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Kinetic modeling of photocatalytic degradation of Acid Red 27 in UV/TiO$_2$ process

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Abstract

The photocatalytic degradation of Acid Red 27 (AR27) was investigated using UV radiation in the presence of TiO$_2$-P25 as a function of initial concentration of AR27 ([AR27]$_0$), oxygen concentration ([O$_2$]), TiO$_2$-P25 amount, light intensity (I_a), pH and temperature. The activation energy of the photocatalytic degradation of AR27 was 8.24 kJ mol^{-1}. The photocatalytic degradation rate follows pseudo-first order kinetic with respect to the AR27 concentration. The following rate of decolorization was achieved by kinetic modeling:

$$r_{\mathrm{AR27}} = 5.294\, I_a \exp\left(\frac{-8247}{RT}\right)\left(\frac{0.79\,[\mathrm{O_2}]}{1+0.79\,[\mathrm{O_2}]}\right)\left(\frac{4.23\,[\mathrm{AR27}]}{1+4.23\,[\mathrm{AR27}]_0}\right)$$

The calculated results obtained from above equation were in good agreement with experimental data. This model predicts the concentration of AR27 during the photodegradation process at different temperatures, light intensities, AR27 and oxygen concentrations.

Keywords: Decolorization; Photocatalytic degradation; Kinetic model; Azo dyes; Acid Red 27

1. Introduction

Textile finishing mills discharge wastewater containing a great variety of organic contaminants in a wide range of concentration. The release of these colored wastewaters poses a major problem for the industry as well as a threat to the environment [1,2]. The number of dyes presently used in textile industry is about 10 000. Among these dyes, azo dyes constitute the largest and the most important class of commercial dyes [3].

For the treatment of these dyes biological process is ineffective. Also common treatment processes, e.g. adsorption on activated carbon, flocculation and electrocoagulation [4] are not efficient methods because they appear in solid wastes, thus creating other environmental problems requiring further treatment.

However, the recent developments of chemical treatment of wastewaters gave birth to an improvement of the catalytic and oxidative degradation of organic compounds dissolved in aqueous media [5–8]. These are generally referred to as “advanced oxidation processes” (AOPs). Among these processes photocatalysis systems such as combination of a semiconductor (TiO$_2$, ZnO, etc.) with UV light is a very promising technique. Due to a faster electron transfer to molecular oxygen, TiO$_2$ is found to be more efficient for photocatalytic degradation of pollutants [7]. It has been demonstrated that in UV/TiO$_2$ process various parameters affect the degradation rate of compounds. A few degradation kinetic expressions of different compounds have been reported, in particular, the Langmuir-Hinshelwood law has been qualitatively proved in numerous cases of photocatalytic degradation [9,10].

In our previous work the effect of operational parameters on photocatalytic degradation of Acid Red 14 by UV/TiO$_2$ process was reported [6], so in the present work a reaction mechanism and kinetic model of photocatalytic degradation of Acid Red 27 (AR27) using TiO$_2$-P25 as catalyst were

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theoretically and experimentally studied, and the kinetic parameters were also obtained.

2. Experimental

2.1. Materials

Acid Red 27 (AR27), a monoazo anionic dye, was obtained from Boyakh Saz Company (Iran) and used without further purification. Its chemical structure and other characteristics are listed in Table 1. Titanium dioxide was Degussa P-25 and it was in the anatase form (approximately 80% anatase, 20% rutile). It had a BET surface area of 50 ± 15 $m^2\,g^{-1}$ and an average particle diameter of 21 nm, containing 99.5% TiO_2. The NaOH and HCl were purchased from Merck (Germany). Solutions were prepared by dissolving requisite quantity of the dye in double distilled water.

2.2. Photoreactor

All experiments were carried out in a batch photoreactor. The radiation source was a mercury UV lamp (30 W, UV-C, manufactured by Philips, Holland) which was placed above a 500 ml jacket Pyrex glass reactor. In the section of the effect of light intensity, irradiations were carried out using a 4, 8, 15 and 30 W mercury UV lamp. The light intensity in the center of photoreactor was measured by a Lux-UV-IR meter (Leybold Co.). The reaction temperature was controlled by circulation of water at the desired temperature through the jacket. For adjusting of oxygen concentration at the desired level, suspensions were continuously purged with O_2, N_2 and mixture of both through a gas disperser placed at the bottom of the reactor before and during the illumination.

2.3. Procedures

For the photocatalytic degradation of AR27, a solution containing known concentration of dye and TiO_2-P25 was prepared and it was allowed to equilibrate for 30 min in the darkness, then 200 ml of the prepared suspension was transferred to the reactor. The liquid film was 40 mm thick. After adjusting of temperature and oxygen concentration, the lamp was switched on to initiate the reaction. During irradiation, agitation was maintained to keep the suspension homogenous, and at certain reaction intervals, 10 ml of sample was withdrawn, centrifuged and filtered through a 0.2 μm membrane filter (Schleicher & Schuell, Germany) and the reminiscent dye concentration analyzed with a 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. The effect of pH was studied with adjusting pH by addition of NaOH or HCl in the range of 2–11. The measurement of oxygen was carried out by an Oximeter (Oxi 538, WTW, Germany). The operative conditions of experiments are summarized in Table 2.

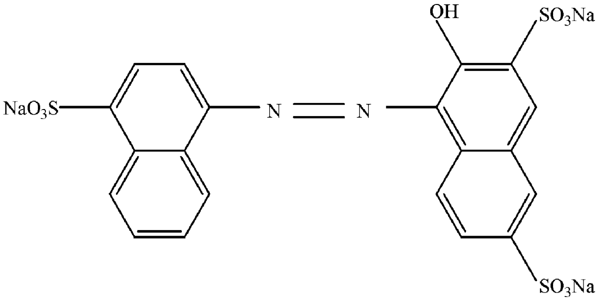
Table 2
Operating conditions of experiments.

Parameters	Values
AR27 initial concentration	5–40 $mg\,l^{-1}$
TiO_2 amount	0–400 $mg\,l^{-1}$
Light intensity	0–8.6 $W\,m^{-2}$
Oxygen concentration	1.8–20 $mg\,l^{-1}$
Temperature	281–315 K
pH	2.75–10.8

3. Kinetic modeling

It has been well demonstrated that when TiO_2 is illuminated by light ($\lambda < 390$ nm) electrons are promoted from the valence band to the conduction band to give electron-hole pairs. The holes at the TiO_2 valence band, having an oxidation potential of +2.6 V versus normal hydrogen electrode (NHE) at pH = 7, can oxidize water or hydroxide to produce hydroxyl radicals. The hydroxyl radical is a powerful oxidizing agent and attacks to organic compounds and intermediates (Int.) are formed. These intermediates react with hydroxyl radicals to produce final products (P), also hydroxyl radicals could be consumed by inactive species [11]. Our paper reports a simple kinetic model according to the

Table 1
Structure and characteristics of Acid Red 27

Structure	Other names	C.I. number	λ_{max} (nm)	M_w ($g\,mol^{-1}$)
	Amaranth Azorubin S	16185	521	604.48

following reactions:

$$TiO_2 + h\nu \xrightarrow{k_1} e^- + h^+ \tag{1}$$

$$e^- + h^+ \xrightarrow{k_2} \text{heat} \tag{2}$$

$$h^+ + H_2O_{ads} \xrightarrow{k_3} {}^{\bullet}OH_{ads} + H^+ \tag{3}$$

$$h^+ + OH^-_{ads} \xrightarrow{k_4} {}^{\bullet}OH_{ads} \tag{4}$$

$$^{\bullet}OH_{ads} + AR27_{ads} \xrightarrow{k_5} \text{Int.} \tag{5}$$

$$^{\bullet}OH_{ads} + \text{Int.} \xrightarrow{k_6} P \tag{6}$$

$$^{\bullet}OH_{ads} + S_{inactive} \xrightarrow{k_7} \text{inactive species} \tag{7}$$

In the photodegradation of AR27 by UV radiation in the presence of TiO_2, it is postulated that the rate-determining step is the reaction between adsorbed $^{\bullet}OH$ radicals and AR27. The rate of disappearance of the AR27 may be represented by:

$$r_{AR27} = k_5[^{\bullet}OH]_{ads}[AR27]_{ads} \tag{8}$$

The concentration of photon-induced holes (h^+) can be obtained by applying steady-state assumption.

$$\frac{d[h^+]}{dt} = k_1 I_a - k_2[h^+][e^-] - k_3[h^+][H_2O]_{ads} - k_4[h^+][OH^-]_{ads} = 0 \tag{9}$$

$$\frac{d[h^+]}{dt} = k_1 I_a - k_2[h^+][e^-]k_8[h^+] = 0 \tag{10}$$

$$k_8 = k_3[H_2O]_{ads} + k_4[OH^-]_{ads} \tag{11}$$

Terzian and Serpone [12] showed at low light intensities, h^+ trapping competes effectively with electron-hole recombination.

$$k_8[h^+] \gg k_2[h^+][e^-] \tag{12}$$

Therefore, Eq. (10) simplifies to:

$$[h^+] = \frac{k_1 I_a}{k_8} \tag{13}$$

Similarly, if the steady-state approximate is used for the concentration of $^{\bullet}OH$ radicals, we obtain:

$$\frac{d[^{\bullet}OH]}{dt} = k_3[h^+][H_2O]_{ads} + k_4[h^+][OH^-]_{ads} - k_5[^{\bullet}OH]_{ads}[AR27]_{ads} - k_6[^{\bullet}OH]_{ads}[\text{Int.}] - k_7[^{\bullet}OH]_{ads}S_{inactive} = 0 \tag{14}$$

If we suppose that deactivation of $^{\bullet}OH$ radicals with inactive surfaces (S) is very important than other processes, we obtain:

$$[^{\bullet}OH]_{ads} = \frac{k_8}{k_7 S_{inactive}}\left[h^+\right] = k'[h^+] \tag{15}$$

Combination of Eqs. (13) and (15) in Eq. (8) yields:

$$r_{AR27} = \frac{k' k_1 k_5}{k_8} I_a [AR27]_{ads} \tag{16}$$

If the Langmuir adsorption model is applied to AR27 in this system, then we obtain:

$$r_{AR27} = \frac{k_p I_a K_{AR27}[AR27]}{1 + K_{AR27}[AR27]} \tag{17}$$

where K_{AR27} is the adsorption equilibrium constant for AR27 and $k_p = k' k_1 k_5 / k_8$. Al-Ekabi and Serpone [9] and Chan et al. [13] have admitted that the rate must include competitive adsorption by solvent, intermediates and pollutants. Under these reasonable conditions, the Eq. (17) can be written as follows:

$$r_{AR27} = \frac{k_p I_a K_{AR27}[AR27]}{1 + K_{AR27}[AR27] + \sum K_i C_i} \tag{18}$$

In this equation K_i is the adsorption equilibrium constant for solvent, intermediates and pollutants. Beltran-Heredia et al. [14] made the following assumption:

$$K_{AR27}[AR27] + \sum K_i C_i = K_{AR27}[AR27]_0 \tag{19}$$

In this equation $[AR27]_0$ is the initial concentration of AR27. With substituting Eq. (19) in to Eq. (18), we obtain:

$$r_{AR27} = \frac{k_p I_a K_{AR27}[AR27]}{1 + K_{AR27}[AR27]_0} \tag{20}$$

The limitation of the rate of photocatalytic degradation is also attributed to the recombination of photogenerated hole-electron pairs. The adsorbed oxygen on the surface of TiO_2 prevents the recombination process by trapping electrons. Kormann et al. [15] and Crittenden et al. [16] indicated a noncompetitive Langmuir-Hinshelwood model for adsorption of oxygen on the surface of TiO_2 as follows:

$$r \propto \frac{K_{O_2}[O_2]}{1 + K_{O_2}[O_2]} \tag{21}$$

where K_{O_2} is the adsorption equilibrium constant of oxygen. The above equation shows that the reaction rate is proportional and independent of oxygen concentration at low and high O_2 concentration, respectively. Finally, the reaction rate can be written as follows:

$$r_{AR27} = \frac{k_p I_a K_{AR27}[AR27]}{1 + K_{AR27}[AR27]_0} \frac{K_{O_2}[O_2]}{1 + K_{O_2}[O_2]} \tag{22}$$

When O_2 concentration and light intensity are constant we may write:

$$k_c = \frac{k_p I_a K_{O_2}[O_2]}{1 + K_{O_2}[O_2]} \tag{23}$$

In this state Eq. (22) can be written as:

$$r_{AR27} = \frac{k_c K_{AR27}[AR27]}{1 + K_{AR27}[AR27]_0} = k_{ap}[AR27] \tag{24}$$

where

$$k_{ap} = \frac{k_c K_{AR27}}{1 + K_{AR27}[AR27]_0} \quad (25)$$

Eq. (24) shows a pseudo-first order reaction with respect to the AR27 concentration.

4. Results and discussion

4.1. Influence of operational variables and estimation of model parameters

4.1.1. Effect of the amount of TiO_2

The effect of different dosages of TiO_2-P25 on the photocatalytic degradation of AR27 was examined. Fig. 1 shows the variation of pseudo-first order rate constant with different TiO_2-P25 dosages. The rate constant was found to increase with the increasing amount of TiO_2-P25 and approaching a limiting value at high dosage. The reason of this observation is thought to be the fact that at the concentration range from 0 to 300 mg l^{-1} the observed enhancement in pseudo-first order rate constant is probably due to an increased number of available adsorption and catalytic sites on TiO_2-P25. A further increase in catalyst loading, may cause opacity and light scattering and thus decrease in the passage of irradiation through the solution. Effect of TiO_2-P25 amount on pseudo-first order rate constant (k_{ap}) was fitted with a Langmuir-type expression:

$$k_{ap} = \frac{k_0 K_{TiO_2}[TiO_2]}{1 + K_{TiO_2}[TiO_2]} \quad (26)$$

Values of k_0 and K_{TiO_2} were obtained from the nonlinear regression of the experimental data to Eq. (26) as $k_0 = 0.1\,\text{min}^{-1}$ and $K_{TiO_2} = 0.003\,\text{mg}^{-1}\,\text{l}$.

4.1.2. Effect of the initial dye (AR27) concentration

It is important from an application point of view to study the dependence of removal efficiency on the initial concentration of dye. Experiments show k_{ap} decreased with increasing of initial concentration of AR27 when other parameters are kept unchanged. Therefore, the degradation rate was pseudo-first order with respect to the initial concentration of AR27. Eq. (25) can be tested using experimental data after transforming it to a straight-line equation as follows:

$$\frac{1}{k_{ap}} = \frac{1}{k_c}[AR27]_0 + \frac{1}{k_c K_{AR27}} \quad (27)$$

A plot of the $1/k_{ap}$ against $[AR27]_0$ gives the values of slope ($1/k_c$) and intercept ($1/k_c\ K_{AR27}$) (Fig. 2). From the intercept and slope of this curve, k_c and K_{AR27} values for AR27 were found to be 1.39 mg l^{-1} min^{-1} and 4.23 mg^{-1} l, respectively.

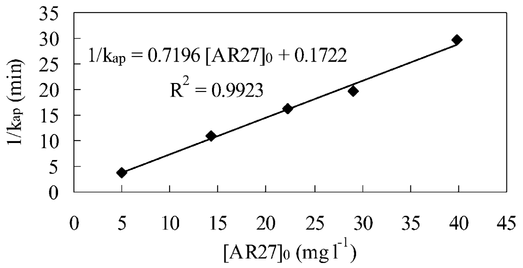


Fig. 2. Plot of reciprocal of pseudo-first order rate constant against initial concentration of AR27. TiO_2 = 300 mg l^{-1}, $T = 295 \pm 2$ K, neutral pH, light intensity = 8.6 W m^{-2}, $[O_2]$ = 7 mg l^{-1}.

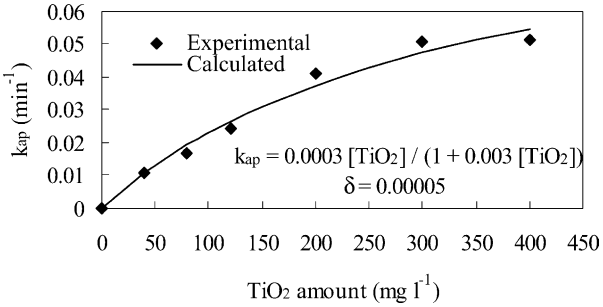


Fig. 1. Effect of TiO_2 amount on the pseudo-first order rate constant. $[AR27]_0$ = 30 mg l^{-1}, $T = 295 \pm 2$ K, neutral pH, light intensity = 8.6 W m^{-2}, $[O_2]$ = 7 mg l^{-1}.

4.1.3. Effect of reaction temperature

In a range of 280–315 K a linear relationship between $\ln k_{ap}$ and $1/T$ was observed. The slope of this straight line was 991.93, which corresponds to an activation energy of 8.24 kJ mol^{-1} (Fig. 3). This value is in agreement with the findings for photocatalytic reactions. A comparison of activation energy obtained in this study with the data in the literature listed in Table 3. The values listed in Table 3 show the semiconductor photocatalysis is usually not very temperature dependent. However, an increase in temperature helped the reaction to compete more efficiently with e$^-$–h$^+$ recombination.

4.1.4. Effect of oxygen concentration

Eq. (22) shows pseudo-first order rate constant (k_{ap}) is a function of oxygen concentration as follows:

$$k_{ap} = \frac{k'_c K_{O_2}[O_2]}{1 + K_{O_2}[O_2]} \quad (28)$$

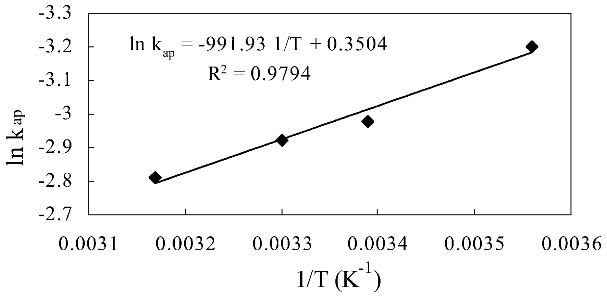


Fig. 3. Effect of reaction temperature on the pseudo-first order rate constant. TiO_2 = 300 mg l^{-1}, $[AR27]_0$ = 30 mg l^{-1}, pH = 5.38, light intensity = 8.6 W m^{-2}, $[O_2]$ = 7 mg l^{-1}.

Table 3
Comparison of activation energy obtained in this study with the data in the literature for photocatalysis systems

Reactant	Catalyst	Activation energy ($kJ\,mol^{-1}$)	Reference
Dodecylbenzene sulfonate	TiO_2 (Merck)	39.1	Daneshvar et al. [5]
Xylenols	TiO_2-P25 (Degussa)	8.8	Terzian and Serpone [12]
4-Chlorophenol	TiO_2-P25 (Degussa)	5.5	Al-Sayyed et al. [17]
Methylene blue	TiO_2-P25 (Degussa)	8.9	Zhang et al. [18]
Acid Red 27	TiO_2-P25 (Degussa)	8.24	This study

where

$$k_c' = \frac{k_p I_a K_{AR27}}{1 + K_{AR27}[AR27]_0} \tag{29}$$

A plot of the $1/k_{ap}$ against $1/[O_2]$ gives a straight line with a slope $23.93\,mg\,l^{-1}\,min$ and an intercept of 18.71 min as shown in Fig. 4. From the intercept and slope of this curve, k_c' and K_{O_2} values were $0.053\,min^{-1}$ and $0.79\,mg^{-1}\,l$, respectively. The positive influence of oxygen concentration in photocatalysis system can be related to enhance the separation of photogenerated electron-hole pairs, hereby increasing hydroxyl radical concentration.

4.1.5. *Effect of light intensity*

The pseudo-first order rate constant (k_{ap}) in the low intensities is proportional with light intensity with the following relationship:

$$k_{ap} = k_c'' I_a \tag{30}$$

A plot of k_{ap} versus I_a yielded straight line and showed that the experimental data are in good agreement with kinetic model (Fig. 5). Previous studies indicated that at low intensity level the reaction rate is proportional to the light intensity [12]. This result indicated that the electron-hole pairs are consumed more rapidly by chemical reactions than by recombination.

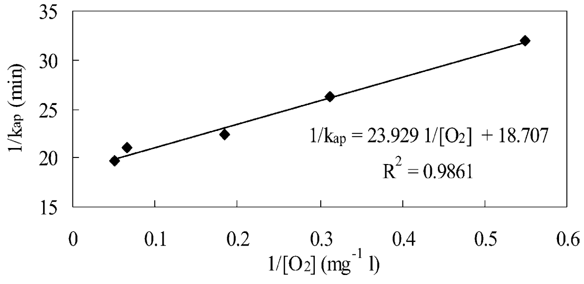


Fig. 4. Plot of reciprocal of pseudo-first order rate constant against reciprocal of oxygen concentration. $TiO_2 = 300\,mg\,l^{-1}$, $[AR27]_0 = 30\,mg\,l^{-1}$, $T = 295 \pm 2\,K$, pH = 5.38, light intensity $= 8.6\,W\,m^{-2}$.

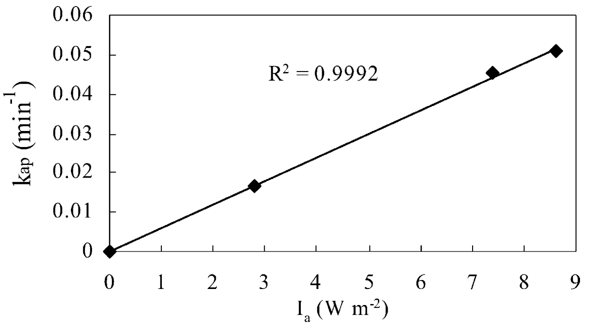


Fig. 5. Plot of pseudo-first order rate constant against light intensity. $TiO_2 = 300\,mg\,l^{-1}$, $[AR27]_0 = 30\,mg\,l^{-1}$, $T = 295 \pm 2\,K$, pH = 5.38, $[O_2] = 7\,mg\,l^{-1}$.

4.1.6. *Effect of the initial pH*

The pH value is an important operational variable in actual wastewater treatment. In photocatalysis systems, pH value is also one of the factors influencing the rate of degradation. Results obtained from experiments with varying pH from 2.75 to 10.80 are illustrated in Fig. 6. From the plot, it is deduced that the pseudo-first order rate constant (k_{ap}) is higher in the acidic and basic conditions. The zero point charge (zpc) for TiO_2 is at pH between 5.6 and 6.4. Hence, at more acidic pH values, the catalyst surface is positively charged, while at pH values above 5.6, it is negatively charged [11]. Since the dye has three sulfonic groups in its structure, which is negatively charged, the acidic solution favors adsorption of dye onto photocatalyst surface, thus k_{ap} increases. Increasing of the k_{ap} under alkaline condition could be attributed to the increase of hydroxyl ions which induces more hydroxyl radical formation.

4.2. *Development of rate equation*

As discussed in the previous sections, the rate of photodegradation is a function of initial concentration of AR27, oxygen concentration, light intensity and temperature as follows:

$$r_{AR27} = k_p I_a \exp\left(\frac{-E_a}{RT}\right)\left(\frac{K_{O_2}[O_2]}{1+K_{O_2}[O_2]}\right)\left(\frac{K_{AR27}[AR27]}{1+K_{AR27}[AR27]_0}\right) \tag{31}$$

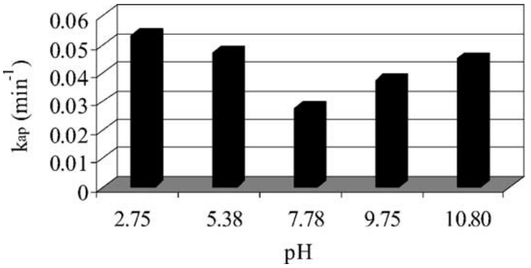


Fig. 6. Effect of pH on the pseudo-first order rate constant. $TiO_2 = 300\,mg\,l^{-1}$, $[AR27]_0 = 30\,mg\,l^{-1}$, $T = 295 \pm 2\,K$, light intensity $= 8.6\,W\,m^{-2}$, $[O_2] = 7\,mg\,l^{-1}$.

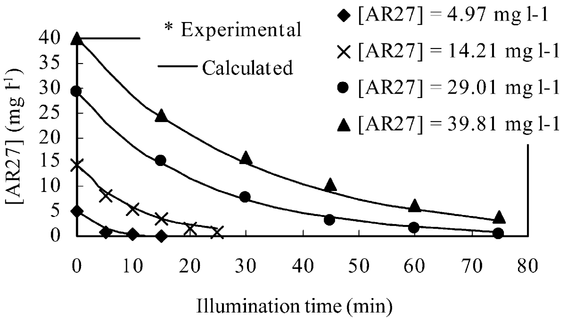


Fig. 7. Experimental and calculated AR27 concentration versus irradiation time for different AR27 initial concentrations. Refer to Fig. 2 for experimental details.

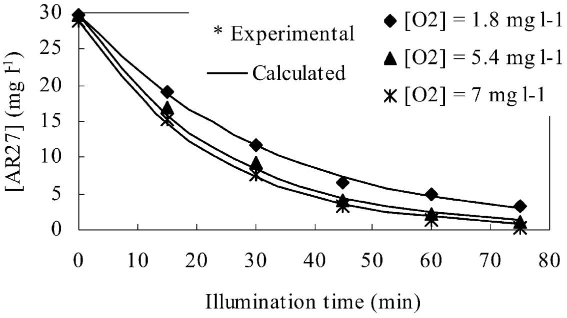


Fig. 8. Experimental and calculated AR27 concentration versus irradiation time for different oxygen concentrations. Refer to Fig. 4 for experimental details.

From previous sections, K_{O_2} and K_{AR27} values were estimated from experimental data. Since, $[AR27]_0$, $[O_2]$, I_a, T, K_{O_2} and K_{AR27} are known, we can calculate k_p as $5.924\,mg\,l^{-1}\,min^{-1}\,W^{-1}\,m^2$. Substituting these values to Eq. (31), we obtain:

$$r_{AR27} = 5.294\, I_a \exp\left(\frac{-8247}{RT}\right)\left(\frac{0.79\,[O_2]}{1+0.79\,[O_2]}\right)\left(\frac{4.23[AR27]}{1+4.23[AR27]_0}\right) \qquad (32)$$

The calculated results from Eq. (32) correlated the experimental results well as shown in Figs. 7–10.

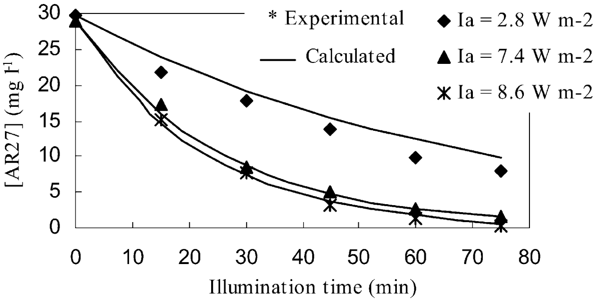


Fig. 9. Experimental and calculated AR27 concentration versus irradiation time for different light intensities. Refer to Fig. 5 for experimental details.

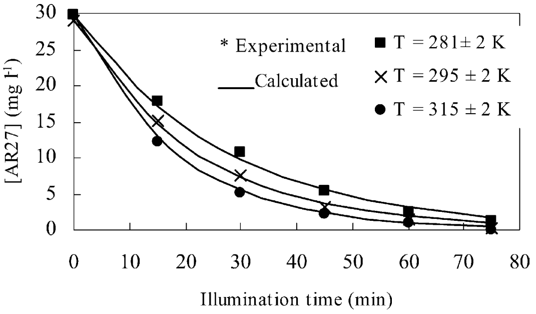


Fig. 10. Experimental and calculated AR27 concentration versus irradiation time at different temperatures. Refer to Fig. 3 for experimental details.

5. Conclusions

AR27, an anionic monoazo dye of acid class, can be easily degraded by UV radiation in the presence of TiO_2-P25 as a photocatalyst. The pseudo-first order rate constant (k_{ap}) is sensitive to the operational parameters. The k_{ap} is a function of pH value and is higher in the acidic conditions. The activation energy of the photocatalytic degradation of AR27 is $8.24\,kJ\,mol^{-1}$. The photocatalytic degradation of AR27 can be explained in terms of Langmuir-Hinshelwood kinetic model. AR27 adsorption occurs in competitive with that of the solvent, pollutants and intermediates and the dissolved oxygen adsorption occurs at different sites. On the basis of these assumptions and kinetic modeling, AR27 concentration profiles can be predicted in different initial concentrations of AR27, oxygen concentrations, light intensities and temperatures. The results obtained from kinetic modeling are in good agreement with experimental data.

Acknowledgements

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