

Chlorpyrifos and Endosulfan degradation studies in an annular slurry photo reactor



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ARTICLE INFO

Article history:

Received 26 November 2014

Received in revised form

30 July 2015

Accepted 15 August 2015

Available online 11 November 2015

Keywords:

Photo catalysis

Toxicity

TiO₂

Slurry

Endosulphan and chlorpyrifos

ABSTRACT

TiO₂ is one of those compounds which are highly used in photocatalytic degradation of substrates using UV radiation. The substrates are degraded oxidatively and hence finds an important position in advanced oxidation for water/wastewater treatment processes. The thrust of this research was to evaluate the effectiveness of Heterogeneous Photocatalysis (HP) technique, for the removal of pesticides from water/wastewater. The photo-catalytic degradation of two pesticides, widely used in India, viz., Endosulfan (ES) and Chlorpyrifos (CPS) was studied in an annular slurry photo reactor under UV illumination at 254 nm. Results revealed that the degradation rate is significantly affected by the initial pesticide concentration, pH of the solution and catalyst concentration. Batch degradation studies on Endosulphan and Chlorpyrifos were conducted in the concentration range from 5 to 25 mg/L at a pH ranging from 3.5 to 10.5 and at a catalyst loading of 0.5–2 g/L. Endosulphan removal efficiency was about 80–99% and chlorpyrifos removal efficiency was about 84–94%. L–H rate constants were determined using L–H kinetics. High removal efficiencies obtained (80–99%) indicate the effectiveness of this process and its potential for practical application.

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1. Introduction

Several pesticides like Chlorpyrifos (O,O-diethyl O-3,5,6-trichloro pyridin-2-yl phosphorothioate) and Endosulfan (6,7,8,9,10,10-Hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepine-3-oxide) are commonly used in India for increasing agricultural productivity (Sankararamkrishnan et al., 2005). Chlorpyrifos is a broad-spectrum organophosphorous pesticide with a principal mechanism of toxicity by inactivation of acetylcholinesterase at nerve junctions. Species sensitivity varies considerably across kingdom. Aquatic invertebrates, particularly crustaceans and insect larvae, are sensitive to exposure: LC50s are generally less than 1 µg/L (Racke, 1993). Endosulfan is an organochlorine pesticide which acts as central nervous system poison (Jones et al., 2009). Acute intoxication may result in neurological manifestations, restlessness, muscular twitching and convulsions that may end in death. It gives highly toxic effects to amphibians and human (Ribeiro et al., 2001). Because of their widespread use, they are detected in various environmental matrices such as soil, water and air. Organochlorine pesticides are known for its toxicity and persistence to biodegradation. They are ubiquitous and found

even in the most remote areas, far from any industrial activity. Direct applications of insecticides rain water runoff from agricultural systems, disposal of out dated stocks and discharge of wastewater from industries are the major sources of insecticides contamination in water bodies in India. Many treatment technologies are available for the remediation of pesticide contaminated water. They can be classified as Physical, chemical and biological methods. Adsorption, incineration and advanced oxidation are the most commonly used conventional treatment methods for the treatment of pesticide contaminated water and wastewater. Physical unit treatment processes such as nanofiltration and reverse osmosis (Boussahel et al., 2002), slow sand filtration, activated carbon adsorption as well as chemical oxidation technologies such as ozonation and advanced oxidation processes have led to partial removal of pesticides residues during water treatment. Imitations of conventional decontamination techniques have resulted in an intensive search for more efficient treatment techniques. Advanced Oxidation Process (AOP), which is based on the principle of highly reactive hydroxyl radical generation, has emerged as a sound alternate technique for treatment of hazardous pollutants (Hoffmann et al., 1995, Devipriya and Yesodharan, 2005, Wang et al., 2006). The highly reactive hydroxyl radicals oxidize a broad range of pollutants non-selectively due to their high oxidative capacity (E₀ = 2.8 V) (Madhavan et al., 2010). Therefore, the thrust of this research was to explain the Advanced Oxidation processes

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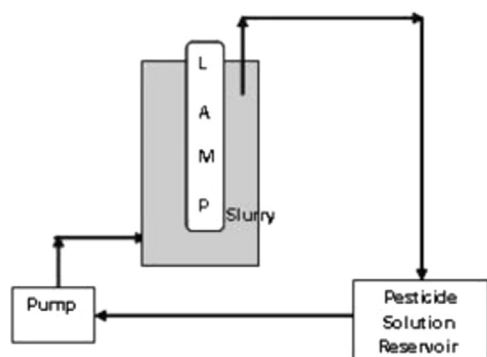


Fig. 1. Schematic diagram of annular reactor.

(AOP) technique, which is suitable for removal of pesticides from water/wastewater in environment. The objective of the present study is to evaluate the effectiveness of photo catalysis in degradation of CPS and ES using three major parameters, initial pesticide concentration, catalyst concentration and pH were studied in batch recirculation mode with the annular slurry reactor.

2. Materials and methods

2.1. Materials

Technical grade pesticide Endosulfan (99.8% purity) (E.I.D. Parry pesticides division, India), Chlorpyrifos (99.5% purity) (Sriram Pesticides, India) and TiO_2 (pure anatase, surface area $15 \text{ m}^2/\text{g}$, CDH, India) were used in the experiments.

2.2. Experimental studies

A schematic of the experimental setup is shown in Fig. 1. Pesticide degradation studies were carried out in an annular photo reactor consisting of an inner quartz tube of 2.5 cm dia, outer cylinder of 3.82 cm dia and length of 30 cm. A UV lamp (Philips, 9W) emitting predominantly at 254 nm wavelength provided the photonic energy required for degradation. Slurry of the solution pesticide mixed with TiO_2 photocatalyst, was continuously circulated from a well-mixed feed tank, using a peristaltic pump (Ravel Hitech, India) through the annular chamber. Samples were collected from the reactor at specific time intervals and analyzed by HPLC after filtration (Gelman GHP acrodisc $0.25 \mu\text{m}$) to remove titania particles. The degradation studies were carried out over a range of pH from (3.5–10.5), initial concentration of pollutant (1–10 mg/L) and different catalyst concentrations (0.5–2 g/L).

2.3. Analytical method

Concentration of residual pesticides in the samples were analyzed with a Gradient HPLC (Jasco Pu-2089 plus, Japan) with PDA detector using Agilent Eclipse PAH $5 \mu\text{m}$ column of dimension $10 \times 150 \text{ mm}^2$. Acetonitrile and milliQ water in the ratio of 70:30 was used as mobile phase at a flow rate of 1 ml/min.

3. Results and discussion

The effects of three major parameters, initial pesticide concentration, catalyst concentration and pH were studied in batch recirculation mode with the annular slurry reactor.

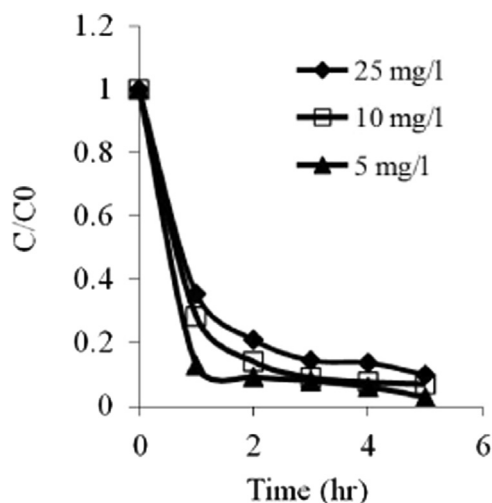


Fig. 2. Effect of initial concentration on CPS degradation TiO_2 – 0.5 g/L, pH-7.

3.1. Effect of initial pesticide concentration

The effect of initial pesticide concentration on photocatalytic degradation was studied in the range of 5–25 mg/l for both CPS and ES. These concentrations are higher than the pesticide concentration in normal water supplies which can be observed in pesticide manufacturing wastewaters and in heavily contaminated ground waters. The results, shown in Figs. 2 and 3 substantiate the photo-catalytic degradation of both the pesticides. The removals range from 80% to 95% in about 4–5 hours. Both the pesticides were degraded rapidly in the first two hours, but the rate of degradation decreased later. This is generally attributed to the formation of intermediates and their competition with the pesticide for the OH-radicals. It was also observed that the rates and extent of degradation decreased with increasing pesticide concentration. This may be explained on the basis of competition for active TiO_2 sites between pesticide adsorption and photonic OH-generations. The reduction in pollutant removal efficiency with increasing pollutant concentration in photo catalysis is widely reported in the literature. (Ochuma et al., 2007; Chong et al., 2009b, 2009c). There are many possible reasons for this behavior. Since adsorption precedes photo catalysis, at higher pesticide concentrations, most of the photo-catalyst surface is adsorbed by the pesticide leaving lesser number of sites for hydroxyl radical generation. High concentrations of organic substrates saturate the TiO_2 surface and reduce the photonic efficiency leading to photo-catalyst deactivation.

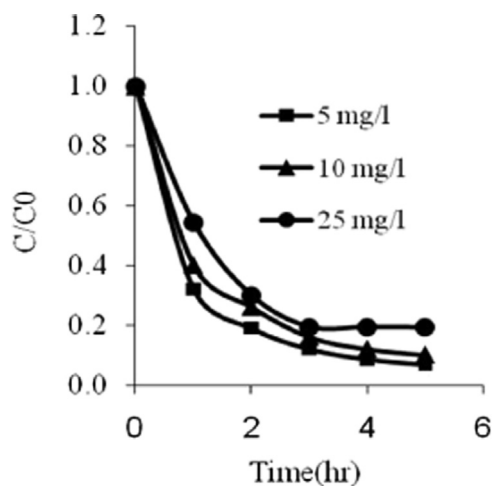


Fig. 3. Effect of initial concentration on ES degradation TiO_2 load – 0.5 g/L, pH-7.

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