

Solar photocatalytic mineralization of isoproturon over TiO₂/HY composite systems[☆]

Mangalampalli V. Phanikrishna Sharma, Kannekanti Lalitha,
Valluri Durgakumari, Machiraju Subrahmanyam*

Inorganic and Physical Chemistry Division, Indian Institute of Chemical Technology, Hyderabad 500007, India

Received 18 May 2007; received in revised form 7 September 2007; accepted 23 September 2007

Available online 5 November 2007

Abstract

The present investigation covers immobilization of titanium dioxide over HY support for the treatment of isoproturon pesticide. Catalysts are characterized by XRD, SEM–EDAX, TEM, BET surface area and UV–vis DRS. A detailed photocatalytic degradation study under solar light in aqueous suspensions with parameters like loading of TiO₂ over HY, amount of the catalyst, concentration of substrate, pH effect, durability of the catalyst and comparison between suspended TiO₂ and supported systems are reported. Mineralization of isoproturon is monitored by total organic carbon, chemical oxygen demand and a plausible mechanism is proposed for photocatalytic degradation based on degradation products.

© 2007 Elsevier B.V. All rights reserved.

Keywords: Titanium dioxide; HY zeolite; Photocatalytic degradation; Isoproturon

1. Introduction

Herbicides are mainly used in agricultural applications. Phenylurea herbicides are broadly applied worldwide on agricultural soils for weed control and isoproturon (Fig. 1) is one among the widely used. It is a systemic herbicide [3-(4-isopropylphenyl)-1,1-dimethylurea] for the control of annual grasses and broad-leaved weeds in agricultural fields. Their application in modern agricultural practices is a cause for concern due to their relatively high solubility in water and low chemical and biological degradation rates. The contamination of ground and surface water [1,2] is because of mal-agricultural practices, careless disposal of empty containers and washing of equipment. As a consequence, different strategies have been devised for the removal of these pollutants from water. Traditionally, pollutants in water are removed using activated carbon [3],

nanofiltration [4], ozonation [5] and isolation of specific bacterial cultures [6]. These processes have inherent limitations in applicability, effectiveness and costs. In recent years, advanced oxidation processes (AOPs) have been proposed [7] as an attractive alternative for the treatment of contaminated ground, surface, and wastewater containing pesticides or non-biodegradable organic pollutants [8–10]. These methods are generally based on the generation of OH radicals which interact with organic pollutants leading to progressive degradation and subsequently complete mineralization. Among the AOPs, titanium dioxide mediated semiconductor photocatalysis is gaining more importance due to its high production of hydroxyl radicals, inexpensive, non-toxic, abundantly available and especially stable under irradiation. In fact, the main drawback of this approach is the need for complex filtration procedure and also decrease of radiation flux due to the turbidity of the catalyst suspension, called as shadowing effect. These problems have motivated the development of immobilization of TiO₂ on different support materials like glass [11], pumice stone [12], Cuddapah stone [13], activated carbon [14] and zeolites

[☆] IICT Communication no.: 070301.

*Corresponding author. Tel.: +91 40 27193165; fax: +91 40 27160921.

E-mail address: subrahmanyam@iict.res.in (M. Subrahmanyam).

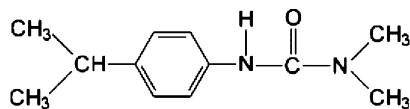


Fig. 1. Structure of isoproturon.

[15], etc. In this context, zeolites have attracted greater attention due to their adsorption capacity that helps in pooling the pollutants to the vicinity of the TiO_2 surface and leads to faster degradation [9,15,16]. Zeolites are microporous crystalline aluminosilicates with structural features that make them attractive hosts for photochemical applications, i.e. ability of photoinduced electron donor and acceptor reactions and strongly influence with extra framework cations in the photochemical reactions [17]. Furthermore, zeolites delocalize bandgap excited electrons of TiO_2 and thereby minimize electron–hole recombination. These interesting properties attract zeolites as catalyst support in the treatment of pesticide contaminated water. TiO_2 supported on zeolites, with their large surface area and light transparent nature increases the adsorption capacity and uniform diffusion of pesticide pollutants leads to efficient degradation. The objective of present investigation is the development of an efficient HY supported TiO_2 photocatalyst for complete mineralization of isoproturon in aqueous solution. Different parameters are optimized for photocatalytic degradation of isoproturon. Mineralization of isoproturon is monitored by total organic carbon (TOC) and chemical oxygen demand (COD) and an attempt is made to identify intermediates and a plausible mechanism is proposed for the isoproturon photocatalytic degradation.

2. Experimental

2.1. Materials and methods

All chemicals used in the present work were of analytical grade and used as such without further purification. Isoproturon (>99% pure, Technical grade) obtained from Rhône-Poulenc Agrochimie, France and titanium dioxide P-25 (Anatase 80%, rutile 20%, surface area $50\text{ m}^2\text{ g}^{-1}$ and particle size 27 nm) is from Degussa Corporation, Germany. Zeolite NH_4Y (Si/Al = 2.7), HCl, H_2SO_4 , NaOH and acetonitrile are obtained from Ranbaxy Limited, India. AgSO_4 , $\text{K}_2\text{Cr}_2\text{O}_7$, and ferrous ammonium sulfate are obtained from S.D. Fine Chemicals Limited, India. All the solutions were prepared with deionized water obtained using a Millipore device (Milli-Q).

2.2. Preparation of HY supported TiO_2 catalysts

Ammonium form of Y-zeolite is converted into H-form by calcination at $500\text{ }^\circ\text{C}$ in air for 7 h. TiO_2 supported on HY photocatalysts are prepared by solid state dispersion (SSD) method as reported earlier [15]. TiO_2 loading over

zeolite was maintained to obtain 3, 5, 10, and 15 wt% in the finished catalyst.

2.3. Characterization

The catalysts are characterized by various techniques like XRD, UV–vis DRS, BET surface area, SEM–EDAX and TEM. The XRD of catalysts are analyzed by Siemens D-5000 X-ray diffractometer using Ni filtered $\text{Cu K}\alpha$ radiation ($\lambda = 1.5406$) from $2\theta = 2$ to 60° . The UV–vis diffused reflectance spectra are recorded on GBC UV–vis Cintra 10_e spectrometer. The surface area of the catalysts was determined by the physical adsorption of a monolayer of nitrogen gas (cross-sectional area = 0.162 nm^2) at $-195\text{ }^\circ\text{C}$ from BET adsorption measurements using an Auto Chem 2910 (Micromeritics, USA) instrument. The SEM analysis samples were mounted on an aluminum support using a double adhesive tape coated with gold in HUS–SGB vacuum coating unit and observed in Hitachi S-520 SEM unit. Elemental analysis was carried out using Link, ISIS-300, Oxford EDAX detector. TEM sample is carried out on TECHNAI F12 Philips unit operated at 80 kV with a filament current of 27 mA.

2.4. Photocatalytic experiments

$1.14 \times 10^{-4}\text{ M}$ isoproturon solution is freshly prepared by taking required amount of isoproturon in 500 ml of deionized water and vigorously stirred until it dissolves. This concentration is used in all the experiments until unless stated. Prior to the start of light experiments, dark (adsorption) experiments are carried out by taking 50 mg of catalyst in 50 ml of isoproturon solution under continuous stirring in dark for better adsorption of the pesticide on to the catalyst surface and change in isoproturon concentration was measured by HPLC. For all solar experiments, isoproturon solution of 50 ml is taken in an open glass reactor with known amount of catalyst. The initial pH of the solution is maintained by adding HCl or NaOH. The slurry solution is continuously stirred in a shaking unit at 120 rpm and illuminated under bright solar light for 4 h. Distilled water is added periodically to avoid concentration changes due to evaporation. The solar experiments (intensity $\sim 75\text{ mW cm}^{-2}$) are carried out at 11:00 a.m. to 3:00 p.m. during November and December 2006 in Hyderabad.

2.5. Analyses

The isoproturon degradation is monitored by Shimadzu 10AvP HPLC using C-18 phenomenex reverse phase column with acetonitrile/water mobile phase of ratio 50/50 (v/v) at a flow rate of 1 ml min^{-1} . The degradation by-products are identified by Agilent 1200 series LC–MS. The mineralization of isoproturon is analyzed through depletion of TOC using Shimadzu TOC–VCPN analyzer and COD is carried out using dichromate solution as the

Download English Version:

<https://daneshyari.com/en/article/80880>

Download Persian Version:

<https://daneshyari.com/article/80880>

[Daneshyari.com](https://daneshyari.com)