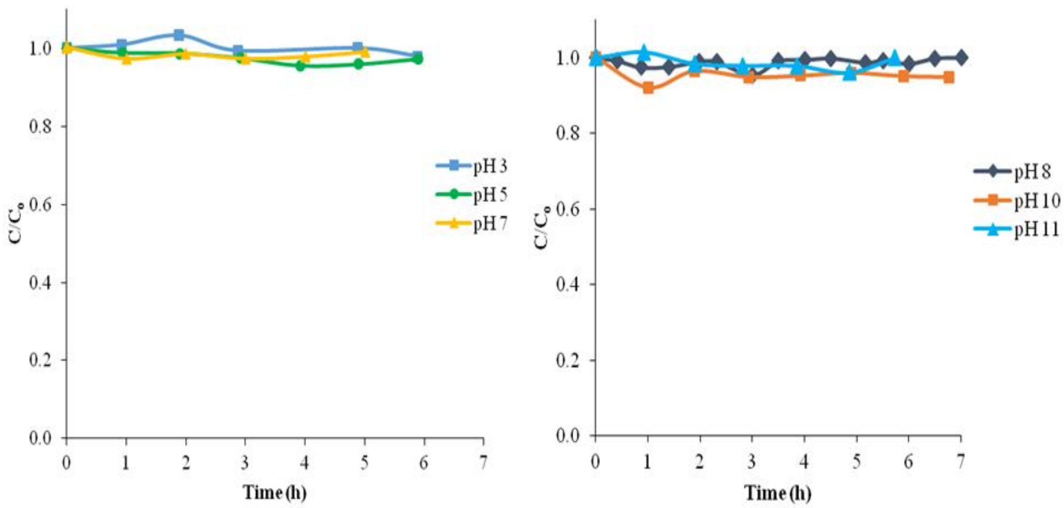


Fig. 1. C/C_o values versus degradation time of $3.5x10^{-5}$ M HABA obtained in $3.57x10^{-2}$ M H_2O_2 medium at different pHs

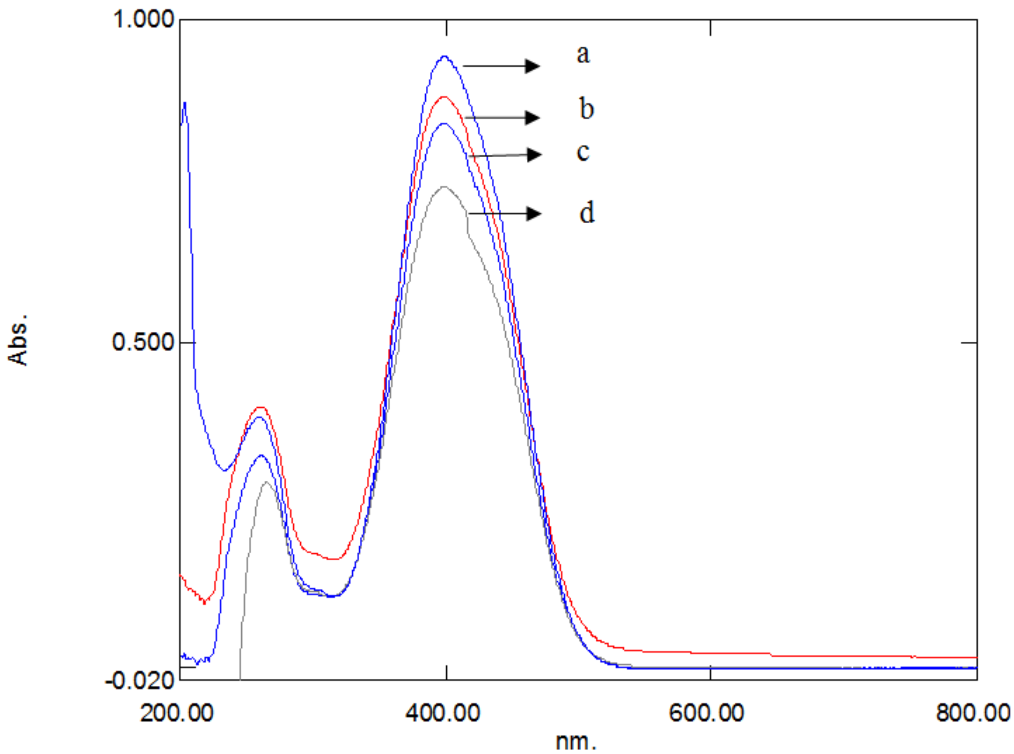


Fig. 2. Absorption spectra of degradation of the $3.5x10^{-5}$ M HABA in 0.04 M BR buffer (pH 10) and $3.57x10^{-2}$ M H_2O_2/UV media a) in the absence of H_2O_2 b) 0 c) 28 d) 76 minutes after addition of H_2O_2 and in the presence of irradiation

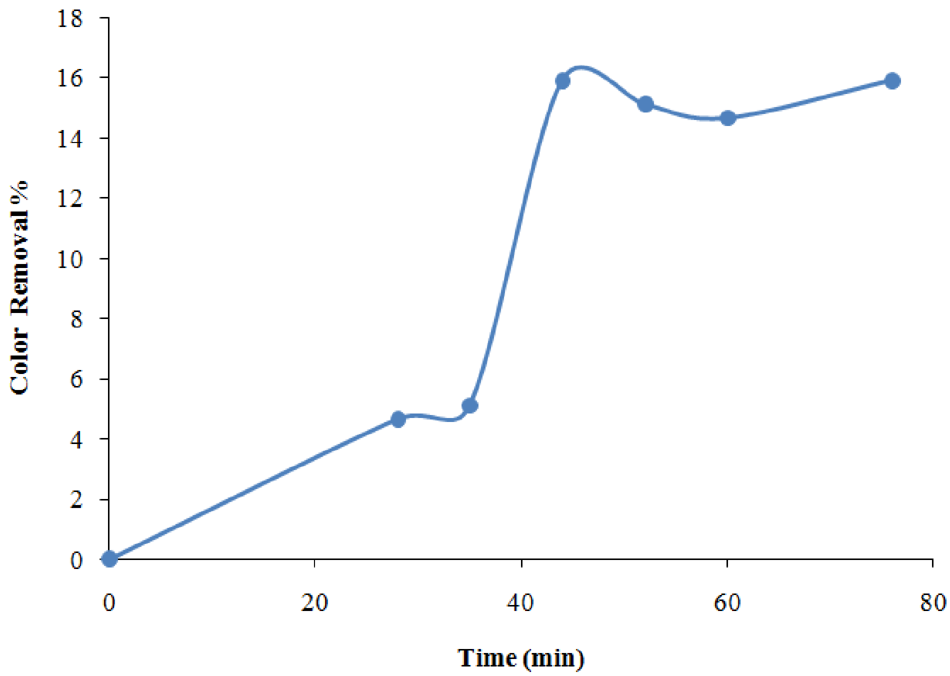


Fig. 3. Color removal of HABA (3.5x10^{-5} M) as a function of time in 0.04 M BR (pH 10) buffer and H_2O_2 (3.57x10^{-2} M) media in the presence of irradiation

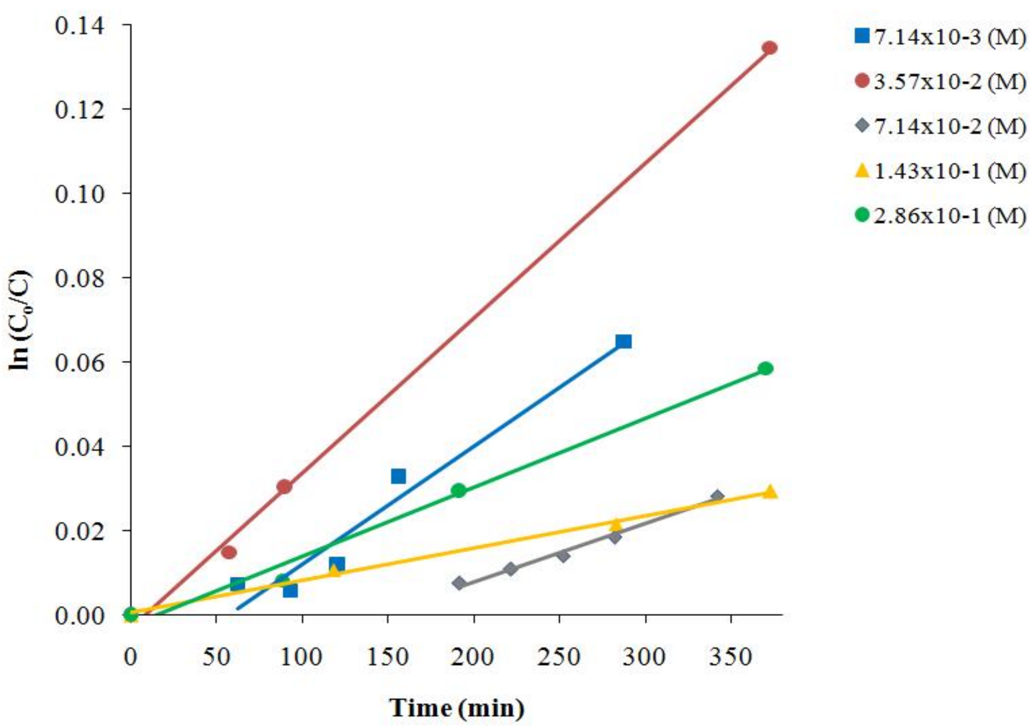


Fig. 4. The linear variations of ln(C_o/C) versus time belonging to the degradation of 3.5x10^{-5} M HABA solutions in 0.04 M BR buffer (pH 10) for different concentrations of H_2O_2

Table 1. Equations and rate constant (k) values of ln (C_o/C)-t lines obtained for 3.5x10^{-5} M HABA in 0.04 M BR buffer at different H_2O_2 concentrations

H_2O_2 concentration	Equation	r^2 (Correlation coefficient)	k (min^{-1})
7.14x10^{-3} (M)	$ln(C_o/C) = -0.0156 + 2.79x10^{-4}t$	0.9539	2.79x10^{-4}
3.57x10^{-2} (M)	$ln(C_o/C) = -0.0029 + 3.67x10^{-4}t$	0.9982	3.67x10^{-4}
7.14x10^{-2} (M)	$ln(C_o/C) = -0.0200 + 1.39x10^{-4}t$	0.9889	1.39x10^{-4}
1.43x10^{-1} (M)	$ln(C_o/C) = 0.0006 + 7.65x10^{-5}t$	0.9958	7.65x10^{-5}
2.86x10^{-1} (M)	$ln(C_o/C) = -0.0025 + 1.63x10^{-4}t$	0.9905	1.63x10^{-4}

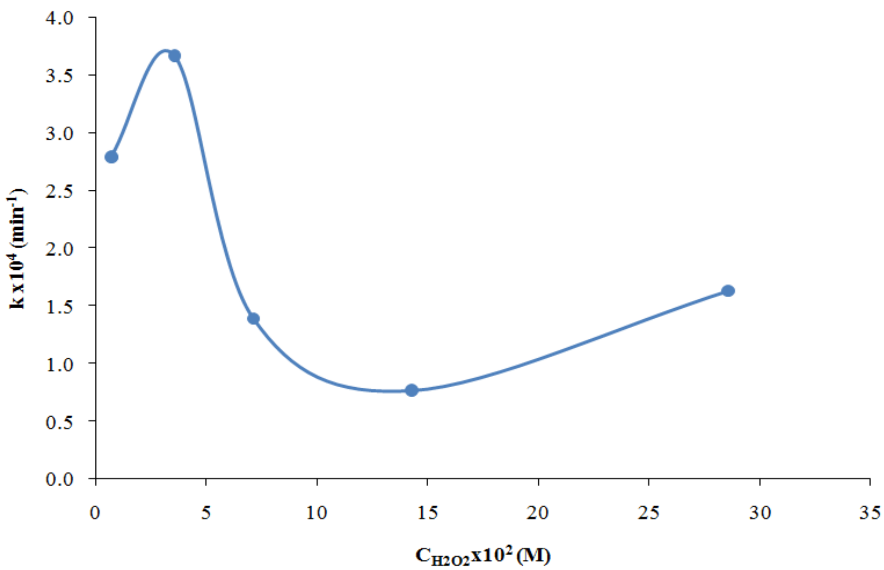


Fig. 5. Variation of rate constant values of 3.5x10^{-5} M HABA with increasing H_2O_2 concentrations in 0.04 M BR (pH 10) buffer

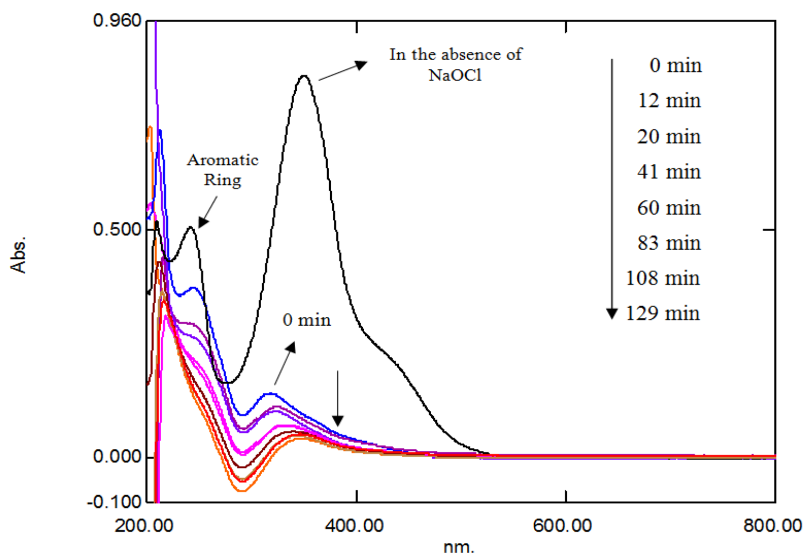


Fig. 6. Absorbance changes of 3.5x10^{-5} M HABA obtained at certain time intervals in BR buffer (pH 8) by NaOCl (1.55x10^{-3} M) process

H_2O_2 concentration with the highest rate constant value was determined as $3.57x10^{-2}$ M for $3.5x10^{-5}$ M HABA (Fig. 5). The rate constant at an optimum H_2O_2 concentration was determined to be $3.67x10^{-4}$ min^{-1}.

The concentration of hydrogen peroxide is an important parameter affecting the efficiency of the color removal of the dye by H_2O_2/UV treatment. Some authors have reported that the peroxide concentration may either increase the photoreaction rate or inhibit it due to the scavenging action of peroxide depending on the concentration. Therefore, the optimum concentration of peroxide in the reaction should be provided [17,19,24]. As the initial concentration of H_2O_2 increases, the color removal rate of the dye increases due to the increase of •OH radicals in the solution [25,26]. However, at a certain H_2O_2 concentration, the •OH free radicals will reach equilibrium with H_2O_2. Increasing H_2O_2 concentration after this point will not increase the concentration of •OH free radicals in the solution. As seen in Fig. 5, degradation rate of HABA increases until the hydrogen peroxide concentration increases to $3.57x10^{-2}$ M. After this point, the reaction rate of dye decreases and it remains approximately constant as reaches to $10x10^{-2}$ M H_2O_2 concentration value. Increasing the H_2O_2 concentration promotes the reaction of the formed hydroxyl radicals with the excess H_2O_2 to produce hydroperoxyl radicals,which have less oxidizing capacity [24,27,28].

3.4 Degradation of HABA in NaOCl and NaOCl/UV Media

The $3.5x10^{-5}$ M HABA solution was prepared in 0.04 M BR (pH 8) buffer. Commercial bleach with a NaOCl concentration of $1.15x10^{-2}$ M was added to the prepared solution. Then, this 50 mL solution with a concentration of $1.55x10^{-3}$ M NaOCl was stirred continuously in a magnetic stirrer and 5 mL samples were taken at certain time intervals and absorbance values were measured. The absorbance changes obtained in NaOCl and NaOCl/UV media were given in Fig. 6 and Fig. 7, respectively. When the absorbance changes of HABA given in Fig. 6 were examined, it was seen that the major absorbance band of HABA at 350.5 nm decreased with time after adding of NaOCl. And it was also observed that the absorbance band of HABA at 241.5 nm (phenyl ring) decreased with time after adding of NaOCl and disappeared after 41 minutes.

It was observed that the major absorbance band of HABA at 352 nm decreased with time after adding of NaOCl and in the presence of irradiation (Fig. 7). And the absorbance band of phenyl ring of HABA at 241.5 nm also decreased with time in NaOCl/UV medium and disappeared after 18 minutes of degradation.

Color removal (%) and C/C_o values of HABA by NaOCl and NaOCl/UV processes were also given in Fig. 8. Oxidation with $1.55x10^{-3}$ M NaOCl produced a color removal of 64% after 129 minutes and NaOCl/UV produced a color removal of 83% after 127 minutes at a pH of 8 (Fig. 8). Color removal % values of HABA as a function of time for different processes were given in Table 2. Equations and k (min^{-1}) values of ln (C_o/C)-t lines obtained for $3.5x10^{-5}$ M HABA in 0.04 M BR buffer (pH 8) by NaOCl and NaOCl/UV processes were given in Table 3.

The fact that the decolorization rate constant of HABA in NaOCl/UV medium ($k = 1.30x10^{-2}$ min^{-1}) is greater than the rate constant in NaOCl medium ($k = 9.80x10^{-3}$ min^{-1}) indicates that there is a faster color removal in NaOCl/UV process compared to the NaOCl process (Fig. 8) [29,30].

Table 2. Performance of different processes in degradation of HABA as a function of time

Process	Color removal %	Time (h)
H_2O_2	12%	6.2
H_2O_2/UV	16%	1.3
NaOCl	64%	2.2
NaOCl/UV	83%	2.1

Table 3. Equations and k values of ln (Co/C)-t lines obtained for 3.5x10-5 M HABA in 0.04 M BR buffer (pH 8) by NaOCl ($1.55x10^{-3}$ M) and NaOCl ($1.55x10^{-3}$ M)/UV processes

Method	Equation	r^2	k (min^{-1})
NaOCl/UV	$\ln(C_o/C) = 0.2871 + 1.30x10^{-2}t$	0.9093	$1.30x10^{-2}$
NaOCl	$\ln(C_o/C) = 0.0883 + 9.80x10^{-3}t$	0.9847	$9.80x10^{-3}$

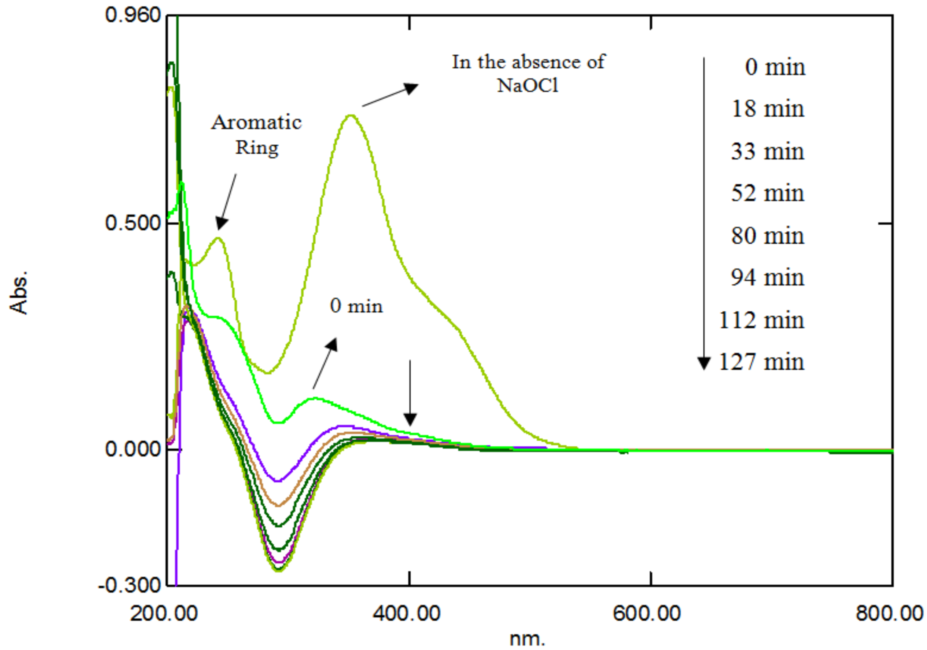


Fig. 7. Absorbance changes of 3.5×10^{-5} M HABA obtained at certain time intervals in BR buffer (pH 8) by NaOCl (1.55×10^{-3} M)/UV process

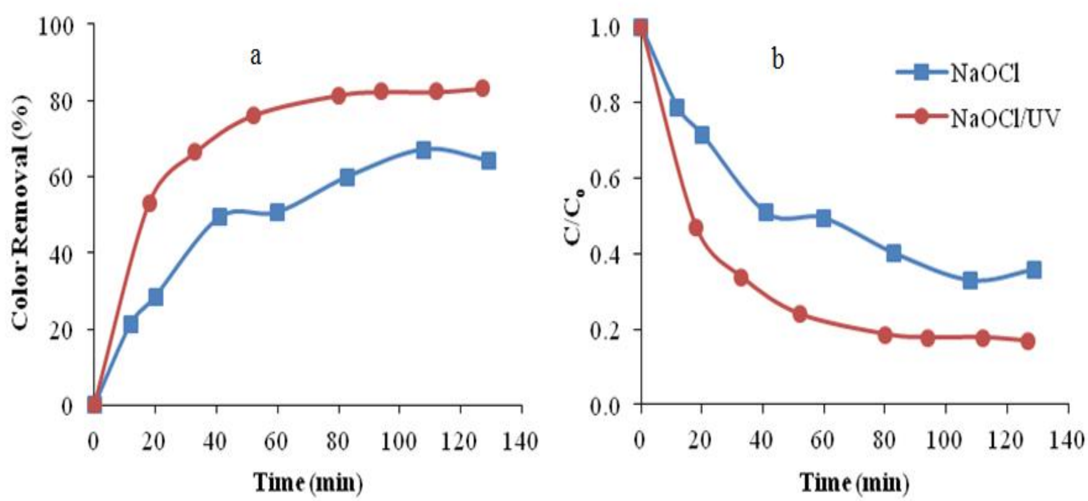


Fig. 8. a) Color removal b) C/C_o values of HABA (3.5×10^{-5} M) as a function of time in 0.04 M BR (pH 8) buffer by NaOCl (1.55×10^{-3} M) and NaOCl (1.55×10^{-3} M)/UV processes

4. CONCLUSIONS

In this study, H_2O_2, H_2O_2/UV, NaOCl and NaOCl/UV processes were used for color removal of HABA dye. The results of this work showed that using UV irradiation in the presence of H_2O_2, led to 16% degradation of the HABA in a time of 1.3 hours while in H_2O_2 process, 12% removal of color removal was obtained within 6.2 hours. Color removal of the HABA compound has not been achieved as desired amounts, by H_2O_2 and H_2O_2/UV methods. These methods used for degradation of dye are compared, it is concluded that NaOCl/UV and NaOCl methods are more effective than H_2O_2/UV and H_2O_2 methods because of color removal of 83% and 64% in two hours, respectively.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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