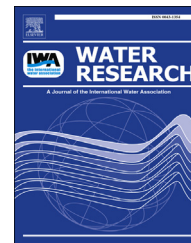


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Effect of dissolved oxygen concentration on iron efficiency: Removal of three chloroacetic acids



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ABSTRACT

The monochloroacetic, dichloroacetic and trichloroacetic acid (MCAA, DCAA and TCAA) removed by metallic iron under controlled dissolved oxygen conditions (0, 0.75, 1.52, 2.59, 3.47 or 7.09 mg/L DO) was investigated in well-mixed batch systems. The removal of CAAs increased first and then decreased with increasing DO concentration. Compared with anoxic condition, the reduction of MCAA and DCAA was substantially enhanced in the presence of O₂, while TCAA reduction was significantly inhibited above 2.59 mg/L. The 1.52 mg/L DO was optimum for the formation of final product, acetic acid. Chlorine mass balances were 69–102%, and carbon mass balances were 92–105%. With sufficient mass transfer from bulk to the particle surface, the degradation of CAAs was limited by their reduction or migration rate within iron particles, which were dependent on the change of reducing agents and corrosion coatings. Under anoxic conditions, the reduction of CAAs was mainly inhibited by the available reducing agents in the conductive layer. Under low oxidic conditions, the increasing reducing agents and thin lepidocrocite layer were favorable for CAA dechlorination. Under high oxidic conditions, the redundant oxygen competing for reducing agents and significant lepidocrocite growth became the major restricting factors. Various CAA removal mechanisms could be potentially applied to explaining the effect of DO concentration on iron efficiency for contaminant reduction in water and wastewater treatment.

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1. Introduction

In recent years, zero valent iron (Fe⁰) and Fe⁰-doped particles have been shown very efficient for the aqueous removal of various inorganic and organic contaminants (Cundy et al.,

2008; Fu et al., 2014; Gunawardana et al., 2011; Henderson et al., 2007; Li et al., 2006; Noubactep, 2008; Scott et al., 2011). The potential effect of the coexisting oxidants on iron corrosion, such as oxygen (O₂) or chlorine, has attracted considerable attention (Jung et al., 2011; Rahman and Gagnon, 2014; Sarin et al., 2004a, 2004b; Stratmann and Müller, 1994; Wang

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et al., 2012; Zhang and Huang, 2006). However, limited investigation have been completed examining the influence of O_2 on the removal of contaminants by iron (Ghauch et al., 2010, 2011; Huang and Zhang, 2005; Wang et al., 2010). The rate constant for carbon tetrachloride reacting with iron under an oxic condition was significantly lower than that under an anoxic condition (Helland et al., 1995). The presence of O_2 in the iron-water system decreased the removal efficiency of nitrate (Westerhoff and James, 2003) and bromate (Xie and Shang, 2007), and slowed the reduction of bromoacetic acid (Zhang et al., 2004), trichloropropanone (Lee et al., 2007) and diclofenac (Ghauch et al., 2011). The rapid removal of carbon tetrachloride and trichloroethylene using iron and palladized-iron cathodes was not interfered even when purging the raw water with air (Li and Farrell, 2000). The nitrate reduction could maintain stable under various oxic conditions if amending iron with the aqueous ferrous ion (Huang and Zhang, 2005). The existing O_2 failed to inhibit with the quick degradation of tribromoacetic acid, trichloronitromethane and trichloroacetonitrile, which were mass transfer limited species (Lee et al., 2007; Zhang et al., 2004). Some attractive results have been reported on the enhancing effect of O_2 on iron efficiency for the removal of dye (Wang et al., 2010) and diclofenac (Ghauch et al., 2010) in a batch system, and the reduction of nitrate (Westerhoff and James, 2003) and chromium (Yoon et al., 2011) in a packed column.

A potential mechanism for different O_2 roles was involved the formation of various iron corrosion products, which served as a physical barrier, a semiconductor or a coordinating surface in the core-shell structure of iron corrosion (Scherer et al., 1998; Sarin et al., 2004a; Yan et al., 2010). Uludag-Demirer and Bowers (2003) reported O_2 acted as an irreversible inhibitor of the trichloroethylene reduction based on the assumption of magnetite and maghemite formed on the iron surface. Huang and Zhang (2005) reported maghemite or lepidocrocite produced under oxic conditions would decrease the removal efficiency of iron, while magnetite even in a substantial thickness might not impede the nitrate reduction. In these researches, Fe^0 was widely regarded as an effective reductant (direct reduction), which was responsible for the decrease of contaminants (Gillham and O'Hannesin, 1994; Li et al., 2006; Matheson and Tratnyek, 1994). Due to ubiquitous oxide films decreasing the accessibility of iron surface, the observed removal of contaminants was predominantly mediated by aqueous and/or solid corrosion products through adsorption, co-precipitation and subsequent indirect reduction by other reducing agents except for Fe^0 (Noubactep, 2008, 2010, 2011, 2014; Noubactep et al., 2010). The reducibility of aqueous and solid iron corrosion was usually ignored, although the aqueous, absorbed or structural divalent iron (Fe^{II}) was thermodynamically capable of reducing some pollutants (Chun et al., 2005, 2007; Pecher et al., 2002; Schlautman and Han, 2001; White and Peterson, 1996). The enhancement on removal efficiency of iron was reported with coexisting iron minerals, such as magnetite (Coelho et al., 2008; Huang and Zhang, 2006; Mak et al., 2011), green rust (Cho et al., 2010), pyrite (Kim et al., 2013) and ferric hydroxide (Song et al., 2013). The availability of aqueous or solid Fe^{II} mentioned above was almost conducted under anoxic condition to restrain the rapid oxidation of Fe^{II} by O_2 . Hence, the

evolution of reducing ability or agents for contaminants in iron/water system under various oxic conditions was necessary to investigate in order to promote its application in the treatment of surface water and wastewater.

As important byproducts of the chlorination of water and wastewater, the chloroacetic acids (CAAs) including monochloroacetic acid (MCAA), dichloroacetic acid (DCAA) and trichloroacetic acid (TCAA) were selected as the model compounds. The batch experiments with a controlled DO concentration ranged from 0 to 7.09 mg/L were analyzed over a 2-h reaction period, during which the liquid and solid were periodically sampled to determine the evolution of CAA degradation processes and iron corrosion products. The removal mechanism of CAAs affected by various DO concentrations was also explored in the iron/water system.

2. Materials and methods

2.1. Chemicals

The analytically pure MCAA, DCAA and TCAA were bought from the Tianjin Fuchen Chemical Regents and Sinopharm Chemical Reagent (China), respectively. The standard solution for CAAs (2000 mg/L each), formic acid, ammonium formate and sodium carbonate (analytical reagent) were purchased from Sigma–Aldrich (USA). The chromatographically pure methanol and acetonitrile were obtained from Fisher Chemicals (USA). The analytically pure sodium chloride, hydrochloric acid, sodium hydroxide, ferrous chloride and magnetite were bought from Beijing Chemical Works (China). Akaganeite substituted for lepidocrocite was synthesized for convenience (Chitrakar et al., 2006).

2.2. Iron pretreatment

The raw iron particles were collected from a local manufacturing factory, which had some grease on the surface. The particles were first passed through two sieves (0.2 and 1.0 mm), then soaked in a detergent solution for 24 h for oil removal, then rinsed in sequence with tap water, deionized water and ultrapure water for residual detergent removal, and finally stored in ultrapure water. The final iron particles contained black coatings and the specific surface area measured via BET was 1.50 m²/g. Fresh iron without oxide films was prepared with 1 M hydrochloric acid lasting for 10 min, then rinsed with deoxygenated water until no chlorine ion detected in water.

2.3. Dissolved oxygen control

Refreshing the headspace at a 10-min interval was an effective way to maintain a stable DO concentration in the reactor. This operation consisted of the following steps. (1) A reactor was taken down from the rotator. (2) Two needles were inserted through the silicone stopper of the reactor and one of them was connected with high-purity nitrogen gas (N_2) lasting for 20 s. (3) A specific amount of O_2 was injected into the headspace. (4) Two needles were pulled out after waiting for 3–5 s, ensuring that the gas pressure in the reactor was the same as

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