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A comparative study on reaction kinetic of textile wastewaters degradation by UV/TiO₂ and UV/ZnO

Mojtaba Ahmadi*, Pegah Amiri

Chemical Engineering Department, Faculty of Engineering, Razi University, Kermanshah, Iran.

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ABSTRACT

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A comparative kinetic study of photocatalytic degradation of textile wastewater by UV/TiO₂ and UV/ ZnO for removal of chemical oxygen demand (COD), and color was carried out. The effects of some parameters such as the initial concentration of catalyst, initial COD concentration and light intensity on the photocatalytic process were also examined. It was demonstrated that the COD removal by TiO₂/UV and ZnO/UV was about 49 %, 33.3 %, and color removal was 30 % and 10 %, respectively. The experiment demonstrated that the photodegradation efficiency of TiO_2 was significantly higher than that of ZnO. On the other hand, the kinetic study shows that decomposition of chemical oxygen demand follows a first-order for processes. The rate of degradation is highly dependent on the initial concentration of TiO₂, ZnO and light intensity. A comparison between experimental and calculated degradation rate constants shows that TiO₂/UV process gives better results than photocatalytic treatment. Maximum degradation rate was achieved for TiO₂/UV at optimum concentrations of TiO₂ ©2016 Razi University-All rights reserved.

1. Introduction

Textile industry is one of the largest industries in various parts of the world. Water pollution by the textile mills is the result of wastewater discharge from various stages of production like desizing, scouring, bleaching, mercerizing, dyeing, and printing (Barka et al. 2011). The colored wastewater discharged from a textile industry exhibits low biochemical oxygen demand (BOD), high values of COD, changeable pH, suspended solids, and organic chlorine compounds (Lee and Yoon. 2004). An important characteristic of the textile mills is the use of different types of dye (Aslam et al. 2004). Thus, removal of color is the most difficult constituent of the textile wastewater (Oguz and Keskinler. 2008). The physical, chemical and mostly biological technologies that have been widely used to treat textile effluents are: advanced oxidation methods, physic-chemical methods like adsorption (Belaid et al. 2013; Rangabhashiyam et al. 2013), electro-catalytic treatment (Ibrahim et al. 2013), and biological sludge methods (Wu et al. 2008).

Advanced oxidation processes (AOP) such as ozonation, UV and ozone/UV combined oxidation, photo catalysis (UV/TiO2), Fenton's reagent, and ultrasonic oxidation are based on the production of hydroxyl radicals as oxidizing agents to mineralize organic chemicals (Blanco et al. 2012). The reaction mechanism of organic compounds with hydroxyl radicals is very complex, so that the mechanism can be briefly described in three stages including: 1. initiation reaction: in this process, free radicals are produced. 2. propagation reaction: the radicals generated in the previous step are converted to other free radicals, and 3. termination Reaction: in this process, the free radicals produce a stable compound (Lin et al. 1998). Among the different existing methods, heterogeneous photocatalysis has been shown to be successful and beneficial for degradation of wastewater organic pollutants (Yahiat et al. 2011). In photocatalysis systems, combination of semiconductors (e.g. TiO₂, ZnO, ZnS, WO₃, CdS and SrTiO₃) and UV or visible lights can be used. Semiconductors are characterized by two separate energy bands: a low-energy valence band and a highenergy conduction band. Photon energy, hv, with a wavelength of 387.5 nm can be used to excite an electron from the valence band into the conduction band (Al-Momani et al. 2002; Ren et al. 2010). The hole generated can degrade organic pollutants in wastewater to CO2 and H_2O_2 (Aguedach et al. 2005). TiO_2 and ZnO among the semiconductors that have been most studied, present good features of stability, nontoxicity and insolubility (Yeber et al. 2000). In addition, since ozone is a *Corresponding author Email: m_ahmadi@razi.ac.ir

powerful oxidant in textile wastewater treatment, it generally used to remove stable organic compounds and dyes from industrial effluents (Langlais et al. 1991). With dissolving ozone in water, it oxidizes the organic compounds in two different ways: one way is by a direct reaction of molecular ozone, and the other way is through free radicals as OH radicals produced by ozone decomposition in water (Perkowski et al. 1996; Baig and Liechti. 2001; Soares et al. 2006; Somensi et al. 2010). Advanced oxidation processes have a significant impact on the ability to reduce COD and treatment of non-biodegradable pollutants. As a result of better access to these methods, the processes can be combined with biological treatment (Tabrizi and Mehrvar. 2004; Wang et al. 2009).

Among the advanced oxidation processes the homogeneous AOPs employing Titanium dioxide photocatalysis process (Ghezzar et al. 2009; Foo and Hameed. 2010), UV/TiO₂ (Yu et al. 2010; Botía et al. 2012; Gupta et al. 2012), ZnO (Chakrabarti and Dutta. 2004; Ahsan Habib et al., 2012) and ozone (Soares et al. 2006; Oguz and Keskinler. 2008; Wu et al. 2008; Somensi et al. 2010) have been found to be very effective to degrade dye and pollutants. Ghezzar et al. (2009) studied the bleaching and textile effluent degradation with the plasma-catalytic process. The best conditions of experimental parameters for the removal of textile wastewaters have been determined and the wastewater was completely decolorized after only 30 min (Ghezzar et al. 2009). The research of Akyol et al. (2004) was conducted using ZnO for degradation of Remazol Red RR dye (Akyol et al. 2004). Peralta-Zamora et al. reported the treatment of effluent from the cellulose and textile industries by applying a heterogeneous photocatalytic procedure using ZnO, TiO₂ by supported ZnO on the photoassisted (Peralta-Zamora et al. 1998). The results showed that the treatment of textile effluents by UV-irradiation in the presence of free TiO₂ and ZnO or silica gel supported ZnO, the color fades progressively to reach substantial decolorization ratios. Using of ozone as pretreatment of textile wastewater in a pilot-scale plant was investigated by Somensi et al. (2010) ozonation was enhanced the biodegradability of textile wastewater (BOD₅/COD ratios) by a factor of up to 6.8-fold (Somensi et al. 2010). Kinetic study of decolorization C.I. Basic Blue 3 with immobilized TiO₂ nanoparticles photocatalytic was studied by Khataee and Mirzajani et al. (2010). The kinetic characteristics of the photocatalytic degradation of BB3 by using immobilized titanium dioxide nanoparticles were experimentally investigated (Khataee and Mirzajani. 2010). Gupta et al. (2012) investigated mineralization of toxic

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dye amaranth on TiO₂/UV. The decolorization and degradation kinetics were investigated followed a pseudo first order kinetics with regards to the substrate concentration under the experimental conditions (Gupta et al. 2012). The research of Bensaadi et al. (2014) was conducted by using photodegradation with TiO₂ for mineralization of acebutolol. The results showed that the photodegradation follows a pseudo-first-order kinetic.

The aim of this study is to compare photocatalysts techniques by TiO₂ and ZnO for reduction of color and COD textile wastewater. The primary focus is to study the effect of TiO₂ and ZnO concentration, light intensity, initial concentration of COD in on the photocatalytic system. In the literature, only a limited number of studies have so far been focused on the kinetic analysis of degradation textile wastewater so the experimental data were also analyzed by using the first-order kinetic model.

2. Materials and methods 2.1. Material

The used wastewater for the laboratory-scale was obtained from the textile plant. The characteristics of the untreated wastewater are given in Table 1. The performance of two catalysts was compared in the photocatalytic Treatment. TiO2 (P-25) photocatalyst was purchased from Degussa Co. Ltd., Germany (anatase 75 %, rutile 25 %, BET specific surface area 48 m²/g, and mean particle size 25 nm) and ZnO purchased from Merk Co.Ltd., Germany. The absorbance at specific wavelengths of the supernatants was concluded by using ultra violet spectrophotometer, visible (model UV-2100 UV/Vis Spectrophotometer). Decolorization efficiency (DE) was estimated from a mathematical equation based on calculation of decolorization used before:

$$DE = \frac{(absorbance)_0 - (absorbance)_t}{(Absorbance)_0} \times 100$$

where $(absorbance)_0$ and $(absorbance)_t$ are the absorbance before irradiation and the absorbance at time t, respectively.

2.2. Experimental set-up 2.2.1. Photoreactor

The photocatalytic experiments used to photodegradation of textile wastewater were performed in a Pyrex glass vessel photoreactor equipped with an aeration system, as shown in Fig. 1. The photoreactor was illuminated by a 125 W mercury lamp with a peak light intensity at 254 nm located in the distance of 12 cm from center of suspension surface. The temperature of the suspension in the photoreactor was kept constant at 30 0C by circulating water in bathroom water, and the irradiation time was 2 h. The photocatalytic degradation researches were done by loading 500ml of the wastewater solutions in the photocatalytic reactor. The effects of initial concentration of textile wastewater and photocatalyst concentrations were examined by using different initial COD concentrations (500-1650 mg/l) and varying

amount of the photocatalyst from 0.125 to 1 mg/l with an initial COD concentration of 1650 mg/l, respectively. Tests under two 125 W and 250 W light sources were separately done and also sampled once every 30 minutes. The samples were taken from the reactor at scheduled times, and COD was measured.



Fig. 1. Schematic of photocatalytic reactor.

3. Results and discussion

3.1. UV/ TiO₂ photocatalytic process

3.1.1. Effect of initial textile wastewater concentration

The effect of initial concentration of the textile wastewater on the COD removal is shown in Fig. 2. The figure indicates that the initial concentration of the textile wastewater is an effective parameter on removal efficiency. The decrease in photooxidative degradation with increasing initial textile wastewater concentration has been observed. As increasing of the concentration from 500 to 1650 mg/l, the decolorization efficiency decreased from 80 % to 49 %. The slope of the plots increases with decreasing initial concentration, i.e., when initial concentration increased, the catalyst efficiency decreased. It was noticed that the maximum percentage of removal occurred when initial concentration was 500 mg/L. The higher textile wastewater concentration, and the more absorption of UV light are the presumed reasons. Photoreactor solution is more resistant to UV radiation (Sleiman et al. 2007; Barka et al. 2010).

Table 1. Characteristics of textile wastewater.

Parameters	Value	
COD (mg O ₂ L ⁻¹)	1650	
BOD (mg $O_2 L^{-1}$)	390	
BOD/COD	0.23	
рH	11	
Color (A ₃₃₅)	0.81	



Fig. 2. Effect of initial COD concentration on TiO₂/UV treatment (TiO₂ concentration: 0.25 g/l, Light source: 250 W).

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3.1.2. Effect of initial concentration of TiO₂

The initial concentration of TiO₂ was found to be an important parameter for the photooxidative photocatalysis process of textile wastewater in the UV/TiO₂ process. The effect of initial TiO₂ concentration on photodegradation efficiency has been expressed in Fig. 3. As the concentration of TiO₂ increased, the photocatalytic efficiency increased proportionally. Also, at higher concentrations, this beneficial effect tends to level off and then decreases because of the screening effect of excess particles which prevent of the photosensitive surface active sites and impediment the UV irradiation(Sleiman et al. 2007). On the other hand, the photocatalytic efficiency on color removal was observed 27 and 30 % for light sources 125 and 250 W respectively. As seen in Fig. 3, the COD removal increases with increasing light power. It is hoped that kinetic modeling and studies will explain the effect of the light intensity on the photocatalytic efficiency.

3.1.3. Kinetics analyses

The kinetics of the photocatalytic degradation rate of most organic com pounds is described by pseudo-first order kinetics:



where t is time, C is the COD concentration, and K_{app} is the apparent rate constant. By integrating the equations (1) with the initial condition $C(0){=}C0$, the following equations could be obtained:

$$\ln\left(\frac{C}{C_0}\right) = -K_{app}t\tag{2}$$

Regression analysis based on the first-order reaction kinetics for the photocatalytic degradation of textile wastewater in the UV/TiO2 process was conducted, and the results were shown in Fig. 4. The results indicated that the photodegradation kinetics of textile wastewater followed the first-order kinetics well. The rate constants at different initial TiO₂ concentration were obtained and the results were shown in Table 2. Increasing TiO₂ particles prevents UV light from the light source, so COD removal was reduced. The results indicate that 0.25 g/l of TiO₂ is sufficient for the maximum rate of decolorization and COD removal. The proposed kinetic model is also in good agreement with our experimental data. Increasing the light intensity from 125 to 250 W could increase the kinetic rate constants of UV/ TiO2 photocatalytic process. This observation agrees to the results of other studied (Khataee et al. 2011). It is reasonable, since the light intensity increased, more hydroxyl radicals are available to attack the dye pollutants and the photocatalytic reaction rate constant increases.



Fig. 3. Effect of initial concentration of TiO₂ for different light intensity (initial COD: 1650mg/l, temperature: 30 °C).

3.2. UV/ ZnO photocatalytic process 3. 2.1. Effect of initial concentration of ZnO

Effect of catalyst mass on COD removal efficiency of real textile industrial wastewater has also been investigated by employing different masses of ZnO varying from 0.125 to 1 g/l under UV irradiation, and the presented results in Table 3 are plotted in Figs. 5 and 6 shows results curves of rate constant. K and R² calculated values are given in Table 4. As can be seen, the removal rate increased with increasing of ZnO particles. The results indicate that 1g/l of ZnO is sufficient for the maximum rate of removal COD increasing of initial concentration of ZnO increases the catalyst surface and light absorption by the catalyst surface. Akyol et al. (2004) studied the effect of the catalyst loading in photodegradation of Remazol Red RR in aqueous ZnO suspensions. They commented that the decolorization efficiency increases by increasing loading of ZnO.

The results of the effect of titanium dioxide (anatase and rutile) and zinc oxide masses on the photocatalytic decolorization of real and simulated textile dyeing wastewater shows that the decolorization efficiency is increased with increasing in the mass of catalysts, and it becomes constant at a certain mass, and then starts to decrease with increasing in the mass of catalyst further. The increasing of catalytic activity with increasing in the mass of catalyst was explained due to increasing availability of photocatalysts sites, and the decreasing of catalytic activity after the plateau region is related to increasing of light scattering due to the excess of catalyst.

3.2.2. Effect of light intensity

The rate of photocatalysis activation, and electron-hole formation in photochemical reaction is strongly dependent on light intensity (Cassano and Alfano. 2000). For studying the influence of UV intensity on photocatalytic efficiency, different UV source lamps with powers of 125 and 250 W were used. All experiments were created in same conditions, so the obtained results are shown in Figs. 3 and 5 obviously, with increasing the UV light intensity from 125 to 250 W the removal COD efficiency was increased. Generally, the light intensity of the solution had a significant effect on kinetics of the reactions. It was observed that by an increase in light intensity, an increase in rate constant occurred. This can be due to leading to more catalyst activation and more photolcatalytic degradation of textile wastewater.



Fig. 4. Effect of initial TiO₂ concentration on decomposition rate constant (initial COD: 1650 mg/l, temperature: 30 °C).

 Table 2. The first-order kinetic rate constants of UV/ TiO₂ photocatalytic process at 30 °C and under different initial TiO₂ concentration.

TiO	UV source lamp (W)				
TiO₂ (g/l)	125		250		
(g/i)	K (1/min) R	R ²	K (1/min)	R ²	
0.125	0.0027	0.987	0.0032	0.934	
0.25	0.0048	0.958	0.0060	0.965	
0.5	0.0032	0.992	0.0043	0.955	
1	0.0038	0.985	0.0023	0.985	



Fig. 5. Effect of initial concentration of ZnO for different light intensity.

Table. 3. Color removal percent after ZnO/UV process.				
	Color (A ₃₃₅)	Color removal (%)		
Raw wastewater	0.81			
After (ZnO/UV: 250 watt)	0.72	10		
After (ZnO/UV:125 watt)	0.74	8		

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Fig. 6. Effect of initial ZnO concentration on decomposition rate constant.

Table 4. Rate constants of UV/ ZnO photocatalytic process at 30 °C and under different initial ZnO.

TIO	UV source lamp (W)				
TiO ₂	125		250		
(g/l)	K (1/min)	R ²	K (1/min)	R ²	
0.125	0.001	0.953	0.0010	0.954	
0.25	0.0014	0.995	0.0018	0.971	
0.5	0.0016	0.987	0.0027	0.986	
1	0.0027	0.989	0.0032	0.989	

4. Conclusions

In this study, photocatalytic processes for the removal of dyes and organic load of textile effluent were examined. The results show that the degree of photodegradation of textile industrial wastewater are obviously influenced by several parameters. Under optimal conditions, the extent of decolorization and COD removal were achieved about 80 % by using TiO₂, and 37 % by using ZnO at 303 K, respectively. The kinetic study shows that COD removal follows first-order models

adequate to describe the kinetic of biodegradation process. It is clear that mineralization of the total organic content of the textile wastewater is not economical during photocatalytic in a pilot-scale, but partial oxidation altering the original product can have beneficial effects by reducing toxicity effects of wastewater, and enhancing the biodegradability of organic wastewater contents in a sequential microbiological treatment of textile wastewaters.

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