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Heterogeneous photocatalytic degradation of rose bengal: Effect of operational parameters

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ABSTRACT

The photocatalytic degradation of rose bengal dye has been investigated using ZnO nanoparticles as photocatalyst. ZnO nanoparticles were found to be efficient catalyst for the degradation of dye and 98% degradation was observed in 90 min. Effect of various operational parameters such as amount of catalyst (0.25–2.00 g/L), concentration of dye (0.01–0.05 mM) and pH (3–11) of dye solution on the rate of dye degradation was studied. The most favorable results for the degradation of rose bengal were observed at pH 5 at a catalyst loading of 1 g/L. Moreover, hydroxyl radicals have been detected in the photocatalytic reaction mixture by using terephthalic acid photoluminescence probing technique. The reusability of the catalyst has also been studied and catalyst was found to be active even after being used for 5 times.

1. Introduction

In recent years, due to industrialization, there is rapid increase in release of pollutants from industries into water bodies. Industries mainly responsible for polluting water bodies are food, textile, dyeing, chemical, printing, etc. Out of these industries, principal source of water pollution is dyeing industry. Discharge from these industries has deleterious effect on aquatic flora and fauna and render water toxic and unfit for use. Also, due to their persistent nature, they remain in environment for very long period of time.

Out of various classes of dyes, xanthene dyes are most widely used. Xanthene dyes are class of dyes, characterized by presence of xanthene nucleus with aromatic groups as chromophore. These dyes are extensively used as colorant in textile, printing and dyeing industries. But these dyes are reported to be genotoxic, mutagenic, cytotoxic and cytostatic [1–5]. Rose bengal is an important xanthene dye and is widely used in textile, dyeing and photochemical industries. Sako and co-workers [6,7] have studied the toxic effect of rose bengal on *Paramecium caudatum* and cultured fetal rat hepatocytes. The authors observed that rose bengal is toxic and also inhibits leucine aminopeptidase.

Thus there is significant need to expunge rose bengal dye from textile wastewater. In this context, semiconductor heterogeneous photocatalysis has emerged as a promising technique for the obliteration of dissolved dyes [8–10]. In this method, a semiconductor catalyst is irradiated with a suitable light source, which results in the in situ generation of hydroxyl radicals. The hydroxyl radicals due to their high redox potential (2.8 V) are able to completely break down recalcitrant dye molecules. This is a very facile, economical and environmentally benign technique for the degradation of dyes. Khataee et al. [8] studied the degradation of C.I. basic red 46 by combining photoelectron-Fenton and heterogeneous photocatalytic process. The authors observed 99.2% mineralization of 15 mg/L dye in 6 h. Similarly, Daneshvar et al. [9] systematically investigated the degradation of acid red 14 using ZnO photocatalyst. The authors also studied the effect of parameters such as pH, amount of photocatalyst, hydrogen peroxide, and ethanol concentration on the rate of dye degradation.

Although considerable advances have already been made for the degradation of dyes, degradation of rose bengal is less explored. Moreover, earlier reports involve longer time period for the degradation of rose bengal. Bhar et al. [11] have studied the degradation of rose bengal using nanocrystalline FeS_2 thin films. The authors observed 84% degradation of dye solution in 300 min. Degradation of rose bengal has also been studied by Farbod and Khademalrasool [12] using TiO_2 nanoparticles. Complete degradation of dye was observed in 210 min. Similarly, Vignesh et al. [13] observed 85% degradation of rose bengal in 150 min with Ag doped SnO_2 nanoparticles modified with curcumin.

The present paper describes our findings for the degradation of rose bengal in the presence of ZnO photocatalyst under UV irradiation. The time interval required for the degradation of rose

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bengal is very short as compared to previously reported methods. Moreover, effect of various operational parameters such as amount of catalyst (0.25–2.00 g/L), concentration of dye (0.01–0.05 mM) and pH (3–11) of dye solution on the rate of dye degradation was studied.

2. Experimental

2.1. ZnO photocatalyst

The photocatalytic degradation of rose bengal dye has been investigated using ZnO catalyst synthesized by our previously reported method [14]. The catalyst was synthesized using thermal decomposition of oxalate precursor method and was characterized using a Powder X-ray diffraction technique and scanning electron microscopy. The average crystallite size of ZnO nanoparticles was ~27 nm.

2.2. Photocatalytic activity testing

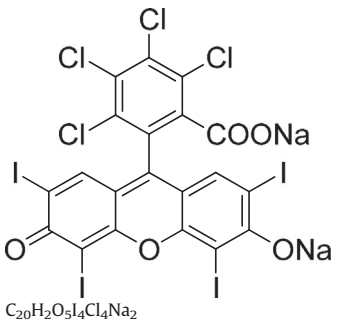
The photocatalytic experiment was performed by evaluating the photodegradation of 100 ml rose bengal aqueous solution (0.03 mM) at a catalyst loading of 1.00 g/L. Molecular structure and characteristics of rose bengal are shown in Table 1. The experiments were performed under UV irradiation. The progress of the reaction was scrutinized by recording the UV–vis absorption spectra of reaction mixture at specific time intervals using a JASCO V-530 spectrophotometer and monitoring the variation of absorption at 547 nm.

Effect of various operational parameters such as amount of catalyst (0.25–2.00 g/L), concentration of dye (0.01–0.05 mM) and pH (3–11) of dye solution on the rate of dye degradation was studied. For studying the effect of pH on the rate of dye degradation, the pH of solution was varied by the addition of 0.1 M HCl or NaOH.

2.3. Detection of hydroxyl radicals

To determine whether reactive oxygen species involved in the photocatalytic degradation of rose bengal is hydroxyl radical or not, terephthalic acid photoluminescence probing technique was used. In this, alkaline solution of terephthalic acid, having ZnO nanoparticles was irradiated with UV light. After 10 min of irradiation, sample was withdrawn from the reaction mixture and was centrifuged to separate catalyst particles. The photoluminescence spectrum of sample was recorded between 325 and 600 nm at an excitation wavelength of 315 nm and variation in intensity of peak at 425 nm was monitored using Perkin Elmer LS 55 Fluorescence Spectrometer.

Table 1
Molecular structure and characteristics of rose bengal.

Molecular structure	
Molecular formula	$C_{20}H_2O_5I_4Cl_4Na_2$
Molecular weight	1017.68 g/mol
C.I. number	45440
λ_{max} (nm)	547

3. Results and discussion

The photocatalytic degradation of rose bengal has been investigated using ZnO catalyst. Fig. 1 shows the time dependent UV–vis spectra of rose bengal in the presence of ZnO catalyst under UV irradiation. It is evident from the figure that with increase in irradiation time there is decrease in absorption maximum and almost complete degradation is observed in 90 min.

Two blank experiments were also performed: one with only UV light and one with only ZnO catalyst. The combined results of degradation of dye for: (i) ZnO+UV, (ii) only ZnO catalyst, and (iii) only UV light, are shown in Fig. 2. It was found that in the absence of either UV light or ZnO, very small degradation was observed. While for UV+ZnO, 98% dye was degraded in 90 min. So it was inferred that presence of both UV light and catalyst is required for efficient dye degradation. The rate constant values for the degradation of dye were calculated by employing first order rate equation [15]

$$\ln \frac{C_0}{C_t} = kt \quad (1)$$

where C_0 is initial concentration of dye, C_t is concentration of dye at time t and k is the first order rate constant. Inset of Fig. 2 shows the linear relationship between $\ln C_0/C_t$ and irradiation time and from the slope of the graph, rate constant value was found to be 0.03 min^{-1}.

3.1. Effect of amount of catalyst

The amount of catalyst used for dye degradation has a profound impact on the rate of degradation. Xia et al. [16] varied the amount of Fe_3O_4/ZnO catalyst from 0.17 to 0.68 g/L. The authors observed an initial increase in rate of methyl orange degradation up to catalyst loading of 0.51 g/L followed by a decrease in rate of degradation with further increase in amount of catalyst. Similar variation in rate of degradation as a function of amount of catalyst was observed by Rao et al. [17].

Thus in order to determine the optimal amount of ZnO catalyst for the degradation of rose bengal, the amount of catalyst was varied from 0.25–2.00 g/L at a constant dye concentration of 0.03 mM. The percentage of dye degraded after 60 min irradiation

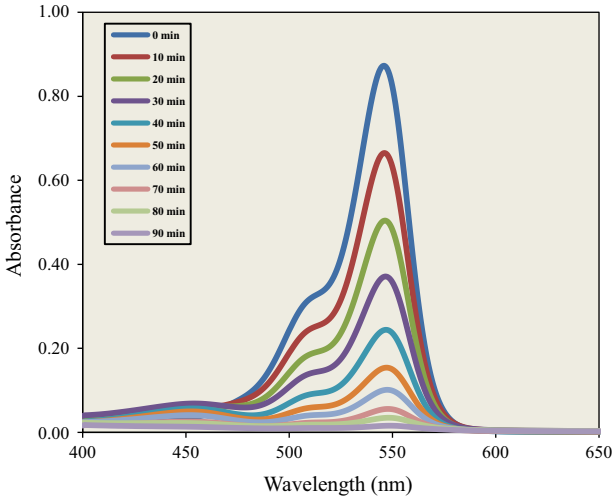


Fig. 1. Time dependent UV–vis spectra of rose bengal.