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Adsorption with catalytic oxidation in a recirculating bed reactor for contaminated groundwater



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ABSTRACT

A novel nanoremediation concept, which is based on in situ trapping of organic contaminants by adsorption and catalytic oxidation in combination with oxidants such as hydrogen peroxide is presented. In earlier works we explored the porous structure of a modified natural clinoptilolite loaded with iron as a supported catalyst (NZ -A-Fe). The supported catalyst prevents iron release during all the process. This paper presents novel results for BTX (Benzene, Toluene and Xylene) removal from aqueous solution considering that adsorption and oxidation processes are taking place simultaneously. The experiment was achieved by fluxing an aqueous solution of BTX 3.3 mM and hydrogen peroxide, at neutral pH, using the same reservoir. After 870 min, C/C0 reaches near 10% for each pollutant. The system removed 65 mg BTX in the present conditions (13 mg/g NZ-A-Fe). The results indicate that the recirculation bed reactor is an excellent system to remove by adsorption and oxidation processes BTX from water due to high mass transfer coefficients and other advantages when compare to batch reactor experiments.

1. Introduction

Water is one of the most valuable resources on planet earth. During the last few decades, the rise of world population as well as industrial revolution has caused serious environmental pollution. In order to reduce risks to human and ecological receptors, considerable time and money have been spent remediating these sites since passage of major environmental legislation [1,2]. Benzene, toluene and xylene (BTX) isomers are monocyclic aromatic hydrocarbons, which have a moderate solubility in water at 20 °C (benzene: 22 mM; toluene: 5.1 mM and xylenes: 1.5 mM). These compounds are the major water-soluble constituents of petroleum derivatives (gasoline) [3].

Several processes have been examined for removal of BTX compounds from aqueous environment including oxidation, bioremediation and adsorption [4]. Adsorption processes involving porous solids are the most popular, and are widely utilized in engineering practice since they permit the recovery of these compounds. Shehdeh Jodeh et al. [5] reported that the use of activated carbon produced from date stones to remove the BTX from water is a good and efficient method and its effect of activating agent on the adsorption efficiency of BTX was also studied. They worked with low BTX solutions concentration (50 mg/L). Bandura et al. [6] examined the adsorption of BTX from aqueous solution by synthetic on zeolite Na-P1 obtained from fly ash and reported that the sorption capacity, expressed as the sum of BTX adsorbed per $1\,g$ of zeolite $Na\text{-}P_1$ was about $0.34\,\text{mg/g}$

To overcome adsorption restriction, the use of advanced oxidation processes (AOPs) has been widely proposed to treat wastes, particularly less concentrated effluents. AOPs are a group of processes based on the generation of highly reactive radicals, especially hydroxyl radicals, which are extremely active and nonselective oxidants, being able to oxidize a wide range of compounds that are otherwise difficult to degrade [7,8]. Currently, the hydroxyl radical OH° is one of the most reactive chemical species known, second only to fluorine in its reactivity [9].

Among the hydrogen peroxide-based AOPs, treatment with Fenton's reagent (dissolved Fe2 + H2O2) is a well- known, effective, and inexpensive way to degrade organic compounds in wastewater [1]. Regarding the simplicity of catalyst removal and recovery, heterogeneous catalysts are more desirable than homogeneous ones.

Beside the use of solid iron-oxide minerals or zero-valent iron, several solid supports such as zeolites [10,11], activated carbon, clays resins or mesoporous materials have been studied for immobilization of iron species in heterogeneous Fenton-like catalysts [12].

Natural zeolites have been the subject of investigation by many researchers who took advantage of the zeolites physical and chemical properties and used them in heavy metals adsorption, water

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purification and softening, soil remediation, etc. During these studies many experimental variables were investigated, such as temperature, solution ionic strength, concentration of adsorbates, contact time and present cationic/anionic species [13–15].

In previous works, the porous structure of this modified natural clinoptilolite as a supported catalyst (NZ-A-Fe) has been reported [16–18], where the behavior of this material (NZ-A-Fe) in both adsorption and oxidation processes were explored.

This paper presents novel results for BTX removal considering that simultaneous processes are taking place. The experiment was achieved by fluxing an aqueous solution of BTX 3.3 mM and hydrogen peroxide using the same reservoir.

2. Experimental

2.1. Materials and methods

Benzene (Bz) was purchased from Merck Química, toluene (Tol) was obtained from Biopack Chemical and *p*-xylene (Xyl) was obtained from Carboclor SA.

Powdered natural zeolite (NZ) with an average particle in the range 0.3-0.5 mm was supplied by the DIATEC S.R.L., extracted from La Rioja, Argentina. Iron sulphate (FeSO4·7H2O) purchased from Cicarelli p.a. was used as an iron precursor for ion exchange.

A stock BTX-contaminated groundwater, was prepared by adding the required volumes (μ l) of Benzene (Bz), Toluene (Tol) and Xylene (Xyl) into a buffered solution to maintain pH = 7 during all the experiences. Hydrogen peroxide was added to the feed solution. The final concentration of pollutants was 1.8, 1.1 and 0.4 mM in Bz, Tol and Xyl respectively. The buffer solution was prepared adding the adequate amounts of Na₂HPO₄ and NaH₂PO₄ in deionized water. The flasks neck containing the stock solution was sealed with Teflon tape and it was stirred for 24 h until complete dissolution of the organic compounds.

2.2. Preparation of fe-loaded zeolite (NZ-A-Fe)

As it was earlier described, samples of this natural zeolite (NZ) were sieved and treated in $NH_4Cl \ 3 M \ at \ 353 K \ during \ 8 h \ (sample \ NZ-A).$ Later these samples were batch-loaded with iron salts (NZ-A-Fe(II)) and chemically reduced with sodium borohydride in order to prepare the catalyst NZ-A-Fe [18]. The used catalyst remained inside the column, at the end of the experiences, was named as NZ-A-Fe(u).

2.3. Characterization analysis

The crystallinity of the zeolite samples was studied using X-ray diffraction (XRD). The spectra were obtained on a Siemens XRD instrument, using filtered Cu Ka radiation with accelerating voltage of 40 kV, current of 30 mA and scanned at 20 from 5° to 55°. The samples profiles were analyzed with the software X'Pert for phase's identification and with PowderCell for refining the structural parameters. Elemental analysis was estimated with Energy Dispersive Spectroscopy (EDS) in a Sigma Zeiss microscope with an Oxford EDS detector. The concentration of freely dissolved BTX was determined by Gas Chromatography (GC) analysis. Iron leaching was analyzed using an atomic absorption spectrometer (AAS) instrument while zeolités iron content was quantitatively determined by redox titration with the phosphoric technique [19]. UV-vis/diffuse reflectance spectra (DRS) of solids were recorded using UV-vis spectrophotometer with an additional integrating sphere Diffuse Reflectance Accessory and BaSO4 as standard. The software used in this analysis is PeakFit by SYSTAT Software Inc.

2.4. Heterogeneous fenton reaction

The reactor consisted of a column 9.0 cm length and 1.0 cm in



Fig. 1. Schematic diagram of the experimental setup for column experiments.



Fig. 2. DRX of zeolite samples: natural zeolite (NZ), natural zeolite treated with amonnium chloride (NZ-A) and this sample loaded with iron and chemically reduced with sodium borohydride in order to prepare the catalyst (NZ-A-Fe).

Table 1

Chemical atomic composition (at%) of the natural clinoptilolite NZ and its modified form loaded with iron.

	NZ	NZ-A-Fe
0	42.37	47.27
Na	3.20	2.80
Mg	1.03	0.71
Al	8.60	7.68
Si	38.3	34.2
S	0.62	1.18
К	1.36	1.10
Ca	2.79	2.91
Fe	0.70	1.50
Si/Al	4.5	4.5

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