



Heterogeneous electro-Fenton treatment: preparation, characterization and performance in groundwater pesticide removal



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ABSTRACT

In this work the preparation, characterization and performance of Y-zeolite as catalyst for electro-Fenton treatment of pesticides was carried out. Initially iron supported Y-zeolite (Fe-Y) was prepared and evaluated for the degradation of imidacloprid and chlorpyrifos. Kinetic studies determined that the pesticides removal followed a pseudo-first-order kinetic model. However, the reusability of this catalyst was not appropriated and to enhance its recyclability, Fe-Y catalyst was embedded in alginate (Al-Fe-Y). The new catalyst showed similar degradation efficiency; and the recyclability was improved. This study demonstrated that Al-Fe-Y could be efficiently used to remove commonly pesticides, imidacloprid and chlorpyrifos, from aqueous medium.

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Introduction

Groundwater pollution is very difficult to mitigate due to the high residence time, the hard access and the ease of spreading long distances. A common approach for managing polluted groundwater is to remove or contain contaminant sources and to address down-gradient contamination using pump-and-treat (P&T) technology [1]. This technology is a classic ex-situ treatment which consists in two stages: pumping, where the polluted plume is extracted by pumping wells; and treatment, in which the pollutants are removed from the extracted water. The water treatment can be performed using mechanical, physical, biological, and chemical methods. However, it is necessary to take into account two main factors, the high amount of water to be treated and the nature of the pollution, in order to achieve an effective treatment.

The appearance of newly emerging organic contaminants is increasing in the groundwater [2]. These pollutants penetrate into the water body due to the runoff from urban areas, return flows from agricultural fields and leaching [3,4]; furthermore, they are

difficult to treat using conventional treatment technologies. Therefore, different approaches are being explored to accomplish an appropriated treatment of these pollutants [4–9]. Recently, the electro-Fenton process has attracted great interest from the scientific community because this treatment can be used to remove a wide variety of organic pollutants, including pesticides [10,11]. This technology combines the classical Fenton treatment with the electrochemical oxidation. Thus, the electrical current induces the in situ generation of H₂O₂ via reduction of oxygen, and the catalytic reaction is propagated by Fe²⁺ regeneration, which can take place by reduction of Fe³⁺ with H₂O₂, hydroperoxyl radical, organic radical intermediates, or directly at the cathode.

In order to couple this process to the P&T technology, it is necessary to design a system that can operate in continuous mode. In this context, the electro-Fenton treatment using heterogeneous catalyst arises. The iron immobilization onto a support provides a physical retention of the catalyst avoiding the iron release in the treated water. For this reason, this specific field attracted the attention of scientific researches worldwide [12–15]. In the last years, our research group has invested great efforts in the design of electro-Fenton reactors as well as developed several catalysts, iron alginate gel beads and iron-sepiolite, for heterogeneous Fenton and electro-Fenton degradation of dyes [11,16–19]. From these previous studies, it was determined that inorganic supports, such as clays, were adequate to perform heterogeneous catalyst.

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Zeolite is a well-known low-cost material widely used for metal removal [20,21] and in recent times, the scientific community increased its attention to use it as catalyst support. Fukuchi et al. [22] used iron-supported natural zeolite for the degradation of 2,4,6-tribromophenol in a heterogeneous Fenton-like system and Kiran et al. [23] utilized iron-modified zeolite catalyst for photocatalytic Fenton oxidation of sodium dodecyl sulfate. However, we could not find information about testing, iron supported zeolite as catalyst for the electro-Fenton treatment of pesticides. Therefore, the aim of this work is the development of a new electro-Fenton process using iron supported zeolite (Fe-Y) to treat pesticides typically present in groundwater. In this case, a Y-type zeolite, NaY, was selected due to its stable structure, large pore volume and cation exchange capacity; the presence of aluminum ions in the framework results in negative charges that are balanced with cations [24]. Initially the preparation and characterization of Fe-Y was performed and then the developed catalyst was tested for the electro-Fenton degradation of two model pesticides of different nature, imidacloprid and chlorpyrifos.

Material and Methods

Materials

Zeolite NaY supplied by Sigma-Aldrich (Barcelona, Spain) was evaluated as support for the catalyst.

Pesticide solutions were prepared using pesticide-grade Pestanal analytical standard provided by Sigma-Aldrich (Barcelona, Spain) at a desired concentration (Table 1).

Iron stock solution was obtained by dissolving $\text{Fe}_2(\text{SO}_4)_3 \cdot n\text{H}_2\text{O}$ (Sigma-Aldrich, Barcelona, Spain) in deionized water, each desired concentration was obtained by diluting this stock solution.

Na-alginate and CaCl_2 were analytical grade and supplied by Sigma-Aldrich (Barcelona, Spain).

Zeolite acid pre-treatment

Acid pre-treatment of Y zeolite was performed following Pazos et al. [25]. A suspension of zeolite and 0.1 M H_2SO_4 (ratio 1:10 g:mL) was maintained in agitation (150 ppm) at 25 °C for 24 hours. After that, the suspension was filtered through filter

paper and the zeolite was dried 24 hours in oven at 60 °C, separated from the paper and preserved.

Adsorption assays

Batch adsorption assays were carried out in 250 mL Erlenmeyer flasks by mixing 3 g of pre-treated Y zeolite with a constant volume (150 mL) of the iron aqueous solution at a desired concentration. The flasks were agitated in an incubator (Thermo Forma) at 150 rpm and 20 °C. In order to analyse iron adsorption, samples were taken of the supernatant and were centrifuged (Sigma 3K-18) during 15 min at 5000 rpm to remove solid. Atomic Absorption Spectroscopy (Agilent 240FS) was used to measure the iron that remained unadsorbed in the supernatant liquid.

Iron uptake (q) concentration was determined by the following equation:

$$q = (A_i - A_f)/m \quad (1)$$

Where A_i is the initial amount of iron in solution (mg), A_f is the final amount of iron in solution (mg) and m is the mass of utilized Y zeolite (g).

All the adsorption studies were repeated three times; the reported value is the average of measurements, the experimental error was calculated as the standard deviation, which was below 3% in all cases.

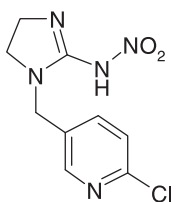
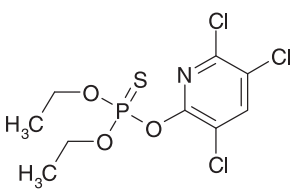
Catalyst preparation

The Fe-Y obtained from the adsorption assays, at a concentration of 52.21 mg g^{-1} , was directly used as catalyst and was also tested embedded in alginate gel. For the later, a suspension of Na-alginate (3%) containing Fe-Y was dropped on a solution of 0.2 M CaCl_2 to create the spherical alginate beads with the Fe-Y zeolite entrapped into its structure.

Characterization of catalyst

Fourier-transform infrared (FT-IR) spectra of the Y zeolite and Fe-Y were recorded on an FT-IR spectrometer (model FT-IR/4100, Jasco). The samples were ground into powder and dried in an oven at 60 °C for 1 h. The potassium bromide (KBr) pellet

Table 1
Type, CAS number, structure, solubility and concentration of studied pesticides.

Pesticide	Type	CAS number	Structure	Solubility water 20 °C ($\text{mg}\cdot\text{L}^{-1}$)	Concentration ($\text{mg}\cdot\text{L}^{-1}$)
Imidacloprid 1-(6-chloro-3-pyridylmethyl)- N-nitroimidazolidine-2-ylideneamine	chloronicotinyl nitroguanidine	13826-41-3		610	100
Chlorpyrifos O, O-diethyl O-(3, 5, 6-trichloro-2-pyridyl)- phosphorothioate	organophosphate	2921-88-2		2	1.3

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