



Development of photoreactor design equation for the treatment of dye wastewater by UV/H₂O₂ process

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Abstract

The treatment of Direct Yellow 86 dye wastewater by the UV/H₂O₂ process in continuous annular photoreactors was studied under various UV light intensities, influx concentrations of dye, dosages of H₂O₂ and dimensions of photoreactor. A photoreactor design equation combined the UV light distribution profile in the reactor and the empirical rate expressions for the decomposition of dye and H₂O₂ was used to predict the destruction of dye within photoreactors of different geometries at various operating conditions. Experimentally observed removal of the dye pollutant in the plug flow annular reactor agreed well with the theoretical solutions modeled by the developed photoreactor design equation. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Dye wastewater; UV/H₂O₂ process; Photoreactor design; Photooxidation; Advanced oxidation processes (AOPs)

1. Introduction

Textile finishing mills discharge wastewater containing a great variety of organic contaminants in a wide range of concentrations. Due to the large degree of aromatics present in these molecules and the stability of modern dyes, conventional biological treatment methods are ineffective for decolorization and degradation [1,2]. During the last decade, attention has been focused on chemical techniques that could be used to discolor textile wastewater [3,4]. Among these techniques, UV irradiation in the combination with some powerful oxidants (e.g. O₃ and H₂O₂) can be a promising alternative for the effective removal of color and refractory organics from the effluent because it does not generate chemical sludge and, moreover, toxicity of wastewater is generally reduced. In particular, the UV/H₂O₂ oxidation process is of potential practical importance [5,6]. It is characterized by the generation of a

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Nomenclature

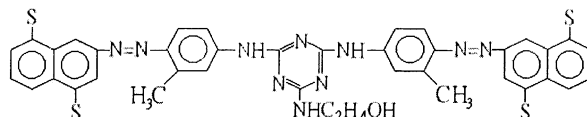
C_{dye}	concentration of dye (ppm)
$C_{\text{dye,eff}}$	the effluent concentration of dye (mM)
$C_{\text{dye,eff,ave}}$	average residual concentration of dye, in the effluent of photoreactor
$C_{\text{dye,in}}$	the inlet concentration of dye (mM)
D_{AB}	binary diffusivity of dye in water ($\text{l}^2 \text{s}^{-1}$)
E	the monochromatic absorbance of water
$[\text{H}_2\text{O}_2]$	H_2O_2 dosage (mM l^{-1})
I	UV light intensity (W m^{-2})
k	rate constant
m	order with respect to I
n	order with respect to H_2O_2 dosage
p	order with respect to C_{dye}
R_{dye}	photooxidation rate of dye ($\text{M s}^{-1} \text{l}^{-1}$)
V	the space volume of the annular reactor
v_z	the velocity of solution in z -direction a cylindrical coordinate
<i>Greek letters</i>	
ε_{dye}	the molar absorption coefficient of Direct Yellow 86 ($\text{M}^{-1} \text{cm}^{-1}$)
$\varepsilon_{\text{H}_2\text{O}_2}$	the molar absorption coefficient of hydrogen peroxide ($\text{M}^{-1} \text{cm}^{-1}$)
τ	retention time of the dye-containing solution within the photoreactor

very powerful oxidizing species, well known as hydroxyl radicals, to decompose and even mineralize organic compounds.

Several studies have been reported about the successful application of the UV/ H_2O_2 process for dyes treatment [7–14]. Galindo and Kalt [7] demonstrated that the UV/ H_2O_2 process is able to destroy totally the chromophore structure of azo dyes and the reaction rate of azo dyes depends on the basic structure of the molecule and on the nature of auxiliary groups attached to the aromatic nuclei of the dyes. Ince [8] established a useful mathematical relation between the rate of dye removal in a UV/ H_2O_2 system and the applied H_2O_2 dosage, as represented by either the fraction of UV light it absorbs, or the ratio of its initial concentration to that of the dye. Colonna et al. [11] reported that the UV irradiation in the presence of hydrogen peroxide leads to complete decolorization and mineralization of sulphonated azo and anthraquinone dyes in a relatively short time. A simple kinetic model was also proposed which can describe the process in a substantially adequate way. Much of the past efforts on the treatment of dye wastewater by the UV/ H_2O_2 process were mainly based on the phenomenological descriptions of the removal of dye pollutants in a batch reactor. However, the study on the development of the design equation of the dye removal by UV/ H_2O_2 process in a continuous photoreactor is scarce.

Most of the past researches on the modeling of photoreactors had been directed to develop expressions for the light intensity distribution for some simple photoreactor configurations [15–17]. Models have also been developed to analyze the problem of irradiation different

reactor geometries transversely, axially or in situ. The purpose of this work is to explore the feasibility and efficiency of the dye removal in aqueous solutions by the UV/H₂O₂ process in continuous annular reactors and to develop a practicable design equation for predicting the destruction behaviors of the dye at various operating conditions. Direct Yellow 86 was used as a surrogate chemical to represent azo dyes as shown in the following chemical structure:



Due to the limitations for determining the distributions of UV light intensity and the dye concentrations within the annular photoreactors, several dimensions of photoreactors were designed in this work to evaluate the treatment efficiency of dye by UV irradiation. The infinite line source model was adopted to describe the profile of UV light intensity and dye concentration in the photoreactors.

2. Experimental

Fig. 1 is a schematic drawing of the typical annular UV photoreactor employed. The photoreactor consisted of one inner diameter quartz tube housing a UVP-XX-15S 254 nm low pressure mercury UV lamp with approximately 5.3 W maximum output. The UV light intensity of the UV lamp was adjusted by a variable voltage transformer and detected by a Spectroline model DRC-100X digital radiometer combined with a DIX-254 radiation sensor. In this work, eight photoreactors with different dimensions as shown in Table 1, were employed, where r_i is the inner radius of the annular photoreactor which is measured as the radius of the inner quartz tube and r_o is the outer radius of the annular photoreactor, which is measured as the radius of the outer Pyrex tube.

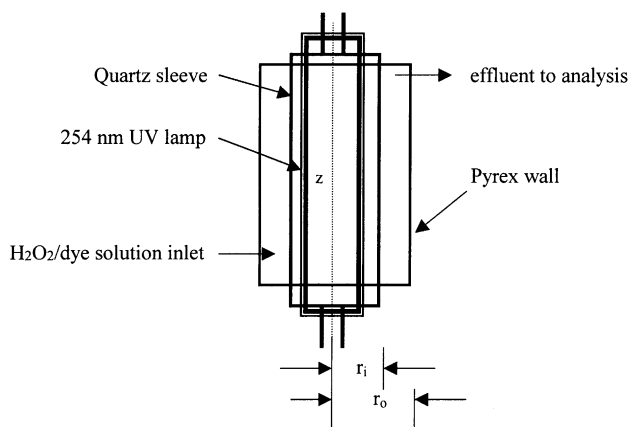


Fig. 1. The schematic drawing of the annular photoreactor employed.

Table 1

The geometry of the annular photoreactors employed in this work^a

Identification of reactor	r_i (cm)	r_o (cm)	r_i/r_o	Effective volume of reactor (cm ³)
R1	1.5	2.5	0.60	503
R2	2.0	2.5	0.80	283
R3	1.5	3.0	0.50	848
R4	2.0	3.0	0.67	628
R5	2.5	3.0	0.83	346
R6	1.5	3.5	0.43	1257
R7	2.0	3.5	0.57	1037
R8	2.5	3.5	0.71	754

^a Length (L) of all photoreactors is 40 cm.

The average UV light intensities on the surfaces of UV lamp and quartz tubes at different electric voltages are listed in Table 2. The dye and other chemical used for analysis were reagent grade and all experimental solution were prepared with deionized water. The dye/H₂O₂ mixture solution with desired molar ratios was pre-mixed in a 10 l vessel and the inlet flow rate into the reactor was kept at the desired retention time by a mass flow meter. The temperature of the reacting solution was water-jacketed to maintain at 25°C for all experiments. At desired time intervals, aliquots of 10 ml solution were withdrawn from the sampling port located at the top of the reactor after the UV lamp was turned on and the concentration of reacted dye reached steady. The inlet and effluent concentrations of the dye were determined by a HP Diode Array spectrophotometer. The standard deviation of the concentration of the dye in aqueous solutions was analyzed to be $\pm 0.3 \text{ mg l}^{-1}$.

3. Model development

As depicted in Fig. 1, the lamp is housed within a quartz tube in this UV reaction system. Distance in the radial direction within the annular UV reactor is represented by r . The variable z represents the vertical distance along the line source, and L is the length of line source. The reflection or refraction effect of light at all interfaces is assumed to be

Table 2

The average surface UV light intensity on the UV lamp and quartz tubes at various electric voltages

Voltage (V)	Average surface UV light intensity (W m ⁻²)			
	UV lamp (254 nm)	Inner radius of quartz tube r_i (cm)		
		1.5	2.0	2.5
90	101	70	52	44
100	120	76	60	52
110	137	88	65	59
120	150	92	74	65
130	164	103	84	73

negligible. Light is assumed to be emitted from a hypothetical line source located at the center of the lamp. In the modeling of a photochemical reactor, the conservation equations of mass of reacting species, momentum, energy, and radiant energy at a given wavelength should be established to analyze a chosen reactor geometry. To reduce the complexity of the model, the plug flow of gas stream in the annular reactor is assumed to be ideal, the momentum conservation equations of the photoreactants can be neglected. In addition, the reaction system is postulated to be isothermal, the energy conservation equations of the reacting species can be ignored. Thereby, only the conservation equations of dye and radiant energy of UV light are left to be concerned. The equation of continuity of dye in cylindrical coordinates for constant density (ρ) and diffusivity (D_{AB}) in the photooxidation system is shown as follows:

$$\frac{\partial C_{\text{dye}}}{\partial t} + \left(v_r \frac{\partial C_{\text{dye}}}{\partial r} + v_\theta \frac{1}{r} \frac{\partial C_{\text{dye}}}{\partial \theta} + v_z \frac{\partial C_{\text{dye}}}{\partial z} \right) = D_{AB} \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_{\text{dye}}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 C_{\text{dye}}}{\partial \theta^2} + \frac{\partial^2 C_{\text{dye}}}{\partial z^2} \right) - R_{\text{dye}} \quad (1)$$

where C_{dye} is the concentration of dye (mM), D_{AB} the binary diffusivity of dye in water ($\text{l}^2 \text{s}^{-1}$), R_{dye} is the photooxidation rate of dye ($\text{M s}^{-1} \text{l}^{-1}$).

Eq. (1) can be further simplified by the following assumptions.

1. The system was assumed to be steady-state, thus, the variation of accumulation is not considered.
2. Since an ideal plug flow is assumed, then $v_r = 0$, $v_\theta = 0$, and $v_z = V/\tau$ constant, where V is the space volume of the annular reactor, τ the retention time of the dye-containing solution within the photoreactor, v_z is the velocity of solution in z -direction a cylindrical coordinate.
3. The diffusion terms are considered to be trivial compared to the convection terms.
4. The intermediates generated during the photooxidation reaction will not interfere the photodecomposition behavior of the parent dye.

Therefore, the Eq. (1) is reduced to

$$v_z = \frac{\partial C_{\text{dye}}}{\partial z} = \frac{\partial C_{\text{dye}}}{\partial \tau} = -R_{\text{dye}} \quad (2)$$

For a plug flow reactor, the design equation can be written as follows:

$$\tau = \frac{V}{v_z} = - \int_{C_{\text{dye,in}}}^{C_{\text{dye,eff}}} \frac{dC_{\text{dye}}}{R_{\text{dye}}} \quad (3)$$

The photooxidation rate (R_{dye}) of the dye by UV/ H_2O_2 process represented in the form of the power law depends on the supplied UV light intensity (I) and the H_2O_2 dosage applied:

$$-R_{\text{dye}} = kI^m [\text{H}_2\text{O}_2]^n C_{\text{dye}}^p \quad (4)$$

where k is the rate constant, I the UV light intensity (W m^{-2}), $[\text{H}_2\text{O}_2]$ the H_2O_2 dosage (mM l^{-1}) and m , n , p are the orders with respect to I , H_2O_2 dosage and C_{dye} and can be determined by experiment.

For a fixed reactor size and UV light intensity, a simple first-order decrease in the concentration of the photoreactant with irradiation time is consistent with a simple direct photoreaction [18]. Nevertheless, the real value of the order, p , with respect to the concentration of dye needs to be resolved by experiments in this work as well as the orders of UV light intensity (m) and H_2O_2 dosage (n).

Because the reflection or refraction effect of light within the annular reactor is assumed to be insignificant, the conservation equations of radiant energy of UV light can be directly represented as the distribution of UV irradiance. In the infinite line source model, the UV irradiance, I , at any point within the reactor is related to the surface flux, I_0 , by a one-dimensional form of Lambert's law of absorption [19]:

$$I = I_0 \frac{r_i}{r} e^{-E(r-r_i)} \quad (5)$$

where E is the monochromatic absorbance of water using logarithms to the base e and is determined to be 0.624 cm^{-1} in this work. With the dye and H_2O_2 in the aqueous stream, Eq. (5) is modified as

$$I = I_0 \frac{r_i}{r} e^{-(E+\varepsilon_{\text{dye}}C_{\text{dye}}+\varepsilon_{\text{H}_2\text{O}_2}C_{\text{H}_2\text{O}_2})(r-r_i)} \quad (6)$$

Where the ε_{dye} and $\varepsilon_{\text{H}_2\text{O}_2}$ mean the molar absorption coefficients of the dye and hydrogen peroxide at 254 nm UV light and were determined to be 12.716 and $19 \text{ M}^{-1} \text{ cm}^{-1}$. The terms $\varepsilon_{\text{dye}}C_{\text{dye}}$ and $\varepsilon_{\text{H}_2\text{O}_2}C_{\text{H}_2\text{O}_2}$ shown in the exponent term in Eq. (6) represent the attenuation of radiation caused by the absorption of the dye and H_2O_2 . On combining Eqs (2)–(4) and (6) it gives

$$\begin{aligned} \frac{\partial C_{\text{dye}}}{\partial \tau} &= v_z \frac{\partial C_{\text{dye}}}{\partial z} = -R_{\text{dye}} \\ &= k \left[I_0 \frac{r_i}{r} e^{-(E+\varepsilon_{\text{dye}}C_{\text{dye}}+\varepsilon_{\text{H}_2\text{O}_2}C_{\text{H}_2\text{O}_2})(r-r_i)} \right]^m [\text{H}_2\text{O}_2]^n C_{\text{dye}}^p \end{aligned} \quad (7)$$

Integration of Eq. (7) for entire reactor length gives

$$\int_{C_{\text{dye},\text{in}}}^{C_{\text{dye},\text{eff}}} \frac{dC_{\text{dye}}}{C_{\text{dye}}^p e^{-\varepsilon_{\text{dye}}C_{\text{dye}}m(r-r_i)}} = k \left[I_0 \frac{r_i}{r} e^{-(E+\varepsilon_{\text{H}_2\text{O}_2}C_{\text{H}_2\text{O}_2})(r-r_i)} \right]^m [\text{H}_2\text{O}_2]^n \frac{L}{v_z} \quad (8)$$

where $C_{\text{dye},\text{in}}$ is the inlet concentration of dye (mM), $C_{\text{dye},\text{eff}}$ the effluent concentration of dye (mM). The integrated value of $C_{\text{dye},\text{eff}}(r)$ is a function of radial distance, r , because the extent of this photodecomposition reaction is radially decreased owing to the attenuation of UV light. The average residual concentration of dye, $C_{\text{dye},\text{eff},\text{ave}}$, in the effluent of photoreactor can be obtained from the following integration by numerical calculations:

$$C_{\text{dye},\text{eff},\text{ave}} = \frac{\int_0^{2\pi} \int_{r_i}^{r_o} C_{\text{dye},\text{eff}}(r) dr d\theta}{\int_0^{2\pi} \int_{r_i}^{r_o} r dr d\theta} = C_{\text{dye},\text{eff},\text{ave}} \left(I_0, C_{\text{dye}}, v_z \frac{r_i}{r_o} L, k, m, n, p \right) \quad (9)$$

With the necessary operating parameters (I_0 , C_{dye} , v_z), geometrical parameters (r_i/r_o , L) and reaction parameters (k , m , n , p), Eq. (9) can be used as the design equation for the photodecomposition of dye in the annular reactor.

4. Results and discussion

The decomposition of Direct Yellow 86 in aqueous solutions by UV/H₂O₂ process was examined under various UV light intensities, H₂O₂ dosages, and dimensions of photoreactor by a 254 nm low pressure mercury lamp to investigate the decomposition efficiency of dye within continuous annular photoreactors. The decomposition behavior of the dye was assumed to be first-order and irreversible reaction in a steady-state plug flow reactor. Thus, the pseudo first-order decomposition rate constant (k_1) of the dye can be calculated as

$$k_1 = \ln \frac{C_{\text{dye,eff}}/C_{\text{dye,in}}}{\tau} \quad (10)$$

Although hydrogen peroxide can be a relatively powerful oxidant, the oxidation of the dye by H₂O₂ alone was found to be very slow with the H₂O₂ dosage ranging from 50 to 200 mg l⁻¹ (dye/H₂O₂ molar ratio = 22–88) within the reaction time of 6.3 min studied in this work. At the reaction conditions, the decomposition of the dye by H₂O₂ alone was always below 1%, consequently the decomposition of dye molecules by UV/H₂O₂ process is mainly carried out by the attack of hydroxyl radicals generated from the photolysis of H₂O₂.

The flow behaviors of a liquid stream in a continuous reactor can be described by the vessel dispersion number $N_{\text{vd}} = D/uL$ [20], where D is the molecular diffusivity coefficient of the dye, u is the flow rate of the stream, and L is the length of the reactor. When the value of N_{vd} is very small, the flow behavior of the stream can be seen as an ideal plug flow and the flow behavior of the stream is close to a mixed flow when the value of N_{vd} is very larger. For a typical run of this work, the flow rate of the stream is 6.4 cm min⁻¹ and the length (L), the inner radius and the outer radius of the annular photoreactor, r_i and r_o are 40, 1.5 and 2.5 cm (R1), respectively. In the dye/H₂O₂ stream, the value of the molecular diffusivity coefficient of Direct Yellow 86 was determined to be 1.1×10^{-2} cm² min⁻¹ in this work. For the above reaction system described, the value of N_{vd} was calculated to be 4.3×10^{-5} that is quite small. Thus, the flow behavior of the dye/H₂O₂ stream can be described likely to be an ideal plug flow that corresponds with the assumption of the previous model of the photoreactor. Furthermore, the variation of the temperature during the reaction course of the photodecomposition system was found to be not noticeable (about kept at 25°C). As a result, it is reasonable to expect the reaction system is nearly isothermal as depicted as the postulation of the above photoreactor model.

The decomposition rates of 50 mg l⁻¹ Direct Yellow 86 at various surface UV light intensity by UV/H₂O₂ process are shown in Fig. 2. It was observed that more than 90% of the dye was decomposed at 102 W m⁻². The decomposition rate of dye was found to increase with increasing UV light intensity. But for the UV light intensity higher than 82 W m⁻², further increase of UV energy only slightly improved the decomposition efficiency of dye indicating the photons provided was excessive.

Fig. 3 illustrates the H₂O₂ dosage effect on the decomposition of the dye by UV/H₂O₂ process at the UV light intensity of 88 W m⁻². The decomposition rate of dye increased with increasing H₂O₂ dosage until the dosage reached 150 mg l⁻¹ (dye/H₂O₂ molar ratio = 66), further addition of H₂O₂ slight decreased the removal of dye. Similar results that the optimum H₂O₂ dosage exists in the UV/H₂O₂ oxidation system were reported by the previous researchers [8–10].

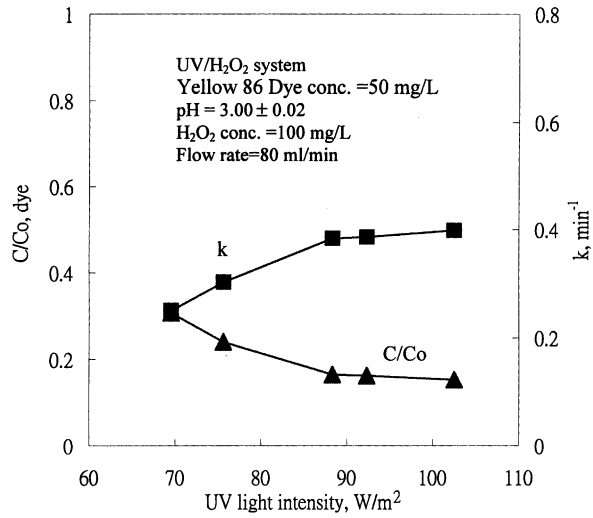


Fig. 2. The UV light intensity effect on the decomposition rates of Direct Yellow 86 by the UV/H₂O₂ process.

The results of the decomposition of dye conducted at various UV light intensity and dimensions of photoreactors are shown in Fig. 4 (where RT means the retention times, τ). The removal rates (represents as $[C/C_0]/RT$) of dye among the photoreactors were found to increase with decreasing the radius of the inner tube, r_1 , of the annular photoreactor using the same outer tube radius. While using the same inner tube radius, the removal

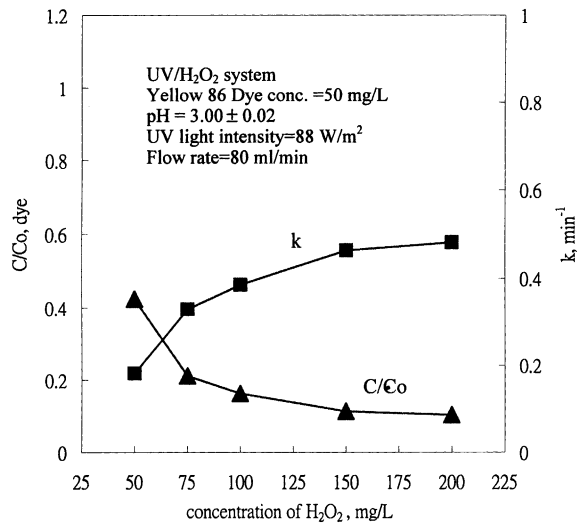


Fig. 3. The H₂O₂ dosage effect on the decomposition rates of Direct Yellow 86 by the UV/H₂O₂ process.

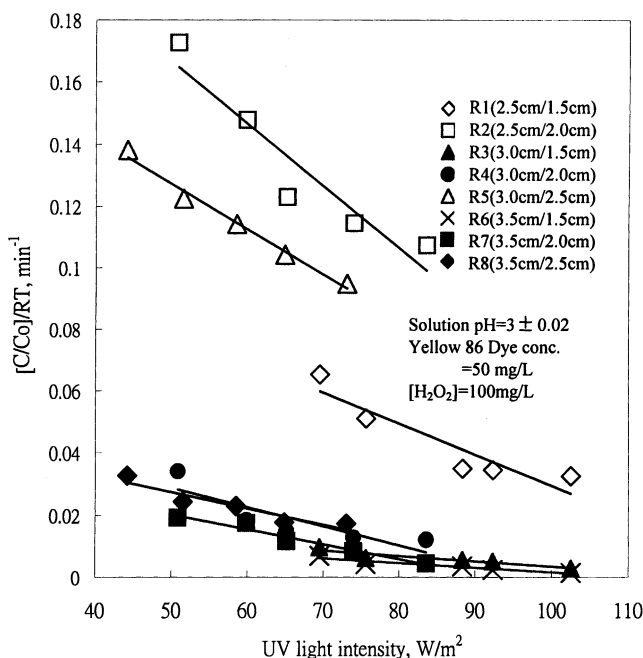


Fig. 4. The results of the decomposition of dye conducted at various UV light intensity and dimensions of photoreactors.

rates of dye increased with increasing radius of the outer tube, r_o . As shown in Fig. 4, the photodecomposition rate of the dye increased with decreasing value of ratio, r_i/r_o (correlated to Table 1). That is, a higher decomposition efficiency of the dye can be achieved by using an annular reactor with a wider gap (i.e. $r_o - r_i$) at the same retention time.

The values of the order, m , of UV light intensity in Eq. (4) for the decomposition of the dye by UV/H₂O₂ process within various dimensions of the photoreactors were regressed and shown in Table 3. The average value of m for the results using the various photoreactors was determined to be 1.02.

By regressing the Eq. (4) with respect to the experimental data of the effect of H₂O₂ dosage (see Fig. 3), the value of order (n) of H₂O₂ dosage was determined to be 0.67

Table 3

The values of the order (m) of UV light intensity for the decomposition of the dye by the UV/H₂O₂ process within various dimensions of the photoreactors

	Reactor (r_o/r_i)							
	R1 (2.5 cm/ 1.5 cm)	R2 (2.5 cm/ 2.0 cm)	R3 (3.0 cm/ 1.5 cm)	R4 (3.0 cm/ 2.0 cm)	R5 (3.0 cm/ 2.5 cm)	R6 (3.5 cm/ 1.5 cm)	R7 (3.5 cm/ 2.0 cm)	R8 (3.5 cm/ 2.5 cm)
m	1.15	0.99	0.83	0.97	0.75	1.13	1.30	0.72
r^2	0.8559	0.9290	0.8784	0.8394	0.9955	0.9463	0.9431	0.9317

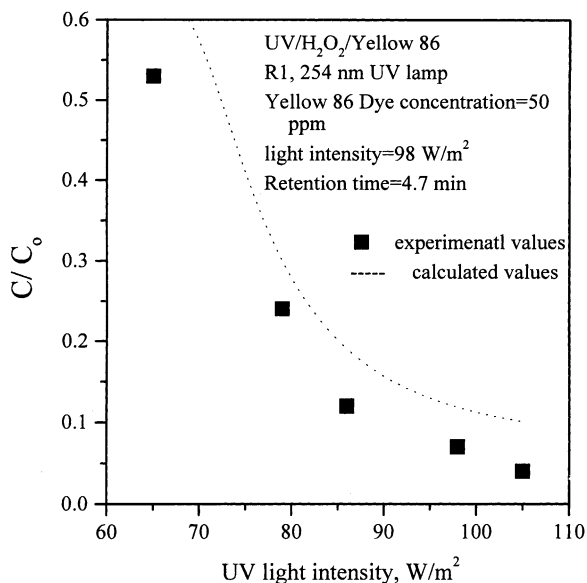


Fig. 5. The modeled results of the effect of UV light intensity on the decomposition of Direct Yellow 86 by Eq. (9).

($r^2 = 0.96$). The value of the order (p) of C_{dye} in Eq. (4) can be determined to be 1 because the pseudo first-order kinetics was assumed.

The modeled results of the effect on UV light intensity for the decomposition of Direct Yellow 86 by Eq. (9) with the calculated values of m , n and p are shown in Fig. 5. Fairly good agreements were found between the experimental results and calculated values indicating the proposed approach is reasonable on the photoreactor design for the decomposition of the dye. The calculated reaction rates were found to be somewhat less than the experimental data possibly because of some oversimplified assumptions of photoreactor characteristics and reaction mechanism.

5. Conclusions

The results obtained have shown that the continuous UV/H₂O₂ process was capable of efficiently decomposing the Direct Yellow 86 dye wastewater. A higher decomposition efficiency of the dye can be achieved by using an annular reactor with a wider gap at the same retention time. By combining the empirical rates and conservation equations, it has demonstrated the successful application of a semi-empirical design equation to predict the treatment performance of an annular photoreactor for the decomposition of Direct Yellow 86 in aqueous solutions by UV irradiation in the presence of H₂O₂. Although the established equation needs more verifications, the methodology seems to be reasonable and can be applied for various photoreaction systems.

Acknowledgements

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