

Degradation of 1,4-dioxane in water using TiO₂ based photocatalytic and H₂O₂/UV processes

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Abstract

1,4-dioxane is a synthetic compound found in industrial effluent and subsequently contaminates water bodies due to its high solubility and high volatility. It is of concern due to its toxic and hazardous nature and has been listed as a class 2B carcinogen. This study involved optimisation of the photocatalytic and H₂O₂/UVC processes for 1,4-dioxane removal. Different photocatalysts and loadings were investigated for the degradation of low concentrations of 1,4-dioxane in water including a commercial P25, a synthesised magnetic photocatalyst and an immobilised sol–gel system. A commercial catalyst (Degussa P25) was the most efficient. A lifetime study of the sol–gel reactor showed that the coating was stable over the time period studied. The optimum H₂O₂ concentration in the H₂O₂/UVC process was found to be 30 ppm. The addition of H₂O₂ to the photocatalytic process for 1,4-dioxane removal caused a decrease in rate for the commercial P25 photocatalyst and an increase in rate for the lab-made magnetic photocatalyst.

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1. Introduction

1,4-dioxane (1,4-diethylene dioxide) is a ring-structured chemical, C₄H₈O₂, used as an industrial solvent stabiliser that prevents the breakdown of chlorinated solvents during manufacturing processes. It has a propensity to enter the environment and contaminate water supplies because of its high solubility in water and high volatility. Industrial solvents are used in degreasing, electronics, metal finishing, fabric cleaning, pharmaceuticals, pesticides, antifreeze, membranes, paper manufacturing and many other applications. It is classified as a toxic chemical, hazardous pollutant and Class 2B carcinogen by the US Environmental Protection Agency [1]. Testicular tumours were seen in rats in a carcinogenicity study and evidence exists in rats that 1,4-dioxane has effects on certain sex hormones (i.e., is an endocrine disrupter) [2,3]. Similar observations have been made regarding human health effects. Effects included an increased incidence of miscarriages, premature births, maternal toxicosis and decreased birth weight [4]. It is thought to be the chief agent implicated in the cancers suffered by Vietnam military personnel (one of the

principal chemical components of Agent Orange) and is associated with increasing the chances of breast and endometrial cancer, stress-related illnesses and lower sperm counts [5]. 1,4-dioxane is resistant to biodegradation [6] and difficult to remove by conventional water treatment methods. Among the physical technologies capable of removing 1,4-dioxane, the most effective is distillation whereas carbon adsorption and air stripping are inadequate [7]. Due to the economic costs of distillation, chemical treatments have been considered as an alternative [8]. Nevertheless, the oxidation with chlorine or permanganate is ineffective, or could even lead to the formation of more toxic compounds [8–11].

Advanced oxidation technologies (AOTs) or processes (AOPs) are a group of related processes emerging to satisfy the demand for more effective and economical water and air remediation [12]. Various photochemical advanced oxidation processes have been suggested for the oxidation of organic compounds in water and wastewater. The most widely applicable ones are based on generation of hydroxyl radicals via photolysis of hydrogen peroxide, ozone and titanium dioxide [12]. Methods, which have been used in industry, include ultraviolet degradation, H₂O₂/UV and ozonation.

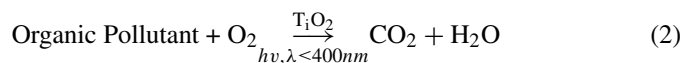
The mechanism most commonly accepted for the photolysis of H₂O₂ is the cleavage of the molecule into hydroxyl radicals

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and other reactive species that attack the organic molecules [13].



Concerning the new oxidation methods under development heterogeneous photocatalysis appears very promising to destroy organic micropollutants, allowing them to reach very low concentrations. Photocatalysis, as the name suggests, involves light and a catalyst to bring about a chemical reaction. In titanium dioxide photocatalysis for water purification, the pollutants are usually organic compounds and, therefore, the overall process can be summarised by the following reaction equation [14,15].



Advanced oxidation processes (e.g. ozone, Fenton's reagent, H₂O₂/ozone) have been evaluated for the degradation of 1,4-dioxane showing some degree of effectiveness [8]. Neither hydrogen peroxide nor ozone [10,11] alone readily oxidise 1,4-dioxane. Using Fenton's reagent the concentration of 1,4-dioxane and the related total organic carbon content was reduced within 10 h by 97 and 11%, respectively [9]. Ozonation and hydrogen peroxide combined are capable of disinfection but have been observed to form undesirable disinfection by-products [10]. We have previously investigated the use of the advanced oxidation technologies, titanium dioxide photocatalysis, photolysis and H₂O₂/UVC, for the degradation of 1,4-dioxane in water [16,17]. The results showed that TiO₂ photocatalysis with both UVA and solar light is effective in degrading 1,4-dioxane and is much more efficient than the H₂O₂/UVC process, UVA or UVC radiation alone. However, previous studies for photocatalysis of 1,4-dioxane in water have all involved a slurry TiO₂ reactor system [8,18–21]. The advantages of a slurry type reactor are the large surface area of catalyst and the intimate contact between the target compounds and the suspended particles, reducing mass transfer effects inherent in an immobilised system. The disadvantages are inhibition of light transmittance by the catalyst and the difficulties encountered when attempting to recover the particles from the treated effluent, due to the need for a solid–liquid separation process which is both time and energy consuming. A recently developed magnetic photocatalyst (MPC) [22], comprising of insulated magnetic core particles coated with a layer of photoactive TiO₂, provides a solution to this problem. The magnetic core allows for increased ease of separation of the particles from the treated effluent whereby the particles can be easily recovered by the application of a magnetic field. Preparation of the MPC consists of coating colloidal magnetite particles with an initial layer of SiO₂ and a subsequent layer of TiO₂. The insulative layer (SiO₂) is necessary to prevent interaction between the magnetic cores and the TiO₂, as any interaction leads to the photodissolution of the cores during irradiation [22]. Immobilisation of TiO₂ on the reactor walls was also investigated in this work as a solution to the solid–liquid separation problem. Immobilisation of the titanium dioxide involved a sol–gel method based on a procedure by Hong [23]. From the point of view of water treatment applications, immobilised TiO₂ has the advantages of easy operation and energy saving, but has a reduced rate of reaction due to mass transfer effects. Currently

many water treatment plants employ the H₂O₂/UVC process as a stage of treatment. Many research groups have investigated the effect of hydrogen peroxide on photocatalytic degradation of organic contaminants under the optimised catalyst loading conditions [24–26]. Poullos et al. [25] and Machado et al. [26] observed an enhancement whereas others have reported a negative effect of adding hydrogen peroxide for degradation of some organic pollutants [27,28]. This theory was investigated for the degradation of 1,4-dioxane using the P25 and MPC catalysts at varying concentrations of H₂O₂.

Therefore, the main aim of this paper is to optimise the AOPs previously investigated (photocatalysis and H₂O₂/UVC processes) for the removal of low concentrations of 1,4-dioxane in water. This involves investigating different photocatalysts (commercial Degussa P25 (P25), lab-made magnetic photocatalyst (MPC) and a sol–gel immobilised system) and loadings, a lifetime study of the immobilised sol–gel reactor, investigating the optimum concentration of H₂O₂ in the H₂O₂/UVC process and the effect of the addition of H₂O₂ on the photocatalytic system.

2. Experimental

2.1. Reactor set-ups and analytical method

The reactors were constructed using borosilicate glass (or quartz) tubing of 6 mm outer diameter and 1 mm wall thickness and constructed as a spiral reactor. Photocatalysis experiments were carried out in a borosilicate glass reactor with the catalyst in suspension or immobilised. A black-light blue fluorescent lamp (NEC, 20W, $\lambda = 300\text{--}400\text{ nm}$, $\lambda_{\text{max}} = 350\text{ nm}$) was fitted through the centre of the coil. A similar set-up was used for the H₂O₂/UVC experiments except the reactor was made from quartz and the light source was a slim line germicidal lamp (UV Air Pty Ltd, $\lambda = 254\text{ nm}$). Degussa P25 was employed as the commercial source of TiO₂ in suspension. Preparation of the MPC consists of coating colloidal magnetite particles with an initial layer of SiO₂ and a subsequent layer of TiO₂, details of which are given in Beydoun et al. [22]. Immobilisation of the titanium dioxide involved a sol–gel method based on a procedure by Hong [23]. The reactor was connected to a peristaltic pump (Masterflex[®] Quick–Load) by Masterflex[®] flexible tubing to enable solution circulation through the reactor at a flow rate of 465 ml/min. A schematic of the reactor set-up can be found in a previous publication [17]. The mineralisation of 1,4-dioxane was monitored by measuring the amount of carbon dioxide generated during the reaction, which was detected by an online conductivity meter [29].

2.2. Experimental procedures

2.2.1. Photocatalysis

Fifty millilitres of photocatalyst suspension (or MilliQ water for the immobilised system) was first introduced into the photoreactor and the pH of the suspension was adjusted to a value of 3 by the addition of perchloric acid (70%, Ajax Chemicals). A pre-determined volume of the 1,4-dioxane (99%, Riedel-

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