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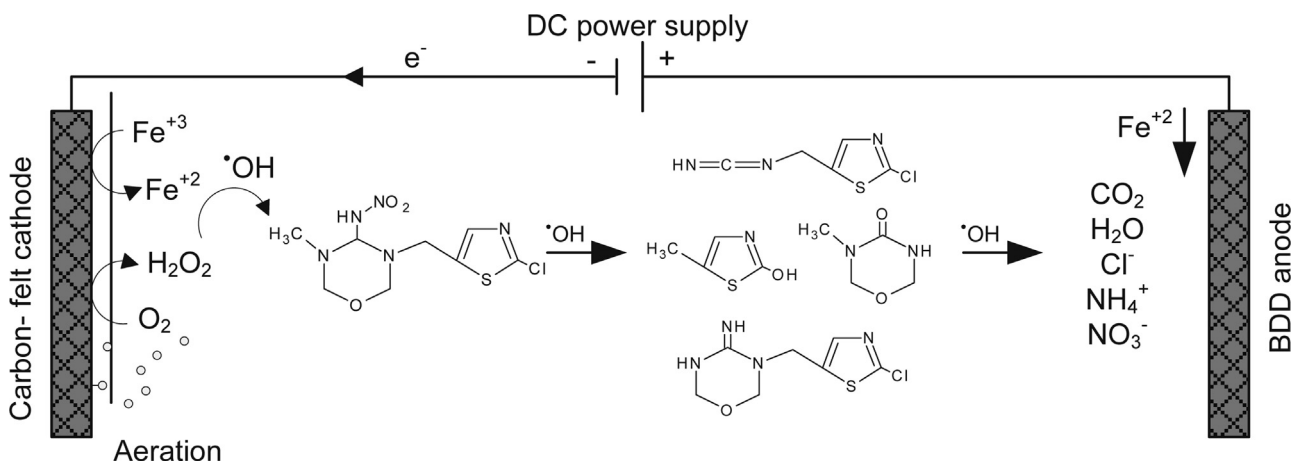


Degradation of thiamethoxam by the synergetic effect between anodic oxidation and Fenton reactions

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GRAPHICAL ABSTRACT



HIGHLIGHTS

- Electro-Fenton process demonstrates excellent ability to treat pesticides as thiamethoxam.
- Thiamethoxam and TOC removals strongly depend on the AOPs applied.
- Kinetics of thiamethoxam degradation and influence of operating conditions were studied.
- Potential mechanism and mineralization pathway of thiamethoxam were proposed.

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ABSTRACT

In this work, a comparative study using anodic oxidation, Fenton and electro-Fenton treatments was performed in order to determine the synergic effect for the removal of thiamethoxam. The results determined that electro-Fenton process showed high efficiency in comparison with Fenton or anodic oxidation. After that, this hybrid process was optimized and the influence of iron catalyst concentration and applied current intensity on the degradation and mineralization were evaluated. Degradation profiles were monitored by high performance liquid chromatography (HPLC) being satisfactorily described by pseudo-first order kinetic model. At the optimal experimental conditions (300 mA and 0.2 mM Fe²⁺), the complete degradation of thiamethoxam was achieved after 10 min. On the other hand, mineralization of thiamethoxam was monitored by total organic carbon (TOC) decay reaching more than 92% of TOC removal

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Fenton
Thiamethoxam

after 8 h. Furthermore, a plausible mineralization pathway for the thiamethoxam degradation was proposed based on the identification of by-products such as aromatic intermediates, carboxylic acids and inorganic ions released throughout electro-Fenton process.

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1. Introduction

The widespread use of neonicotinoid pesticides in agriculture represents a hazardous threat to the environment. These organic compounds have become one of the major polluting agents in the surface water and groundwater because of their leaching potential [1]. Neonicotinoid pesticides have a great insecticide activity which provides a continuous antimicrobial protection against a wide variety of commercially important sucking and chewing pests including some lepidopteran species, aphids, whiteflies, beetles, thrips and leaf miners [2].

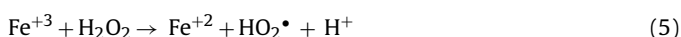
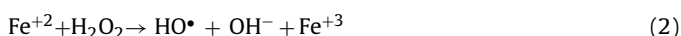
These pesticides act on the insect nervous system by attaching the nicotinic acetylcholine receptors [3]. Several authors reported negative influence of some neonicotinoids exposure on honeybee population [4–6]. As a consequence, the European Commission has restricted the use of three pesticides belonging to the neonicotinoid family (clothianidin, thiamethoxam and imidacloprid) for a period of two years [7].

Thiamethoxam belongs to the second generation neonicotinoid and possesses relatively low mammalian toxicity by oral, dermal and inhalation routes of exposure [8]. This pesticide contains a nitroguanidine functional group. The presence of thiamethoxam and other neonicotinoid pesticides has been detected at parts per million and parts per trillion concentrations in the water cycle [9]. Hence, several alternative technologies have been developed over the past two decades in order to achieve the water quality necessary to ensure safe and healthy aquatic ecosystem.

Among the different process available (physical, chemical or biological), advanced oxidation processes (AOPs) have been applied successfully for the degradation of toxic and persistent organic pollutants. The *in situ* generation of highly reactive species, such as the hydroxyl radical, allows that the degradation of organic matter takes place (Eq. (1)). AOPs are considered promising technologies with significant advantages such as operational cost and high mineralization degree of pollutants [10,11].



Among the AOPs, the Fenton treatment has attracted particular attention on the research community. Fenton's reagent (H_2O_2/Fe^{+2}) has been widely used for the generation of hydroxyl radical (Eq. (2)) and the effective degradation of organic pollutants. However, several drawbacks may hinder the large-scale application of Fenton's process including the cost of reagents and ferric sludge generation. Therefore, the combination of Fenton's reagents and electrochemical system (electro-Fenton process) represents an attractive method including continuous electrogeneration of H_2O_2 (Eq. (3)) and the regeneration of Fe^{+3} , generated from Fenton's reaction (Eqs. (4)–(7)), by direct reduction on the cathode or reduction processes involving H_2O_2 or intermediate organic radicals [12,13]. This allows for catalytic propagation of Fenton's reaction.



Up to now, only few studies have reported the application of electro-Fenton to the elimination of pesticides from aquatic environment. Oturan and Oturan [14] studied the degradation of three pesticides: Mistel GD (Cymoxanil + Mancozeb + additives), Cuprofix CZ (Cymoxanil + Zineb + $CuSO_4$ + additives) and Lannate 20L (Metomyl + ethanol + other additives) used in viticulture by electro-Fenton process. They showed that the electro-Fenton process is efficient for degradation of the active ingredients, and also the additives of the formulations. Other pesticides such as chlortoluron, carbofuran and bentazone were mineralized by electro-Fenton process using carbon-felt cathode and platinum anode [15]. Carbaryl, the second most frequently found insecticide in water, was degraded by electro-Fenton process using platinum or boron-doped diamond (BDD) anodes coupled with carbon-felt cathode [16]. Chloroacetanilide herbicide alachlor, a suspected human carcinogen classified as a priority water pollutant by the European Commission, has been quickly degraded and even totally mineralized by different electrochemical advanced oxidation processes (EAOPs) [17–20].

In our previous studies, electro-Fenton process was successfully applied to the degradation of different organic pollutants [21,22] and pesticides such as pirimicarb and imidacloprid [23,24]. The aim of this work was to evaluate the potential application of electro-Fenton process for the removal of a pesticide that has not been studied until now such as thiamethoxam. Initially, a comparative study using anodic oxidation and Fenton treatments was performed in order to determine the synergetic effect on electro-Fenton process. The effect on the degradation rate of different operating parameters such as catalyst concentration and applied current was studied. The degradation of thiamethoxam was followed by high performance liquid chromatography (HPLC) analysis. The decay of organic matter in the solution was followed by total organic carbon (TOC) measurements that permitted the evaluation of the mineralization efficiency. Besides, the identification of the degradation products of the thiamethoxam was accomplished using gas chromatography coupled with mass spectrometry (GC–MS) equipment and ion-exclusion HPLC.

2. Experimental

2.1. Pollutant and reagents

Thiamethoxam (3-[(2-chloro-5-thiazolyl)methyl]tetrahydro-5-methyl-N-nitro-4H-1,3,5-oxadiazin-4-imine) was evaluated as model neonicotinoid pesticide to determine its degradation ability by electro-Fenton. Thiamethoxam analytical standard (Pestanal[®] grade), iron(II) sulphate heptahydrate (catalyst, 99%) and anhydrous sodium sulphate (supporting electrolyte, 99%) were supplied by Sigma-Aldrich. Organic solvents were HPLC or analytical grade from Fisher Chemicals and Rathburn Chemicals. Oxalic, malonic, succinic, glycolic, formic, acetic and oxamic acids were provided from Sigma-Aldrich and Merck.

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