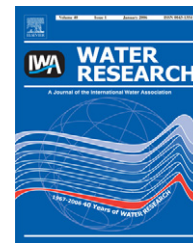


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Iron(3) oxide-based nanoparticles as catalysts in advanced organic aqueous oxidation

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ABSTRACT

Water contaminated with dissolved organic matter is an important issue to resolve for all-purpose uses. The catalytic behavior of iron-based nanocatalysts was investigated for the treatment of contaminated water in the advanced chemical oxidation process. In this study, typical organic contaminants, such as ethylene glycol and phenol, were chosen to simulate common contaminants. It was shown that the two substances are efficiently destroyed by the Fenton-like reaction using iron(3) oxide-based nanocatalysts in the presence of hydrogen peroxide without the need for UV or visible radiation sources at room temperature. A strong effect of nanocatalyst concentration on reaction rate was shown. The kinetic reaction was found and the reaction rate coefficient k was calculated.

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1. Introduction

Organic pollutants are often present in drinking water, groundwater, and domestic and industrial wastewaters. A water treatment based on the chemical oxidation of organic compounds by advanced oxidation processes (AOPs) that is useful for purifying drinking water, groundwater and for cleaning industrial wastewater has been reported recently (Sigman et al., 1997; Yeber et al., 2000; Perez et al., 2002). Several of these studies have focused on using these systems as a pre-treatment for biological systems when the dissolved organic matter is toxic, inhibitory or recalcitrant to micro-organisms. The degradation and mineralization of organic pollutants in wastewater by AOPs is based on the generation of a very reactive free hydroxyl radical (OH^*). This radical is

highly reactive, non-selective and may be used to degrade a wide range of organic pollutants. It reacts with most organic compounds by forming to a double bond or by abstracting hydrogen atoms from organic molecules (Safarzadeh-Amiri et al., 1997; Buxton et al., 1988). The resulting organic radicals then react with oxygen, which leads to the complete mineralization of CO_2 , H_2O and mineral acids (Oliveros et al., 1997; Neyens and Baeyens, 2003). Fenton and Fenton-like systems ($\text{Fe}^{+2}/\text{Fe}^{+3}/\text{H}_2\text{O}_2$) are often used for industrial water treatment based on AOPs (Fenton, 1894; Neyens and Baeyens, 2003). The degradation rate of organic pollutants with Fenton reagents strongly depends on irradiation with ultraviolet (UV) light, and increases with increased UV irradiation intensity (Safarzadeh-Amiri et al., 1996). The use of UV light results in a significant increase in the cost of

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Notation			
AOP	advanced oxidation process	k	reaction rate coefficient, 1/min
TOC	total organic carbon	m	order of reaction
UV	ultraviolet	R	initial ratio of organic solute to hydrogen peroxide concentration
C	concentration of organic pollutants (ethylene glycol or phenol) in solution, ppm	r	ethylene glycol to hydrogen peroxide initial concentrations ratio, ppm/ppm
C_0	initial concentration of organic pollutants, ppm	t	time, min
C_{Fe}	nanocatalyst concentration, ppm	$t_{1/2}$	half-life time, the time required to decrease the initial concentration by half
$C_{H_2O_2}$	initial hydrogen peroxide concentration	α	constant, min
EG	ethylene glycol	β	constant, dimensionless

industrial water treatment. Post-treatment requires the elimination of Fenton reagents as colloidal precipitates. The separation of colloidal precipitates requires the use of additional processes such as coagulation, sedimentation and filtration.

Nanoparticles of inorganic materials, such as metal oxides and semiconductors, have generated considerable attention due to their novel properties compared with their bulk materials (Iwasaki et al., 2000; Kamat and Meisel, 2002). A number of reports have shown that iron oxide has special photochemical and catalytic properties that strongly depend on particle size (Stramel and Thomas, 1986). However, only a few studies of the catalytic activity of colloidal iron-based nanoparticles have been made.

Catalyst recovery is required following water treatment using AOPs. In order to avoid this step, nanocatalysts may be immobilized on inert surfaces, foams or nanofibers without a reduction in catalytic activity (Bauer et al., 1999; Yeber et al., 2000). Therefore, investigation of the effectiveness and applicability of new nanoparticle catalysts may be divided into the following main steps:

- Synthesis of nanoparticle catalysts and testing of their catalytic behavior.
- Immobilization of nanocatalysts on inert surfaces, foams or nanofibers, and investigation of their catalytic behavior.
- Investigation of the regeneration of the adsorbent and the nanocatalyst in successive catalytic runs.

The objective of this research work is to continue the efforts of the first step: to test the catalytic behavior of iron oxide-based nanocatalysts through the degradation and mineralization of organic pollutants in wastewater as a possible method of treating contaminated groundwater and industrial wastewater.

For this study, typical organic contaminants, such as ethylene glycol and phenol, were chosen as simulating pollutants. Ethylene glycol is used in large quantities as a car-cooling fluid or as an airplane and runway deicer. Large quantities of ethylene glycol have created environmental hazards, leading to the serious pollution of drinking water. Several types of industrial waste contain phenols; they are very harmful and highly toxic towards microorganisms (Cheng et al., 1995). Many phenol compounds are used as

solvents or reagents in industrial processes and are therefore very common contaminants in industrial wastewater and contaminated drinking water sources.

2. Experimental

2.1. Materials

Iron chloride hexa-hydrate, $FeCl_3 \times 6H_2O$ (analytical grade; Merck KGaA, Germany), 30% hydrogen peroxide (analytical grade; PA, Panreac Quimica SA), phenol (analytical grade; Fluka) and chemically pure ethylene glycol (BIO LAB Ltd., Israel) were used as received.

2.2. Analysis and equipment

A morphology study was performed using cryogenic transmission electron microscopy (cryo-TEM), a Philips CM120 TEM optimized for cryo-TEM work equipped with an Oxford Instruments CT-3500 cooling holder system. The pH was determined using a Consort P-901 electrochemical analyzer. Total organic carbon (TOC) and phenol content analyses were carried out using a TOC-5000A Shimadzu analyzer and a Hach DR/2010 data-logging spectrophotometer with the phenols 4-aminoantipyrine method.

2.3. Preparation of iron(3) oxide-based nanocatalysts

The preliminary material used for preparing the colloidal iron nanoparticles was iron chloride hexa-hydrate, $FeCl_3 \times 6H_2O$ (analytical grade; Merck). Hydrolysis was used to prepare a 10% sol with iron nanocatalysts with initial acidity (pH = 0.8). A series of iron oxide-based nanocatalysts was then prepared by diluting the initial solution. Cryo-TEM was used for characterizing the prepared nanoparticle material. Fig. 1 shows an example of a cryo-TEM image of a cluster of needle-like nanoparticles. The corresponding electron diffraction pattern (Fig. 1B) reveals that the needle-like nanoparticles seem to be crystals with lattice constants (characteristic inter-planar distances) of 3.3, 2.48 and 1.75 calculated by the Bragg equation. Analogous cryo-TEM images of nanocatalysts were obtained for pH values used in the catalytic process in the pH range 2–3.5. Currently, there is no clear identification of the crystalline structure of the

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