

Research Article

Decolorization of Methylene Blue with TiO₂ Sol via UV Irradiation Photocatalytic Degradation

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TiO₂ sol was prepared for the degradation of methylene blue (MB) solution under ultraviolet (UV) irradiation. The absorption spectra of MB indicated that the maximum wavelength, 663 nm, almost kept the same. The performance of 92.3% for color removal was reached after 160 min. The particle size of TiO₂ sol was about 22.5 nm. X-ray diffraction showed that TiO₂ consisted of a single anatase phase. The small size and anatase phase probably resulted in high photocatalytic activity of TiO₂ sol. The degradation ratio decreased as the initial concentration of MB increased. The photodegradation efficiency decreased in the order of pH 2 > pH 9 > pH 7. Regarding catalyst load, the degradation increased with the mass of catalyst up to an amount of 1.5 g · L⁻¹ then decreased as the mass continued to increase. The addition of H₂O₂ to TiO₂ sol resulted in an increase on the degradation ratio.

1. Introduction

The textile industry produced dye pollutants that were becoming a major source of environmental contamination. It was estimated that 10 to 15 percent of dyestuff without treatment was released in water during dyeing processes [1]. A variety of physical, chemical, and biological methods, such as adsorption, coagulation, membrane process, and oxidation-ozonation were presently available for treatment of dye wastewater [2–4]. The conventional processes were insufficient to purify the wastewaters. They just transferred the compounds from aqueous to another phase, thus causing secondary pollution problem [5, 6]. Semiconductor photocatalysts TiO₂ was acting much interest for their potential applicability in degradation of dye pollutants [7–10]. The TiO₂ catalyst could transform organic pollutants into biodegradable compounds of low molecular weight.

Many papers [11–15] reported that TiO₂ was prepared by the sol-gel progress for it was nonselective, room-temperature and inexpensive manufacturing cost. Huang et al. [16] prepared TiO₂ sol by a sol-gel method. The microstructure and morphology of sol sample were characterized. Its performance for photodegradation of formaldehyde was investigated. However, the operational

effects, such as catalyst concentration, initial formaldehyde concentration, and pH, in the degradation process were not considered. The TiO₂ sol usually contained organic solvent as a major constituent [17–20]. Silva and Faria [21] first mixed tetrabutyl titanate, absolute alcohol, and polyethyleneglycol. Few quantity of deionized water was added and the volume fraction of organic solvent reached almost 98.2%. The degradation of TiO₂ prepared using this method was only around 60%. The organic solvent slowed down the hydrolysis rate of tetrabutyl titanate and then decreased the photocatalytic activity of TiO₂ [22]. It also resulted in more serious environmental pollution [23, 24]. To overcome the disadvantages, the use of aqueous solution instead of organic solvent was desired.

In this paper, aqueous base titanium sol was synthesized via a sol-gel method. Its photocatalytic activity was evaluated on the degradation of MB. The effects of key experimental parameters were investigated.

2. Experimental

2.1. Materials. The absolute ethanol (EtOH, AR), Hydrochloric acid (HCl, AR), Hydrogen Peroxide (H₂O₂, AR), and methylene blue (MB) were purchased from

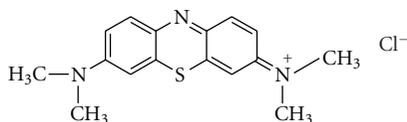


FIGURE 1: Chemical structure of MB.

Sinopharm Chemical Company (China). Tetrabutyl titanate ($\text{Ti}(\text{OC}_4\text{H}_9)_4$, CP) was chosen as a precursor. The chemical structure of MB was shown in Figure 1. The water used in the experiments was double distilled and deionized.

2.2. Preparation of TiO_2 Sol. 8 mL $\text{Ti}(\text{OC}_4\text{H}_9)_4$ was dissolved in 4 mL EtOH. After stirring vigorously by a dynamoelectric stirrer for 10 min at room temperature, it was added dropwise to the mixed solution of H_2O and HCl ($1 \text{ mol} \cdot \text{L}^{-1}$) with a burette under stirring. An aqueous solution of H_2O_2 was then added to the mixture, and clear sol was obtained after 7 days. The mole ratio of $\text{Ti}(\text{OBU})_4 : \text{H}_2\text{O} : \text{C}_2\text{H}_5\text{OH} : \text{HCl}$ was about 1.3 : 320.6 : 3.9 : 1. The water quality content of the sol was about 90%.

2.3. Characterization. The Zeta potential of reaction solution and particle size of TiO_2 sol were measured with the Zeta potential and particle size analysis apparatus (Nano-ZS90, Malvern Co., UK), respectively.

The crystalline phase of TiO_2 was studied by X-ray diffractometer (Rigaku Corporation, Japan) with $\text{Cu K}\alpha$ radiation. A 2θ range of 15° to 60° was scanned with a step of 0.02° using an integration time of 3 seconds.

UV-vis spectroscopy of the TiO_2 sol was recorded on a UV-vis spectrophotometer (Puxi Tongyong Apparatus Ltd. of Beijing TU-1901) with wavelength range of 500–700 nm.

2.4. Photocatalytic Activity Tests. The photocatalytic activity of TiO_2 sol was evaluated on the degradation of MB in an aqueous solution under illumination of UV light (mercury vapor lamp, 40 W, 40 cm long, predominantly wavelength 253.7 nm) in a photoreactor system. The UV lamp was fixed down below the top of the reactor and vertically hung. Four test tubes contained with reaction solution were equably around the lamp. The depth of the solution was 8 cm and the internal diameter of test tube was 1.6 cm. The distance from test tubes to lamp was 10 cm. In a typical experiment, the TiO_2 sol was added into aqueous solution of MB, and the initial MB concentration and the amount of TiO_2 were set at $0.2 \text{ g} \cdot \text{L}^{-1}$ and $1.5 \text{ g} \cdot \text{L}^{-1}$, respectively. The reaction time ranged from 0 to 160 min. The degradation of MB was calculated by formula: $\text{Degradation} = (A_0 - A)/A_0$, where A_0 and A were the absorbance of the primal and remaining MB, respectively. The absorbance was measured with UV/vis spectrophotometer (UV-2100, China).

3. Results and Discussion

3.1. Absorption Spectra of MB Degradation. Figure 2 displayed maximum UV-vis absorption at wavelength of

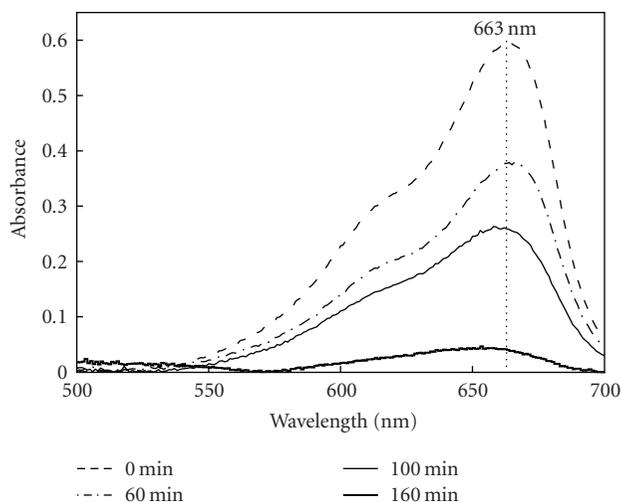


FIGURE 2: UV-vis absorption spectra of MB solution catalyzed by the TiO_2 sol.

663 nm. The maximum absorption almost kept the same during the photodegradation progress. This indicated that it was a sign of MB concentration level after degradation. So it was possible to measure the absorbance at 663 nm each time, and the resulting data of degradation ratio were valid. The absorption peak of the spectra rapidly decreased with increased time and almost disappeared for 160 min light irradiation as seen from Figure 2. The chromophores responsible for characteristic color of the MB were broken down and MB had been degraded. The maximum absorption value and calculated degradation ratio were shown in Table 1 COD values of MB were presented in Table 2. The performance of 92.3% for color removal and 71.4% for COD value removal were reached, which indicated that TiO_2 sol prepared in the experiment had high photocatalytic activity.

3.2. Particle Size and Its Distribution of TiO_2 Sol. The nanometer particle size was probably one of the reasons for the high photocatalytic activity of the catalyst. Figure 3 showed the particle size of TiO_2 sol. It distributed between 10 nm and 80 nm and the mean particle size was 22.5 nm. The small crystal size of the TiO_2 sol caused blue shifts, which was consistent with findings reported earlier [25, 26]. The blue shift probably resulted in an increment of reduction potential for the electron and oxidizing potential for the hole. The electrons and holes with high reduction and oxidation power increased the photodegradation ratio. Particle size also had an influence on the surface area of the reactive species. The decrease of the particle size caused an increase of specific surface area, which was beneficial to accelerate the photocatalytic reaction.

3.3. The Crystallinity of TiO_2 Sol. The crystalline phase affected the activity of TiO_2 . It was observed from Figure 4 that a broad peak at $2\theta = 25.3^\circ$ (corresponding to (101)) was observed. The rutile diffraction peak at $2\theta = 27^\circ$ of TiO_2 film (corresponding to (110)) was not found. These indicated

TABLE 1: The maximum absorbency of MB during the photodegradation progress.

Irradiation time (min)	0	60	100	160
MB maximum Absorbency	0.599	0.383	0.264	0.046
Degradation ratio (%)	0	36.1	56.0	92.3

TABLE 2: COD values of MB solution during the photodegradation progress.

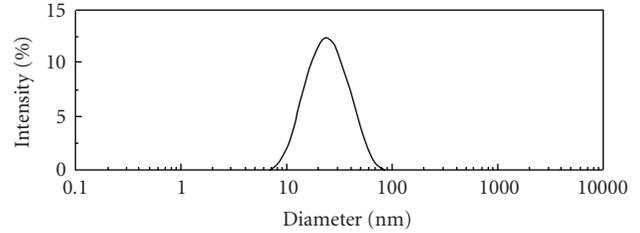
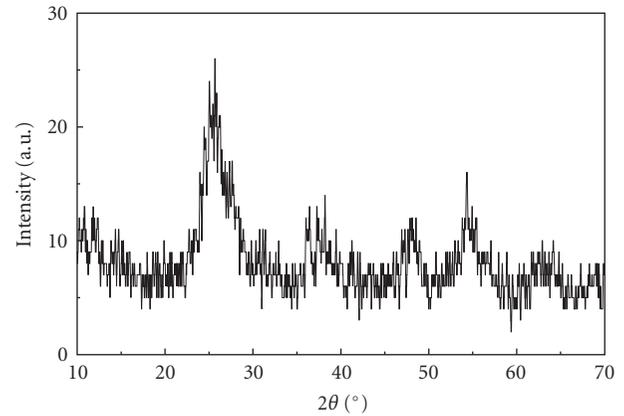
	Before degradation	After degradation	COD removal (%)
COD (mg·L ⁻¹)	112	32	71.4

that TiO₂ thin film without high-temperature treatment was mainly based on the existence of anatase phase. Between the two crystalline phases, anatase was proved to have better optoelectronic properties [27]. Simultaneously, the full width at half maximum of XRD peak corresponded to the crystal size of TiO₂ catalyst. Scherrer formula was used for the calculation of crystallite size [28]. The size was about 23.8 nm. Both the anatase phase and small size improved the photocatalytic activity of TiO₂ sol.

3.4. Photocatalytic Activity of TiO₂ Sol

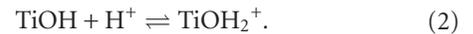
3.4.1. Effect of Initial Concentration of Dye. The initial concentration of MB influenced the UV light absorption of TiO₂ catalyst. It could be seen from Figure 5 that MB was degraded slowly at a high concentration. As the MB concentration increased, some of the UV light photons were absorbed by the substantial amount of MB molecules. The quantity of effective photons which was absorbed by the surface of catalyst was reduced. The quantity of excited TiO₂ electrons produced by effective photons decreased, making the generating holes lessened. Simultaneously, a large amount of impurities and other organic substances were probably adsorbed on the surface of TiO₂, making it more difficult for holes to enter the solution. So fewer and fewer holes entered the MB solution and reacted with adsorbed hydroxide ions to produce hydroxyl radicals, the main oxidizing species responsible for the photooxidation of organic compounds [29]. It was noted that the degradation of MB solution decreased with the increasing concentration. The almost complete degradation took place for dye concentrations at 0.20 g·L⁻¹.

3.4.2. Effect of pH Value of the Solution. Solution pH was an important variable in the evaluation of aqueous phase mediated photocatalytic degradation reactions. It influenced the surface electric charge of TiO₂ catalyst. Figure 6 revealed that the photodegradation efficiency decreased in the order of pH 2 > pH 9 > pH 7. Rideh et al. [30] concluded that the interaction of TiO₂ with cationic electron donors and electron acceptors would be favored for photocatalytic activity at high pH greater than the zero point charge (ZPC) of TiO₂, while anionic electron donors and electron

FIGURE 3: Particle size and its distribution of TiO₂ sol.FIGURE 4: XRD pattern of TiO₂.

acceptors would be favored at low pH less than ZPC. ZPC of TiO₂ was found to be 6.25.

The reaction of surface charge of TiO₂ catalyst performed in alkaline and acid media as follows:



Equation (1) showed that at pH higher than pH_{ZPC}, TiO⁻ appeared to be the predominant form. The zeta potential of the sample was -25.49 mV as shown in Figure 7 (a), which indicated that TiO₂ was negatively charged. The catalyst absorbed MB molecules with electropositive charge based on the law of electrostatic attraction, which made the degradation increased. The final degradation ratio was 33.8% at pH 9, much higher than 18.0% at pH 7 as shown in Figure 6. Regarding pH values of 2, the average zeta potential of TiO₂ was approximately 19.04 mV in Figure 7(b). Because TiO₂ was in the protonated form (TiOH₂⁺) as shown in (2), TiO₂ catalyst had an effect of electrostatic repulsion against the MB molecules.

However, the effect on MB degradation was only in a low degree. In acidic media, electrons tended to move into the surface of catalyst for the electrostatic attraction between the positive charged TiO₂ and negative charged electrons. They reacted with the oxygen molecules absorbed around the TiO₂ surface to form oxidizing species such as •O₂⁻ and •OOH.

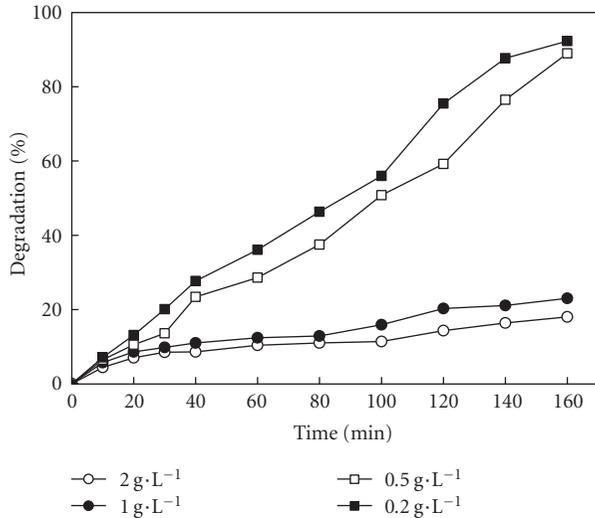


FIGURE 5: Effect of dye concentrations on photodegradation.

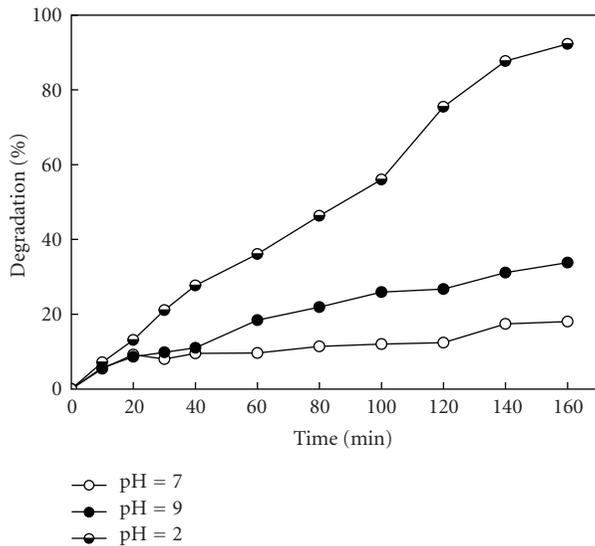
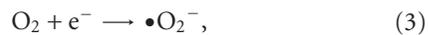


FIGURE 6: Effect of solution pH on photodegradation.

The reaction formulas were as follows [31]:



These oxidizing radicals made the degradation accelerate, which played a more important role than the electrostatic effect discussed above in the reaction progress. Thus, the catalytic activity of TiO_2 sol improved in alkaline and acid media, especially in acid media.

3.4.3. Effect of Catalyst Load. In order to avoid an ineffective excess of catalyst and to ensure a total absorption of efficient photons, the fitting mass of catalyst needed to be found. The concentration was varied from 0.5 to 2.5 $\text{g}\cdot\text{L}^{-1}$ in this study. In Table 3, the degradation ratio increased with the mass of

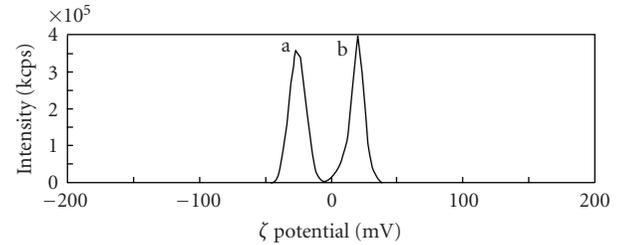


FIGURE 7: Zeta potential of MB solution (a) pH = 9; (b) pH = 2.

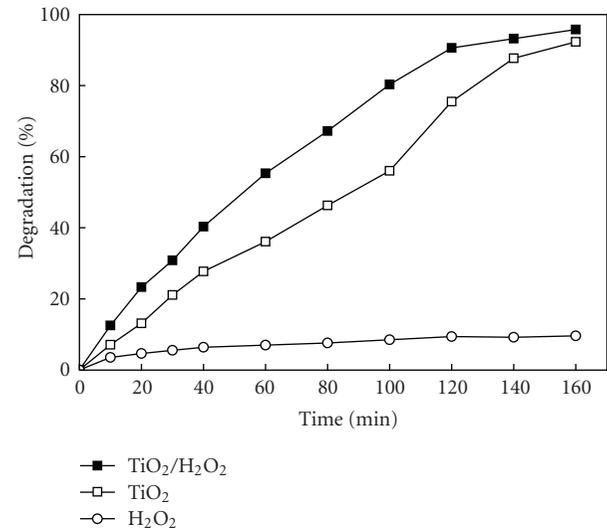
FIGURE 8: Effect of added H_2O_2 on photodegradation.

TABLE 3: Effect of catalyst load on photodegradation.

C_{TiO_2} ($\text{g}\cdot\text{L}^{-1}$)	MB Absorbency before reaction (A_0)	MB Absorbency after reaction (A)	Degradation ratio (%)
0.5	0.544	0.313	42.5
1.0	0.560	0.146	74.0
1.5	0.599	0.046	92.3
2.0	0.635	0.091	85.6
2.5	0.683	0.134	80.4

catalyst up to an amount of 1.5 $\text{g}\cdot\text{L}^{-1}$ for an increment of the active sites available for MB adsorption and degradation. However, an increase on the catalyst loading to 2.0 and 2.5 $\text{g}\cdot\text{L}^{-1}$ resulted in a decrease in the degradation ratio. This could be attributed to deactivation of activated molecules by collision with ground state molecules of TiO_2 . Following these observations, it was decided to keep the amount of TiO_2 at the value of 1.5 $\text{g}\cdot\text{L}^{-1}$ in subsequent photocatalytic degradation experiments.

3.4.4. Effect of Added H_2O_2 . Hydrogen peroxide played a dual role in photocatalytic reaction acting as electron acceptor and could decompose to produce $\bullet\text{OH}$ radicals. As shown in Figure 8, the addition of H_2O_2 (0.03 $\text{mol}\cdot\text{L}^{-1}$) to TiO_2 sol

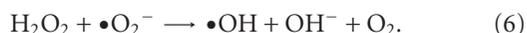
resulted in a significant increase on the degradation ratio of MB. At the end of 160 min of irradiation total removal, the degradation ratio of TiO₂ with H₂O₂ reached 95.8%, more than 92.3%. The decolorization of MB by H₂O₂ itself was in a low degree. The degradation ratio was only 9.6%.

Photocatalytic reaction major energy wasting step consisted in the recombination of photo-generated electrons and holes leading to the low quantum yield of the process. Electron-hole recombination could be prevented by adding a proper electron acceptor to the system. So the addition of H₂O₂ to TiO₂ sol was considered. H₂O₂ showed a positive effect on the degradation by avoiding electron/hole recombination, as it acted as electron acceptors.

Adding H₂O₂ was also a possible way to increase the concentration of •OH radicals to increase the degradation as shown in the following [32]:



Hydroxyl radicals could as well be produced by reaction of H₂O₂ with superoxide radical anion as follows:



4. Conclusion

TiO₂ sol synthesized via a sol-gel method was used in the photocatalytic degradation of MB under UV light. The maximum wavelength of MB in the absorption spectra was 663 nm and almost kept the same. The absorption peak decreased during the degradation progress. The particle size of TiO₂ sol was 22.5 nm, which was one cause of the high photocatalytic activity of the sol. The high initial concentration of MB interfered with the penetration of UV light transmission. Fewer holes reacted and resulted in a decrease in the degradation ratio. The activity of TiO₂ sol improved in alkaline media due to the electrostatic attractions between oppositely charged MB molecules and negatively charged TiO₂ catalyst. In acidic media, more electrons reacted to form oxidizing species to increase the degradation. Regarding catalyst load, the degradation increased with the mass of catalyst up to an amount of 1.5 g·L⁻¹. It decreased as the mass continued to increase attributed to the deactivation of activated molecules. The addition of H₂O₂ to TiO₂ sol resulted in an increase in the degradation ratio. The H₂O₂ acted as electron acceptors to make electron/hole recombination avoided and increased the concentration of •OH radicals.

Acknowledgments

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