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Kinetics of decolorization of an azo dye in UV alone and UV/H₂O₂ processes

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Abstract

The decolorization of C.I. Acid Red 27 (AR27), a monoazo anionic dye, was studied in the ultraviolet radiation (UV) alone and UV plus hydrogen peroxide (UV/H₂O₂) processes. The experimental results indicated that the kinetics of both oxidation processes fit well by pseudo-first order kinetics. The reaction rate was sensitive to the operational parameters and increased with increasing H₂O₂ concentration and light intensity. The reaction orders of H₂O₂ concentration and light intensity in both processes were obtained with linear regression method. A regression model was developed for pseudo-first order rate constant ($k_{ap,UV/H_2O_2}$) as a function of the H₂O₂ concentration and UV light intensity.

 $k_{\rm ap,UV/H_2O_2} = (2 \times 10^{-4} I_0^{0.75} + k_3 I_0^{1.38} [H_2O_2]_0^n) \varphi_{\rm AR27}$

As a result of two opposing effects of H_2O_2 concentration at low and high concentrations, *n* has a value of 0.49 and -0.39 and k_3 has a value of 3×10^{-4} and 0.1 for the regions of $0 \text{ mg } l^{-1} < [H_2O_2]_0 < 650 \text{ mg } l^{-1}$ and $650 \text{ mg } l^{-1} < [H_2O_2]_0 < 1500 \text{ mg } l^{-1}$, respectively. φ_{AR27} is the initial dye concentration correlation index for developing of model for different initial concentrations of AR27. This rate expression can be used for predicting $k_{ap,UV/H_2O_2}$ at different conditions in UV alone and UV/H_2O_2 processes. The results show that UV alone cannot be an efficient method for decolorization of AR27 in comparison with UV/H_2O_2 process, therefore the first term of the model can be neglected. © 2006 Elsevier B.V. All rights reserved.

Keywords: Advanced oxidation processes (AOPs); UV/H2O2; Decolorization; Kinetics; C.I. Acid Red 27

1. Introduction

Azo dyes constitute a major part of all commercial dyes employed in a wide range of processes in the textile, leather, pharmaceutical, plastics, paints and food industries. They are characterized by the presence of the azo group (-N=N-)attached to two substituents, mainly benzene or naphthalene derivatives, containing electron-withdrawing and/or electrondonating groups [1]. The major sources of dyes in the environment are effluents from the textile industry. Textile finishing mills discharge wastewater containing a great variety of organic contaminants in a wide range of concentration. The disposal of these colored wastewaters poses a major problem for the industry as well as a threat to the environment [2]. For removal of such pollutants, physical techniques such as adsorption on activated

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carbon, ultrafiltration, reverse osmosis, coagulation and electrocoagulation [3] can be used efficiently. Nevertheless, they are non-destructive, and merely transport contaminants from water to sludge [3]. Advanced oxidation processes (AOPs) are attractive alternatives to non-destructive physical water treatment processes, because they are able to mineralize organic water contaminants [4,5]. UV radiation is the base of AOPs which is applied to water treatment for the degradation of individual pollutants. Among the AOPs, chemical oxidation using UV in the presence of H_2O_2 is a very promising technique [3,4,6,7]. AOPs are destructive types of water pollution removal processes because they eliminate compounds rather than transferring them into another medium such as a solid phase. Processes involving the use of UV radiation and H2O2 are characterized by the generation of hydroxyl radicals. The hydroxyl radicals are short-lived and highly reactive chemical species that reacts non-selectively with organic matter present in wastewater. These radicals can oxidize organic compounds producing organic radicals, which are highly reactive and can undergo further oxidation. The possi-

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ble reactions, which occur during UV/H_2O_2 process, are hydrogen abstraction, electrophilic addition and electron transfer reactions [8]. The UV/H_2O_2 technique has several advantages such as no sludge formation in the treatment, operation at ambient temperature and also the oxygen formed in this process is useful for aerobic biological decay process [9]. Furthermore, if the operation is carried out under suitable conditions, the final products are H_2O , CO_2 and low molecular weight aliphatic acids [10].

Various types of kinetic models have been postulated to describe the kinetics of the UV/H₂O₂ process. An overview of these models that they are divided into two sections. The first type of kinetic models for the UV/H₂O₂ process is on the basis of reaction mechanism and known as chemical and photochemical reactions. These kinetic models were developed for many dye compounds such as azo acid dyes [2] and Reactive Black 5 [11]. The second type of these models is an empirical rate expression, which rate constants and reaction orders are determined by dealing the experimental results with linear regression. Zang and Farnood [12], Elkanzi and Kheng [13] and Ku and Ho [14] tested the power law model and obtained reaction orders for methyl tert-buthyl ether, isoprene and chlorophenols, respectively. Malik and Sanyal [15] reported an empirical rate expression for azo dyes in wastewater, which was only related the k_{ap} to initial H₂O₂ concentration in the UV/H₂O₂ process. These literature surveys show that the study on the development of the empirical rate expression for the dye removal by UV only and UV/H2O2 processes and inserting the operational parameters such as H_2O_2 , AR27 concentration and I_0 (light intensity) is scarce. The power law models have been examined for various organic compounds, but for removal of colored organic compounds such models are rare. Malik and Sanyal [15] only reported a power law model for mixture of three azo dyes (Direct Bordeaux BW (C.I. Direct Red 7), Congo Red (C.I. Direct Red 28) and Direct Blue 2B (C.I. Direct Blue 54)). This model was tested in the low concentration of H2O2 and the effects of light intensity and initial concentration of dye compounds were neglected.

The objective of the present study is to establish the empirical reaction rate expressions for the decolorization of AR27, as a model compound from monoazo dyes, by UV radiation alone and combined with H_2O_2 , to obtain reaction orders for H_2O_2 , AR27 and I_0 , and also compare the efficiency of the two oxidation methods in the decolorization of AR27.

2. Materials and methods

2.1. Reagents

AR27, a monoazo anionic dye, was obtained from Boyakh Saz Company (Iran) which was commercial dye and used without further purification. Fig. 1 displays the structure of this dye. The H_2O_2 solution (30%) was purchased from Merck (Germany). Solutions were prepared by dissolving the requisite quantity of the dye in double distilled water.



Fig. 1. Chemical structure of AR27 (C.I. 16185).

2.2. Experiments

All experiments were carried out in a batch photoreactor. The radiation source was a low pressure mercury UV lamp emitting at 254 nm (30 W, UV-C, manufactured by Philips, Holland) which was placed above a batch photoreactor of 0.51 volume [6]. The light intensity was measured by a Lux-UV-IR meter (Leybold Co., GmbH) with UV-C probe at the surface of the solution in the reactor. Variation in the distance between UV lamp and Pyrex reactor caused the change in the light intensity. In each experiment, 200 ml of the dye solution of nearly constant concentration $(30 \text{ mg } 1^{-1})$ and variable concentration of H_2O_2 (from 0 to $1500 \text{ mg } 1^{-1}$) was transferred to a Pyrex reactor (liquid depth was 40 mm in the photoreactor). Then the lamp was switched on to initiate the reaction. During irradiation, the solution was agitated in a constant rate. At certain reaction intervals, 2 ml of sample was withdrawn and the concentration of the dye was determined by means of a spectrophotometer.

2.3. Analysis

The decolorization of AR27 was measured with UV–vis spectrophotometer (Ultrospec 2000, Biotech Pharmacia, England) at 521 nm. A calibration plot based on Beer–Lambert's law was established by relating the absorbance to the concentration.

3. Results and discussion

The changes in the absorption spectra of AR27 solutions during the UV/H₂O₂ process at different irradiation times are shown in Fig. 2. The decrease of the absorption peak of AR27 at $\lambda = 521$ nm in Fig. 2 indicates a rapid degradation of azo dye. The decrease is also meaningful with respect to the nitrogento-nitrogen double bond (-N=N-) of the azo dye, as the most active site for oxidative attack.

The photooxidative decolorization of AR27 with H_2O_2 in the presence of UV radiation is thought to be a pseudo-first order reaction with respect to the AR27 concentration. The semi-logarithmic graphs of the concentration of AR27 in the presence of different concentrations of H_2O_2 versus irradiation time (Fig. 3) yield straight lines indicating pseudo-first order reaction. The apparent reaction rate constants ($k_{ap,UV/H_2O_2}$) for photooxidation of AR27 in the presence of different concentrations of H_2O_2 were evaluated from experimental data (Fig. 3)



Fig. 2. Spectral changes of AR27 during illumination in the presence of H₂O₂ at 0.5 min intervals. [AR27]₀ = $30 \text{ mg } 1^{-1}$, [H₂O₂]₀ = $650 \text{ mg } 1^{-1}$, $I_0 = 44 \text{ W m}^{-2}$, $T = 295 \pm 2 \text{ K}$ and pH = 5.3.

using a linear regression. In all cases R^2 (correlation coefficient) values are higher than 0.99, which confirm the proposed kinetics for decolorization of AR27 in this process. Fig. 4 shows the $k_{ap,UV/H_2O_2}$ values as a function of H₂O₂ concentration. As can be seen from Fig. 4, $k_{ap,UV/H_2O_2}$ increases with increasing H₂O₂ concentration until 650 mg l⁻¹. This is due to an increase in the hydroxyl radical concentration. The enhancement of H₂O₂ concentration above 650 mg l⁻¹ retards the process. This is reasonable, because •OH efficiently reacts with H₂O₂, so that the photooxidative degradation promoter itself contributes to the •OH-scavenging capacity and reduces the decolorization rate of AR27.

The overall reaction rate expression for removal of AR27 in the UV/ H_2O_2 process can be subdivided to three parts: the reaction rate caused by H_2O_2 oxidation alone, by UV radiation



Fig. 3. Derivation of the pseudo-first order rate constants of AR27 in the UV/H₂O₂ process by linear regression. $[AR27]_0 = 30 \text{ mg } l^{-1}$, $I_0 = 44 \text{ W m}^{-2}$, $T = 295 \pm 2 \text{ K}$ and $4.5 \le \text{pH} \le 5.2$.



Fig. 4. Pseudo-first order rate constants of AR27 as a function of H_2O_2 initial concentration at different light intensities. For experimental details refer to Fig. 3.

alone and by UV/H2O2 as following:

$$-\frac{d[AR27]}{dt} = k_1[H_2O_2]_0^a[AR27] + k_2I_0^b[AR27] + k_3I_0^m[H_2O_2]_0^n[AR27]$$
(1)

where k_1 is the rate constant with H₂O₂ alone, k_2 the rate constant with UV alone, k_3 the rate constant with UV/H₂O₂, I_0 the light intensity and *a*, *b*, *m* and *n* are the reaction orders. The results show that the removal of AR27 was negligible in the absence of UV radiation, therefore we can write:

 $k_1 \cong 0$

In the direct photolysis, UV radiation can be absorbed, promoting the AR27 molecule to an excited state. That is, one electron of the molecule goes to a higher energy state or excited state. The molecule in the excited state has a very short life time, after which it returns to the ground state or decomposes to yield a different molecule. The overall reaction rate expression for UV alone process is as follows:

$$-\frac{d[AR27]}{dt} = k_2 I_0^b [AR27]$$
(2)

The semi-logarithmic graphs of the concentration of AR27 in the different light intensities versus irradiation time in UV alone process (Fig. 5) yield straight lines. The apparent reaction rate constants $(k_{ap,UV})$ for photooxidation of AR27 in the different light intensities were evaluated from Fig. 5 using a linear regression. The light intensity reaction order in UV alone process obtained with linear regression from plot of $\ln k_{ap}$ versus $\ln I_0$ as 0.75 (Fig. 6). UV photolysis has been used to eliminate chlorinated and nitrated aromatics, halogenated aliphatics and other hazardous wastes present in water. The results of investigation on direct photolysis of aromatics indicate that 90% removal of nitrobenzene, phenol and 4-nitrophenol is achieved within less than 60 min of irradiation time [8]. But our results demonstrate that direct photolysis with UV radiation at 254 nm cannot cause a considerable decolorization. Results indicate that only 18.5% of initial $30 \text{ mg } 1^{-1}$ AR27 was decomposed within 60 min UV irradiation under 44 W m⁻² light intensity. Consequently, direct photolysis alone cannot be used as an effective procedure for removal of AR27 from water.



Fig. 5. Derivation of the pseudo-first order rate constants of AR27 with UV alone by linear regression. $[AR27]_0 = 30 \text{ mg l}^{-1}$ and $T = 295 \pm 2 \text{ K}$.

If H_2O_2 is applied simultaneously with UV radiation, a considerable decrease in the concentration of the AR27 achieved. This loss can be related to the production of hydroxyl radicals, a powerful oxidizing agent. With considering negligible effect of H_2O_2 alone in photooxidation of AR27, the overall reaction rate expression for UV/ H_2O_2 process is as follows:

$$-\frac{d[AR27]}{dt} = (k_2 I_0^b + k_3 I_0^m [H_2 O_2]_0^n) [AR27]$$
(3)

where

$$k_{\rm ap,UV/H_2O_2} = k_2 I_0^b + k_3 I_0^m [H_2O_2]_0^n$$
(4)

The values of $k_{ap,UV/H_2O_2}$ were obtained from semi-logarithmic graphs of the concentration of AR27 versus irradiation time at different initial concentrations of H₂O₂ and light intensities. With subtracting $k_{ap,UV}$ from $k_{ap,UV/H_2O_2}$ we can obtain the following equation:

$$k'_{\rm ap,UV/H_2O_2} = k_{\rm ap,UV/H_2O_2} - k_{\rm ap,UV} = k_3 I_0^m [\rm H_2O_2]_0^n$$
(5)

In order to determine the H₂O₂ reaction order in the UV/H₂O₂ process, a plot of $\ln k'_{ap,UV/H_2O_2}$ versus $\ln[H_2O_2]_0$ at different light intensities are presented in Figs. 7 and 8 for low (0 mg l⁻¹ < [H₂O₂]₀ < 650 mg l⁻¹) and high (650 mg l⁻¹ < [H₂O₂]₀ <



Fig. 6. Derivation of the reaction order of light intensity in the UV alone process by linear regression. For experimental details refer to Fig. 5.



Fig. 7. Derivation of the reaction order of H_2O_2 in the UV/H₂O₂ process by linear regression at different light intensities for low concentration of H_2O_2 (0 mg $l^{-1} < [H_2O_2]_0 < 650$ mg l^{-1}). For experimental details refer to Fig. 3.



Fig. 8. Derivation of the reaction order of H_2O_2 in the UV/ H_2O_2 process by linear regression at different light intensities for high concentration of H_2O_2 (650 mg l⁻¹ < [H_2O_2] < 1500 mg l⁻¹). For experimental details refer to Fig. 3.

1500 mg l⁻¹) initial concentrations of H_2O_2 , respectively (Eq. (6)). The H_2O_2 reaction order obtained by linear regression from slope of these lines and presented in Table 1. The reaction order versus H_2O_2 has an average value of 0.49 and -0.39 for low and high initial concentrations of H_2O_2 , respectively. Malik and Sanyal [15] reported the H_2O_2 reaction order of 0.69 for mixture of three azo dyes. This difference between H_2O_2 reaction order is in the result of different chemical structure of these colored organic compounds.

$$\ln k'_{\rm ap,UV/H_2O_2} = \ln k' + n \, \ln \, [\rm H_2O_2]_0 \tag{6}$$

$$k' = k_3 I_0^m \tag{7}$$

A plot of $\ln k'_{ap,UV/H_2O_2}$ against $\ln I_0$ gives straight lines for different H_2O_2 initial concentrations (Eq. (8)) with slope of *m*. The values of *m* (light intensity reaction order) obtained by linear regression from Fig. 9 presented in Table 1. The reaction order versus light intensity has an average value of 1.38. The increase of reaction rate constant with increasing of light intensity is due

The reaction orders of light intensity and initial concentration of H_2O_2 for decolorization of AR27 in UV/ H_2O_2 process

Table 1

$I_0 ({\rm W}{\rm m}^{-2})$	$[H_2O_2]_0 \ (mg \ l^{-1})$	т	п	R^2
22	60–650	_	0.486	0.9761
30	60-650	_	0.506	0.9624
44	60-650	_	0.474	0.9558
8.7-44	300	1.377	_	0.9989
8.7-44	450	1.379	_	0.9967
8.7-44	650	1.379	-	0.9943



Fig. 9. Derivation of the reaction order of light intensity in the UV/H_2O_2 process by linear regression at different concentrations of H_2O_2 . For experimental details refer to Fig. 3.

to the enhanced production of hydroxyl radicals. At low UV power the rate of photolysis of H_2O_2 are limited and at high UV power more hydroxyl radicals are formed, upon the photodissociation of H_2O_2 , hence decolorization rate increases.

$$\ln k'_{\rm ap, UV/H_2O_2} = \ln k'' + m \ln I_0 \tag{8}$$

$$k'' = k_3 [H_2 O_2]_0^n \tag{9}$$

A comparison reaction orders for H_2O_2 and light intensity in UV alone and UV/ H_2O_2 processes obtained in this study and data in the literature are listed in Table 2. In order to insert the effect of initial dye concentration in the rate expression, the ratio of k_{ap} of the decolorization at each initial concentration of AR27 divided into k_{ap} at 30 mg l⁻¹ of AR27 and initial dye concentration correlation index φ_{AR27} was obtained. A plot of φ_{AR27} versus 1/[AR27]₀ is presented in Fig. 10 and using linear regression the following equation can be found for φ_{AR27} (Eq. (10)):

$$\varphi_{\text{AR27}} = 11.065 \frac{1}{[\text{AR27}]_0} + 0.6532 \tag{10}$$

The results show reaction rate constant decreases as initial dye concentration is increased at the same concentration of H_2O_2 . This can be postulated by considering that, the molar extinction coefficient of the dye at $\lambda < 260$ nm is very high, so that a rise in its concentration induces an inner filter effect and hence the solution becomes more and more impermeable to UV radiation. With substituting the reaction orders

Table 2

Comparison of reaction orders obtained in this study with the data in the literature for organic compounds by UV alone and UV plus $\rm H_2O_2$

Organic compound	Process	Reference		
	UV alone	UV/H ₂ O ₂ process		
	I_0 order	I_0 order	H ₂ O ₂ order	
Phenol	0.52	0.2	0.41	[14]
2-Chlorophenol	0.45	0.99	0.49	[14]
2,4-Dichlorophenol	0.72	1.22	0.83	[14]
Azo dyes mixture	_	_	0.63	[15]
<i>p</i> -Hydroxybenzoic acid	_	_	0.75	[16]
Acid Red 27	0.75	1.38	0.49	This study



Fig. 10. AR27 index (φ_{AR27}) as a function of initial dye concentration for the decolorization of AR27 in the UV/H₂O₂ process. [H₂O₂]₀ = 650 mg l⁻¹, I_0 = 44 W m⁻² and T = 295 ± 2 K.

for H₂O₂ and light intensity and φ_{AR27} in Eq. (3) following rate expression obtained for decolorization of AR27 in the UV/H₂O₂ process for 0 mg l⁻¹ < [H₂O₂]₀ < 650 mg l⁻¹ (Eq. (11)) and 650 mg l⁻¹ < [H₂O₂]₀ < 1500 mg l⁻¹ (Eq. (12)) initial concentrations of H₂O₂, respectively. In these equations light intensity is between 0 and 44 W m⁻² and initial concentration of AR27 is between 10 and 30 mg l⁻¹.

$$-\frac{d[AR27]}{dt} = (2 \times 10^{-4} I_0^{0.75} + 3 \times 10^{-4} I_0^{1.38} [H_2O_2]_0^{0.49}) \varphi_{AR27}$$
$$\times [AR27]$$
(11)

$$-\frac{d[AR27]}{dt} = (2 \times 10^{-4} I_0^{0.75} + 0.1 I_0^{1.38} [H_2O_2]_0^{-0.39}) \varphi_{AR27} \times [AR27]$$
(12)

The above equations can be used for predicting pseudo-first order rate constant (k_{ap}) for different light intensities, initial concentrations of H₂O₂ and AR27. The calculated results from Eqs. (11) and (12) correlated the experimental results well as shown in Fig. 11 for different conditions. The R^2 (correlation coefficient) has a value of 0.99, which indicates the model is able to predict the reaction rate constant at different conditions such as different initial concentrations of H₂O₂ and AR27 and light intensities with high accuracy and reliability. Although the rate expression is empirical in nature, it provides information about



Fig. 11. Comparison between experimental and calculated data for the decolorization of AR27 in the UV/H₂O₂ process. $0 \text{ W m}^{-2} < I_0 < 44 \text{ W m}^{-2}$, $0 \text{ mg} \text{ l}^{-1} < [\text{H}_2\text{O}_2]_0 < 1500 \text{ mg} \text{ l}^{-1}$ and $10 \text{ mg} \text{ l}^{-1} < [\text{AR27}]_0 < 30 \text{ mg} \text{ l}^{-1}$.

Table 3 Pseudo-first order rate constants for decolorization of AR27 by UV alone and UV plus H_2O_2 at different conditions

$I_0 ({\rm W}{\rm m}^{-2})$	$\begin{array}{c} [H_2O_2]_0 \\ (mgl^{-1}) \end{array}$	$k_{\rm ap,UV}$ (min ⁻¹)	$k_{\mathrm{ap,UV/H_2O_2}}$ (min ⁻¹)	$\frac{k_{\rm ap,UV/H_2O_2}}{k_{\rm ap,UV}}$
8.7	650	0.0006	0.1420	237
22	650	0.0021	0.6014	286
30	650	0.0027	0.8566	317
44	1500	0.0035	0.9024	258
44	1200	0.0035	1.0301	294
44	900	0.0035	1.1278	322
44	650	0.0035	1.3057	373
44	450	0.0035	1.2318	352
44	300	0.0035	1.1030	315
44	180	0.0035	0.8493	243
44	120	0.0035	0.6774	194
44	60	0.0035	0.4133	118

 $[AR27]_0 = 30 \text{ mg } 1^{-1}$ at all runs.

the effect of operational parameters on the reaction rate that is useful for design.

Table 3 lists the rate constants for reactions with UV alone and UV plus H₂O₂. The last column in Table 3, which gives the ratio of the rate constants for UV plus H₂O₂ to the rate constants for UV alone, provides a measure of the degree of enhancement due to H_2O_2 . The results show that, as the ratio of H_2O_2 to AR27 and light intensity increases, more hydroxyl radicals are available to attack the AR27 and the rate of reaction is increased. The results show that in the optimum conditions the reaction rate for decolorization of AR27 is about 373 times greater with H_2O_2 , whereas Sundstrom et al. [17] reported only the values 47, 26 and 3.3 times increase with H_2O_2 for diethyl phthalate, phenol and 2-chlorophenol, respectively. This comparison shows that the synergistic effect of UV light and H₂O₂ was most pronounced for AR27 (as a model compound from monoazo dyes) in contrast with other aromatic compounds. These results are in good agreement with other findings in literature such as, Aleboyeh et al. [18] who showed combination of UV plus H₂O₂ in comparison with UV alone increases removal rates of Acid Orange 8 and Methyl Orange, 172 and 137 times, respectively. All of these results show that the first term in the rate expression (decolorization with UV alone) can be neglected in the decolorization of azo dyes.

4. Conclusions

The UV/H_2O_2 process is proved to be powerful method for destroying AR27. The semi-logarithmic graphs of the concen-

tration of AR27 versus illumination time were linear suggesting pseudo-first order reaction. The results indicate that apparent reaction rate constant in the UV/H₂O₂ process is a function of H₂O₂ concentration and light intensity. The correlation between apparent reaction rate constant with H₂O₂ amount and light intensity were obtained with linear regression. The results show that UV alone cannot be an efficient method for decolorization of AR27. The rate of reaction was up to 373 times faster with UV light plus H₂O₂ than with UV light alone. An empirical expression, with two terms, was developed to correlate the reaction rate to light intensity and H₂O₂ and AR27 initial concentrations. The k_{ap} values predicted by the rate equations are in good agreement with the experimental data.

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