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Photocatalytic Degradation of Monoazo Dye in Ethanol using Zinc Oxide in Ultra-Violet Radiation

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The photocatalytic degradation of azo dye 2-(2-naphthyl azo)-4,5-diphenyl imidazole was studied. Solutions of different concentrations of this dye using zinc oxide under UV irradiation. Different parameters were measured such as the catalyst amount, dye concentrations, light intensity and effect of pH. The optimum concentration of catalyst was 0.15 g/100 mL while the optimum concentration of dye was 50 ppm. It has been shown that the best pH to degrade monoazo dye 8.1 with 8.44 mW/cm^2 light intensity. The photocatalytic degradation of azo dye follows the first order reaction. In addition, photocatalytic degradation of the dye has been studied using different concentrations of aluminium oxide suspended in an aqueous solution of this dye irradiated by ultraviolet lamp in a vessel reactor at room temperature, with 10 cm^3/min air bubble has been passed through the solution. In addition, when the dye concentration increased, the photocatalytic degradation efficiency of dye decreased to 83.54 %.

Keywords: Photcatalytic degradation, Azo dye, Zinc oxide, Degradation efficiency.

INTRODUCTION

Water pollution is one of the serious problems where efforts have been devoted to sort out. This pollution results from different pollutants and one of them is azo dyes [1,2]. These dyes have been enormously used in textile, painting and cosmetics industries [3,4]. Due to their bright colour, water solubility and toxicity [5-7], many methods have been used to purify water from them. Such methods include adsorption, chemical and biological removal and photochemical processes [8-10]. Among these processes, photocatalytic process can be considered as the effective method where many metal oxides are used to degrade these dyes in the presence of light. One of the most important oxides is ZnO due to its low-cost and its absorption over a wide range of spectrum than the other metal oxides [11,12].

These metal oxides could be discharged without treatment of water. The photochemical process is applied for the treatment of water and wastewaters relies on advanced oxide processes [13-15]. Whereas, this method produces the reactive hydroxyl radicals (•OH) and these radicals are able to mineralizing organic pollutants [16-18]. Furthermore, when these oxides are exposed to photons the electrons are promoted from the valence band to the conduction band leaving the electrons in the conduction band where they undergo photo reduction and this leads to leave a positive hole in valence band [19,20].

In this paper, the kinetic of colour removal of an azo dye was shown using the heterogeneous photocatalytic degradation.

EXPERIMENTAL

An azo dye 2-(2-naphthyl azo)-4,5-diphenyl imidazole was prepared according to the literature [21]. Zinc oxide was obtained from Merck while other solvents NaOH, ethanol and HCl were purchased from BDH.

Preparation of dye solutions: A stock solution of prepared azo dye (10^{-3} M) was prepared in 50 mL absolute ethanol. The desired dye solutions with different concentrations were prepared and their optical densities were determined at λ_{max} (469 nm). Different dosages of catalyst ZnO were added to these solutions and then irradiated with UV- lamp.

Photoreactor and procedure: The photocatalytic degradation of azo dye was investigated using photoreactor. This reactor consists of two thimbles, one of them is outside and the second is inside. Water was passed through the outside thimble to cool the reaction mixture until its temperature reached at room temperature. The reaction solution (100 mL)

was put in the reaction chamber. The photocatalytic degradation of dye was conducted under 125W low-mercury lamp. All experiments of the photocatalytic degradation processes of dye have been performed by mixing 0.15 g/100 mL of the catalyst with 50 ppm of dye solution. To maintain an adsorption equilibrium between the surface of catalyst and dye, the suspension solution was stirred in the dark for 30 min and air (10 mL/min) was passed through the dye solution during irradiation. 2 mL of suspension reaction mixture was withdrawn every 10 min, and finally centrifuged at 4000 rpm to remove any residual of ZnO particles. The absorptions of dye samples were then measured using UV-visible spectrophotometer.

RESULTS AND DISCUSSION

Effect of zinc oxide loading mass on photocatalytic degradation of azo dye: Number of experiments have been performed using different amount of zinc oxide (0.03-0.60 g/100 mL), in order to obtain the best conditions for dye degradation. All experiments were carried out under optimum conditions including 50 ppm concentration of this dye at pH = 8.1 and 10 mL/min the flow rate of passed air at room temperature. The degradation of the monoazo dye with decreasing dosage of zinc oxide is shown in Fig. 1. The best photodegradation was obtained using 0.15 g/100 mL of catalyst, which combined with high light absorption. In addition, by increasing the dosage of zinc oxide, the photodegradation effecincy increased as its active sites increased which results in enhancing the number of adsorbed dye molecules. However, beyond the maximum catalyst amount (0.15 g/100 mL), the percentage degradation of dye was decreased. This decrease is due to the light absorption by dye only on the first layer of ZnO. Moreover, when the turbidity of solution increased due to the high dosages of zinc oxide the light penetration through the dye solution decreased and this led to light scattering. In this case, some zinc oxide surface area decreased and became unavailable to absorb photons and eventually to adsorb the dye and this led to reduce the photocatalytic reaction [22,23].

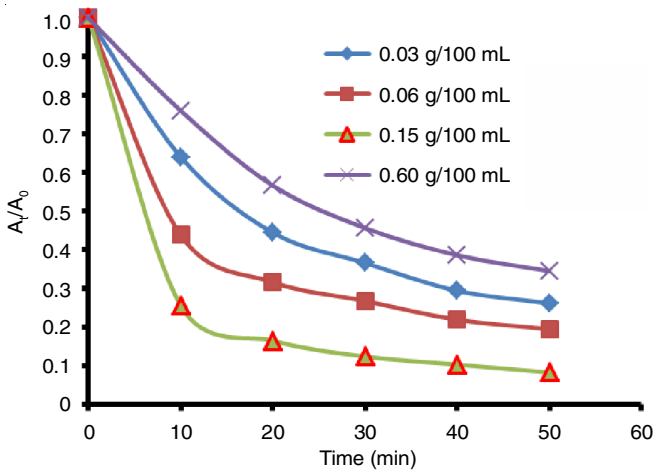

Fig. 1. Effect of ZnO loading on the monoazo dye degradation using ultraviolet radiation, 50 ppm azo dye and pH = 8.1

Effect of azo dye concentration on photocatalytic degradation: Different concentrations of azo dye (50-100 ppm) were chosen to show their effect on the photocatalytic degradation process. Using 0.15g/100 mL, as optimum value of ZnO as catalyst at pH= 8.1 and was irradiated with 8.44 mW/cm^2 UV light with a flow rate of air 10 mL/min at room temperature. (Figs. 2 and 3). When the dye concentration increased, its photocatalytic degradation decreased due to covered large area of ZnO and this leads to the absorption of maximum amount of photons to give the high concentration of activated ZnO. Moreover, at higher concentrations of dye at > 50 ppm, the active sites of ZnO are saturated by the dye and this leads to reduction in electron-hole pair generation and that leads to reduce the photodegradation efficiency. Furthermore, this reduction in photodegradation efficiency due to the excess amount of dye on ZnO surface which decrease the light penetration. Photocatalytic degradation of different organic pollutants was studied by other researchers and similar results were observed in which ZnO is the best photocatalyst in comparison with TiO_2. For instance, the percentage of reactive Red 15 dye removal was 87 % and 90.5 % using TiO_2 and ZnO, respectively [24-26].

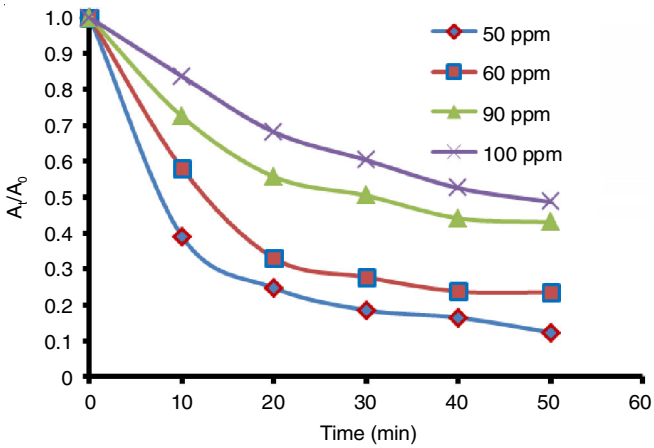

Fig. 2. Change in (A/A_0) with irradiation time at different concentrations of monoazo dye

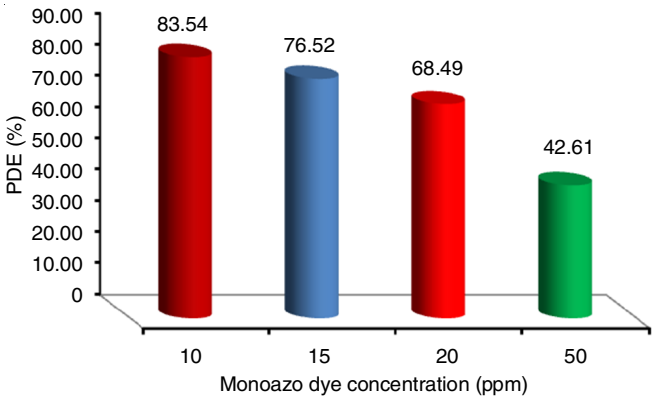

Fig. 3. Relation between photocatalytic degradation efficiency (PDE %) with the dye concentration using 0.15 g/100 mL ZnO

Effect of pH: The photocatalyic degradation efficiency of dye was affected by pH changes, where various pH values 4 -11 were taken and these values were adjusted using 0.01 N HCl and 0.01 N NaOH as shown in Fig. 4. The point of zero charge of ZnO 8.1. The zinc oxide surface has a positive charge in acidic solutions (pH < 8.1) and it is with negative charge in alkaline media (pH > 8.1). Therefore, in the acidic medium where the dye adopted the cationic form there was a poor adsor-

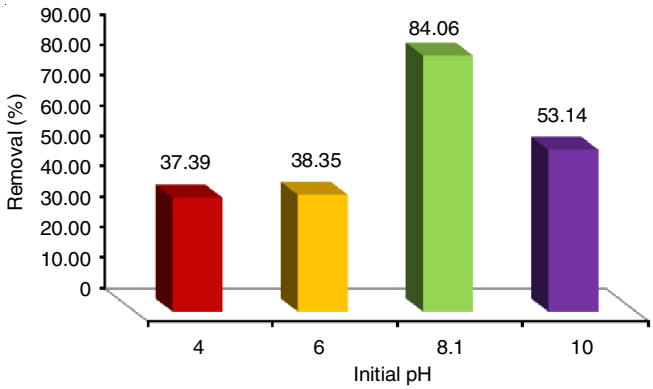

Fig. 4. Monoazo dye degradation at different initial pH values using ultraviolet light, initial azo dye concentration (50 ppm) and (0.15 g/100 mL) of ZnO

ption, consequently causes decreased in the photocatalytic degradation efficiency (PDE %) of dye. At higher pH value in the alkaline medium, more dye molecules would adsorb on the catalyst surface resulted in high PDE % of dye due to electrostatic attraction of the negatively charged zinc oxide with anionic dye.

The photocatalytic degradation of dye was found to be maximum at pH 8.1 with photocatalytic efficiency (84.06 %) because of generation of hydroxyl radicals from the oxidation of hydroxyl ions. Moreover at pH 8.1, the dye absorption increased on the surface of zinc oxide. While in strong alkaline medium, the degradation of dye gradually decreased because of the decrease in dye absorption on the negatively charged ZnO. In addition in alkaline medium, the hydroxyl radicals could be scavenged very fast [27].

Conclusion

The first order photocatalytic degradation process of monoazo dye relied on the amount of catalyst which was 0.15 g/100 mL. In addition, when dye concentration increased the photocatalytic degradation processes decreased and this is due to the decrease of concentration of OH^- which is adsorbed on zinc oxide surface with 50 ppm concentration of dye as an optimum value. The best pH value of photocatalytic degra-dation was 8.1 and the photocatalytic degradation of dye incre-ased when the light intensity increased due to the increase of photoelectron in the conduction band. This is arttibuted due to the increase of electron-hole pairs and a decrease of recombination process between photoelectron and hole in the valance band. The best value of light intensity was 8.44 mW/cm^2 and the efficiency degradation of monoazo dye was found to be 84.06 %.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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