

Decomposition of Anthraquinone Vat Yellow 1 Using Zinc Oxide Photocatalyst

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Abstract

Decomposition of anthraquinone Vat Yellow 1 in an aqueous solution was investigated for this study. Heterogeneous photocatalysis using zinc oxide photocatalyst in aqueous solution under UV and solar irradiation was proposed to decompose anthraquinone Vat Yellow 1. Decomposition of the dye was effective under both irradiations using the zinc oxide photocatalyst. The effects of zinc oxide dose and dye concentration on the decomposition of Vat Yellow 1 under UV irradiation were investigated to assess the optimum conditions. Decomposition efficiency of Vat Yellow 1 dye increased as zinc oxide amount increased and decreased as the initial dye concentration increased. The decomposition kinetics was established to follow first-order kinetics. A study on the presence of inorganic additives such as sodium carbonate (Na₂CO₃) and sodium chloride (NaCl) was found to decrease the decomposition.

Keywords

Heterogeneous Photocatalysis, Solar Light, UV Light, Vat Yellow 1, Zinc Oxide Photocatalyst

1. Introduction

Anthraquinone dyes are largely used class of synthetic dyes after azo dyes [1] [2]. Anthraquinone dyes consist of anthraquinone with a quinoid ring as the basic chromophore. Hydroxyl groups or amino groups may also exist as chromophores in their structures. Anthraquinones are extensively used for the production of vat dyes which are most used dyes for the dyeing of cellulosic fibres such as cotton. The characteristic brightness with the good fastness of these dyes has made it extensively applicable in the textile industry [2] [3], however, anthraquinone vat dyes and their auxiliary wastes from textile industries constitute serious health and environmental hazards [4] [5] [6]. The characteristic chemical

stability of anthraquinone vat dyes and other textile dyes with their accompanying additives have proven difficult to treat in wastewater by techniques such as adsorption, coagulation, filtration, precipitation and biodegradation [7] [8] [9] [10] [11]. Promising results have however been achieved in the treatment of textile wastewaters using heterogeneous photocatalysis, an advanced oxidation process (AOP). This process involves use of insoluble semiconductor in the presence of suitable light source to generate highly reactive radicals to destroy target organic pollutant [12] [13] [14] [15]. This photocatalytic process is an environmentally friendly simple process that can be carried out under ambient condition. Zinc (ZnO) semiconductor is presently proven to show higher photocatalytic activity under UltraViolet (UV) radiation than other insoluble semiconductors due to its favourable band gap energy, high quantum efficiency and good stability. [16] [17]. In this paper, heterogeneous photocatalytic decomposition of Vat Yellow 1 was investigated using zinc oxide as photocatalyst. Vat Yellow 1 (Figure 1) is one of the frequently used anthraquinone vat dyes in the dyeing of cotton. It is often used to give a light golden yellow background colour in printing. In this present study, the decomposition efficiency of ZnO photocatalyst was assessed on the Vat Yellow 1 under both UV and solar irradiation. The effect of dye concentration and catalyst amount on the decomposition as well as the effect of the presence of inorganic ions were also studied.



Figure 1. Structure of vat yellow 1.

2. Experimental

2.1. Materials

Commercial Vat Yellow 1 (C.I.70600) with it source from Jiangsu World Chemical Co., Ltd, China was obtained from the Dyeing unit of the Department of Textile Design and Technology, Takoradi Technical University, Ghana. It was used without any purification. ZnO (99%) was purchased from Central Drug House (P) Ltd., India. It has a specific surface area of 5.0 Sq mtr/gm. Commercial Na₂CO₃ (99.5%) was obtained from Zibo Widespread International Trading Co., Ltd., China. ZnO and Na₂CO₃ were used as received. Experimental solutions were prepared using double distilled water.

2.2. Methods

Photocatalytic decomposition experiments were carried out under UV light and in direct sunlight to investigate the optimum photocatalytic activity of ZnO catalyst.

Three factors namely dye concentration, catalyst loading and inorganic additives were assessed under the two light irradiations to investigate their effect on decomposition. The sample of varying dye concentration (10 to 60 mg/L) and catalyst (1 to 6 mg/L) were magnetically stirred at 500 rpm under UV and sun irradiation at 150 min. Sodium carbonate (10 mg) and sodium chloride (10 mg) were added separately to dye solution with concentration of 40 mg and 5 mg/L catalyst loading and made to undergo the same irradiation treatments. The UV treatment was carried out in the reactor vessel (Chromato-Vue cabinet C-70G) at 254 nm whiles catalyst excitation energy source was obtained from solar light at intensity of 20,000 lux. Aliquots were drawn periodically at 30 min interval from the various treatments for decolourization and decomposition analysis. Dye decolourization was determined by the use of spectrophotometer (Model: JENWAY 7315) to measure the absorbance at λ max 425 nm. The decomposition of the dye was assessed over 230 min using COD test. Dye solution pH was measured using a Jenway 3510 pH Meter from Bibby Scientific Ltd. A pH of 6.2 was obtained for the dye solution.

3. Results and Discussion

3.1. Photocatalytic Decomposition of Vat Yellow 1 under UV and Solar Irradiation

Photocatalytic experiments were done to examine the decomposition of Vat Yellow 1 under UV and solar light irradiation using ZnO catalyst. Figure 2 shows the aliquots of dye solution taken under UV/ZnO photocatalysis at different time intervals. A gradual decolouration of vat yellow 1 from yellow to nearly colourless after 150 min of light irradiation at 30 min interval was observed. UV-Vis absorption spectra of Vat Yellow 1 as shown in Figure 3 shows gradual decreased in absorption peaks at ~425 nm which disappeared after 150 minutes of UV/ZnO photocatalysis. The decrease in absorption peaks indicated the reduction of Vat Yellow 1 concentration in the various solutions. This reduction may indicate the breakdown of the anthraquinone group in Vat Yellow1 since it is responsible for the colour of the dye [18] [19]. Similar results were obtained in studies on



60min 30min 90min 120min 150min

Figure 2. Colour fading of vat yellow 1 with time under UV/ZnO (ZnO loading = 5 mg/L; Solution pH = 6.2).



Figure 3. UV-vis spectral changes of an aqueous solution of vat yellow 1 ([Vat Yellow 1] =40 mg/L, ZnO = 5 mg/L, pH = 6.2).

removal of anthraquinone dye, polan blue E2R under UV photochemical process [20] and removal of Vat Yellow 4 using UV with TiO_2 as a catalyst [2]. The gradual reduction of colour and peak of Vat Yellow 1 therefore confirms conclusively that ZnO is effective in degrading Vat Yellow 4 under both UV and sun irradiation.

3.2. Comparison of the Extent of Dye Decomposition under Various Conditions

Three major experimental treatments were carried out on the dye solution. These treatments include: 1) agitation of the dye solution with ZnO in dark; 2) agitation of the dye solution without ZnO under UV and solar light irradiation; and 3) agitation of the dye solution under UV and solar light irradiation without ZnO. The results obtained from the studies as a shown in **Figure 4** indicate a slight colour loss of dye with the treatment in the dark in the presence of ZnO. Slight colour loss may be due to the adsorption of the dye on the surface of ZnO. UV and solar irradiation without ZnO did not cause any considerable decolouration of the dye. This indicated that vat yellow 1 was stable in the presence of UV light hence, photolysis has little influence on the decomposition of VY1 [21]. Agitation under UV and solar light in the presence of ZnO, however, caused drastic decolouration of the dye after 150 minutes.

Irradiation with ZnO caused 86% degree of decomposition under UV irradiation and 74% degree of decomposition with solar irradiation in 150 minutes. The observed variation in the rate of the degree of decomposition under UV and solar irradiation is due to the difference in irradiation energy input. ZnO absorbs only UV radiation for optimum bandgap excitation which leads to optimum photocatalysis. Maximum light absorption by ZnO, therefore, occurs with a direct UV source as indicated in **Figure 5**. Sunlight, on the other hand, has about 5% of UV



Figure 4. Effect of UV/solar irradiation on the decomposition of vat yellow 1 in the presence and absence of ZnO.



Figure 5. Percentage decomposition of vat yellow 1 under UV/Solar irradiation in the presence of ZnO.

irradiation energy needed by ZnO for optimum photocatalytic activity [22] [23] [24] [25]. Less percentage decomposition was therefore observed with solar irradiation of the dye. It can be concluded therefore, that ZnO is highly active in the UV region and hence need UV radiation for optimum catalytic decomposition.

3.3. Effect of ZnO Loading

To ensure maximum absorption of radiation photons for effective dye decomposition, the addition of the optimum amount of ZnO is vital. A varying amount of 1 to 6 mg/L ZnO loading at 40 mg/L dye concentration was investigated to examine the optimum amount of ZnO for maximum percentage removal of the dye under UV irradiation. The results as given in **Figure 6** indicate that the increase in catalyst dose from 1 to 5 mg/L steadily increased the dye removal from 50% to 89% in 150 min. At concentrations higher than 5 mg/l there was a



Figure 6. Effect of catalyst loading on the decomposition of vat yellow 1 ([Vat Yellow 1] = 40 mg/L; Solution pH = 6.2).

decrease in decomposition rate from 89% to 81%. The increase in the decomposition of dye at higher catalyst loading is due to the increase in the amount of the catalyst active sites which increases the number of dye molecules absorbed to effect decomposition. Further addition of catalyst above a certain level lead to a decrease in the decomposition rate since the dye concentration is kept constant. At constant dye concentration, the amount of dye is less sufficient to fill the surface active sites of ZnO. Results from works done on different dyes clearly indicate that the optimum catalyst loading is not fixed for all photocatalytic reactions [26] [27] [28] [29]. In the present study, 5 mg/l of catalyst loading was used as the optimum catalyst amount for the decomposition of Vat yellow 1 and for the study of other parameters. Consequently, maximum ZnO catalyst amount is needed to increase photocatalytic decomposition; however, excess ZnO catalyst can decrease decomposition.

3.4. Effect of Initial Dye Concentration

The effect of initial Vat Yellow 1 concentration on the percentage decomposition was studied by varying the initial dye concentration from 10 to 60 mg/L. The results as shown in **Figure 7** indicate that the decomposition rate decrease with an increase in dye concentration. Increasing the dye concentration from 10 to 60 mg/L results in the decrease in decomposition from 96% to 54% in 150 min. As the initial dye concentration increases, the amount of dye adsorbed per an active site of the catalyst increases causing a decrease in the efficiency of the catalyst. Sufficient amount of radiation may also be absorbed by the dye molecules at high dye concentration which reduces the efficiency of the catalyst by reducing the number of active sites on the catalyst. There is also interference of photons at high dye concentration before reaching the surface of the catalyst which reduces dye decomposition efficiency [30] [31] [32]. Conclusively, higher dye concentration ZnO catalyst loading can increase decomposition activity.



Figure 7. Effect of initial dye concentration on the decomposition of vat yellow 1 (ZnO Loading = 5 mg/l; Solution pH = 6.2).

3.5. Kinetic Study

A kinetic study was conducted in this research to assess the effect of initial dye concentration on the rate of reaction on Vat Yellow 1. The rate law employed to determine the reaction rate which as expressed in Equation (1) indicates that the photocatalytic decomposition of the dye obeys pseudo-first-order kinetics.

$$-\frac{d[C]}{dt} = k'[C] \tag{1}$$

where k' is the pseudo-first-order rate constant. The equilibrium concentration of the dye solution after adsorption-desorption equilibrium between dye and ZnO reached in 5 min was taken as the initial dye concentration for the kinetic study. Integrating Equation (1) with $C = C_0$ at t = 0 gives Equation (2)

$$\ln\left[\frac{C_0}{C}\right] = k't \tag{2}$$

where C_0 is the equilibrium concentration of the dye and C is the concentration at time t.

The plots of $\ln C_0/C$ versus *t* as shown in **Figure 8** indicate a linear fit between $\ln C_0/C$ and irradiation time which supports the conclusion that the decomposition follows first-order kinetics. **Table 1** gives the rate constants for the decomposition of the various dye concentrations. It can be observed from **Table 1** that k' decreases with increasing initial dye concentration which is in agreement with results obtained from studies on other dyes [33]. This decrease is due to an increase in initial dye concentration which results in higher competitive adsorption of dye molecules onto ZnO photocatalyst.

3.6. Mineralization of Vat Yellow 1

Complete mineralization of organic pollutant is vital in treating organically polluted wastewater. Chemical Oxygen Demand (COD) test was used to determine the mineralization efficiency of Vat Yellow 1 under UV/ZnO photocatalysis in optimized condition. **Figure 9** shows Vat Yellow 1 decomposition and COD removal at different time intervals. As shown in **Figure 9**, a progressive increase in COD removal was observed during 210 minutes of UV irradiation. This removal confirmed the mineralization of Vat Yellow 1 resulting from the breakdown of Vat Yellow 1 molecules and its intermediates. At the point where decomposition



Figure 8. Kinetics of photocatalytic degradation of C. I. Reactive Red 194 at different initial dye concentrations (ZnO loading = 5 mg/L, Solution pH = 6.2).

Table 1. The k' and \mathbb{R}^2 values at distinct initial dye concentrations.

Initial Vat Yellow 1 Concentration (mg/L)	$k'(\min^{-1})$	R²
10	0.0131	0.9734
20	0.0094	0.9907
30	0.0064	0.9859
40	0.0042	0.9873
50	0.0029	0.9893
60	0.0005	0.9761
60 40 20 20	DD Removal at Yellow-1 Removal	
0		
0 30 60 90 Time (120 150 180	210

Figure 9. Variation of vat yellow 1 and COD Percentage using UV/ZnO ([Vat Yellow 1] = 40 mg/L; ZnO Loading = 5 mg/L; Solution pH = 6.2).



Figure 10. Effect of the presence of Na_2CO_3 and NaCl on the decomposition of vat yellow 1 ([Vat Yellow 1] = 40 mg/L, Amount of ZnO = 5 mg/l).

achieved 91% at 150 min of UV irradiation, COD removal reached 80% indicating still the presence of intermediates in Vat Yellow 1 solution. COD removal however reached up to 90% when the irradiation time was extended to 210 minutes which indicated that mineralization required a longer time [34] [35].

3.7. Effect of Na₂CO₃ and NaCl

The effect of addition Na₂CO₃ and NaCl on the photocatalytic decomposition of Vat Yellow 1 is shown in **Figure 10**. Increasing carbonate (CO_3^{2-}) and chloride ion (Cl⁻) concentration gradually decrease the decomposition percentage of the dye as observed in **Figure 10**. This decrease is due to the hydroxyl scavenging property of carbonate ions to scavenge hydroxyl radicals which is the primary source for the photocatalytic decomposition [2] [10] [29]. Consequently, there is an ultimate decrease in the percentage decomposition of the dye significantly. Furthermore, the isoelectric point (IEP) of ZnO is \geq 8.7 [36] [37] [38] [39]. Hence at pH 6.2 of dye solution, ZnO is positively charged since dye solution pH is below its isoelectric point. This results in migration of Cl⁻ ions to the positively charged ZnO surface to hunt for h⁺ and *OH*• causing reduction effect in dye decomposition.

4. Conclusion

Anthraquinone Vat Yellow 1 decomposition is resistant to direct photolysis. ZnO photocatalyst was revealed to be effective in the decomposition of vat yellow 1 under UV as well as solar irradiation. Decomposition of the dye with ZnO catalyst under UV irradiation was however higher compared to that of the solar irradiation. The optimum decomposition efficiency was obtained with a catalyst loading of 5 mg/L. Decomposition rate decreased with an increase in the initial concentration of the dye. Decomposition was also found to decrease in the presence of NaCl and Na_2CO_3 .

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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