Electrocatalytic performance of PbO_2 films in the degradation of dimethoate insecticide

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Abstract

This study was performed to find the best experimental conditions for the electrochemical removal of the insecticide dimethoate ($C_5H_{12}NO_3PS_2$) from aqueous solutions using a lead dioxide niobium anode. The process was studied under galvanostatic polarisation mode. The influence of applied current density (10–50 $mA{\cdot}cm^{-2}$), initial chemical oxygen demand COD_0 (100–550 $mg{\cdot}\ell^{-1}$), temperature (30–70°C) and pH (3–11) on COD and instantaneous current efficiency (ICE) was studied. The results showed that almost 90% of COD removal was achieved under optimal experimental conditions, indicating that electrochemical oxidation on a PbO_2 anode is a suitable method for treatment of water polluted with dimethoate. It was found that the decay of COD generally followed a pseudo first-order kinetic and the oxidation rate was favoured by increasing the applied current density, temperature, pH and initial COD. The greatest COD removal (90%) was obtained when using an applied current density of 50 $mA{\cdot}cm^{-2}$, COD_0 = 320 $mg{\cdot}\ell^{-1}$, pH = 11, T = 70°C and electrolysis time = 8 h.

Keywords: Electrochemical degradation; hydroxyl radicals; organic pollutants; lead dioxide; wastewater

Introduction

Dimethoate (O,O-dimethyl S-[2-(methylamino)-2-oxoethyl] phosphorodithioate) (Fig. 1) is a widely used organophosphorus (OPs) insecticide and acaricide applied to kill houseflies, as well as a wide range of insects and mites on a variety of fruit, vegetable, field and forestry crops. Like all OPs, dimethoate acts by interfering with the activities of cholinesterase, an enzyme essential for the proper functioning of the nervous system of insects and humans. A study showed that toxic effects and cholinesterase inhibition were observed in adult humans who ingested 30 mg a day or a higher dosage of dimethoate for 57 days (Hayes and Laws, 1990). Dimethoate is very toxic to birds, bees, fish and aquatic invertebrates (Cheminova, 1991). Dimethoate is degraded in the environment to another, more toxic, pesticide, omethoate; the proportion of omethoate in the total residue reaches about 50% after 5 weeks (FAO/WHO, 1985). Powerful degradation methods are required to destroy these pollutants in order to avoid their dangerous accumulation in the aquatic environment.

Among the different technologies for wastewater treatment, biological oxidation is frequently used. However, if wastewater contains highly toxic compounds, biological treatment may be useless. For this reason, there has been an increasing interest in the use of new methods such as electrochemical oxidation. It has been proven to be a promising and attractive technique for the effective oxidation of wastewater containing organic compounds (Chen, 2004; Fernandes et al., 2004; Sanroman et al., 2004). Many studies have demonstrated that the complete mineralisation of organics can be obtained with high efficiency by direct electro-oxidation using only high oxygen overvoltage anodes such as SnO_2(Belhadj-Tahar and Savall, 1998; Comninellis and Pulgarin, 1993; Polcaro et al., 1999 Stucki et al., 1991), PbO_2 (Andrade et al., 2007; Feng et al., 1995; Feng and Li, 2003; Martinez-Huitle et al., 2004a; Panizza and Cerisola, 2008; Quiroz et al., 2005; Samet et al., 2010a) and boron-doped diamond (BDD) anodes (Bechtold et al., 2006; Iniesta et al., 2001; Panizza and Cerisola, 2005; Samet et al., 2010b; Weiss et al., 2008). For example, some studies reported that the current efficiencies obtained with Si/BDD in oxidising 4-chlorophenol (Gherardini et al., 2001), 2-naphthol (Panizza and Cerisola, 2004), chloranilic acid (Martinez-Huitle et al., 2004a), and chlorpyrifos pesticide (Samet et al., 2010b) were higher than those obtained with PbO_2 anodes. In contrast, Martinez-Huitle et al. (2004b) showed that the oxidation of oxalic acid was faster at Ti/PbO_2 than at BDD, because the interaction of the oxalic acid with the PbO_2 surface was particularly strong, and its anodic oxidation was limited only by mass transfer at higher current densities and lower substrate concentrations. Recently, Flox et al. (2009) have demonstrated that *m*-cresol is more rapidly removed with PbO_2 than with BDD.

Both PbO_2 and BDD anodes exhibit good chemical and electrochemical stabilities, long lifetimes, and wide potential windows for water discharge. However, in the case of BDD, the financial expense can represent a serious drawback for industrial-scale wastewater treatment. In contrast, PbO_2 anodes are mainly used in the electrolytic production of perchlorates and chlorates (Kuhn and Wright, 1971). These anodes are electrochemically deposited on the metal surface. Titanium, niobium, tantalum and zirconium are widely used as substrate materials for their well-known properties, such as resistance to corrosion and chemical attack by acids, alkalis or salt solutions, and high mechanical strength.

Various innovative technologies have been proposed for treatment of wastewaters containing dimethoate. These include the use of photocatalytic oxidation, using TiO_2 as catalyst (Chen et al., 2007; Evgenidou et al., 2005), photo-Fenton process (Nikolaki et al., 2005), thermal decomposition (Andreozzi et al., 1999), ozonation (Liu et al., 2008), and microwave

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Received 1 March 2012; accepted in revised form 16 November 2012.

http://dx.doi.org/10.4314/wsa.v39i1.5
Available on website http://www.wrc.org.za
ISSN 0378-4738 (Print) = Water SA Vol. 39 No. 1 January 2013
ISSN 1816-7950 (On-line) = Water SA Vol. 39 No. 1 January 2013