



Fenton oxidation of organic contaminants with aquifer sediment activated by ascorbic acid

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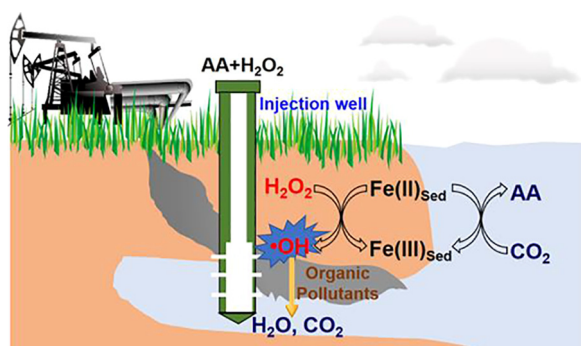
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HIGHLIGHTS

- H₂O₂ is activated by iron-containing minerals in aquifer sediments and AA.
- Goethite and hematite are more reactive to activate H₂O₂ in the presence of AA.
- Organic pollutants could be efficiently removed with Sed/AA/H₂O₂.
- The injection of AA/H₂O₂ is promising to remediate contaminated groundwater.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, we report the desirable conversion of H₂O₂ to ·OH with aquifer sediment activated by ascorbic acid. Although aquifer sediment contained various iron minerals, such as goethite, hematite, montmorillonite, muscovite, chlorite and phlogopite, it could not efficiently activate H₂O₂ to produce ·OH. Interestingly, the presence of ascorbic acid dramatically promoted the ·OH conversion efficiency of H₂O₂ by aquifer sediment to remediate organic contaminants in groundwater, which was ascribed to the efficient iron cycle of aquifer sediment induced by ascorbic acid. Among these minerals in sediment, goethite and hematite were more reactive than other minerals to activate H₂O₂ in the presence of ascorbic acid. Although the initial pH could affect the ·OH conversion efficiency of H₂O₂, H₂O₂ could be decomposed by aquifer sediment at pH 3–11 after the introduction of ascorbic acid. Along with the complete degradation of organic contaminants, the final pH of solution increased to 8.3, which was close to the initial pH (8.9) of groundwater. Moreover, packed column of aquifer sediment could also catalyze the H₂O₂ conversion to ·OH for the removal of different organic contaminants with adding ascorbic acid, suggesting a new strategy for in situ chemical oxidation remediation of organic contaminated groundwater.

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1. Introduction

Recently there is growing concern about groundwater pollution of organic matters releasing from industrial production, agricultural production and domestic waste. Many organic contaminants in groundwater, such as nitrobenzene compounds, organochlorine pesticides and dyes, are persistent in the environment and carcinogenic to humans [1–7]. Organic contaminated groundwater can be remediated with physical treatment, bioremediation and chemical oxidation techniques [8–12]. Among these techniques, in situ chemical oxidation (ISCO) with using strong oxidants (such as permanganate, persulfate, and H_2O_2) become more and more attractive because of their high efficiencies. For instance, permanganate with redox potential of 0.59–1.7 eV could oxidize several contaminants quickly, such as chlorinated ethenes, 2,4,6-trinitrotoluene, pesticides aldicarb, dichlorvos and methyl tert-butyl ether [13–16]. Persulfate and H_2O_2 could be decomposed to produce sulfate radical ($\text{SO}_4^{\cdot-}$) (2.5–3.1 eV) and hydroxyl radical ($\cdot\text{OH}$) (2.8 eV), which can degrade or even mineralize a wide range of organic pollutants [17–20]. In comparison with permanganate and persulfate chemical oxidation, Fenton oxidation with using H_2O_2 is the most widely used ISCO method for the remediation of organic contaminated groundwater, because H_2O_2 is inexpensive and eco-friendly as its decomposition byproducts are O_2 and H_2O [21–24]. Unfortunately, traditional ISCO remediation of contaminated groundwater with Fenton oxidation technology suffers from extra iron injection and low efficiency caused by the poor Fe(III)/Fe(II) cycle.

In general, the near neutral or slightly alkaline nature of groundwater results in the wide existence of iron bearing minerals in aquifer sediments around groundwater [25]. These iron minerals can influence the migration and transfer of contaminants in groundwater. For instance, Yuan et al. reported that Fe(II)-containing minerals, such as montmorillonite, biotite and chlorite, could react with O_2 to produce $\cdot\text{OH}$ at the oxic/anoxic interface in the aquifer, which induced the oxidation of As(III) in the aquifer sediments to the less toxic As(V) and the degradation of tetracycline [25]. Moreover, Sedlak and his coworkers found that iron oxide and iron-containing clay minerals in the aquifer materials could decompose H_2O_2 of high concentration (250 mmol/L) to produce $\cdot\text{OH}$, leading to the degradation of phenol at pH 8.2–8.7 [17]. Thus, iron minerals in aquifer sediments might be the inherent iron source for the ISCO remediation of contaminated groundwater with Fenton process. Unfortunately, the Fenton oxidation efficiency of aquifer sediments was found to be very poor for the remediation of contaminated groundwater (SM Fig. S1), which might be ascribed to the poor Fe(III)/Fe(II) cycle of iron minerals according to our previous studies [4,26,27].

Ascorbic acid, a natural reductant and ligand, can influence the migration and transformation, the dissolution and settlement, and the bioavailability of iron minerals [28–34]. Meanwhile, the coupling interaction of ascorbic acid and iron cycles could also affect the transformation of toxic elements and organic pollutants [35,36]. For instance, ascorbic acid could promote the Fe(III)/Fe(II) cycle to efficiently degrade alachlor in the Fenton-like (Fe(III)/ H_2O_2) system [37]. However, it is still unknown whether ascorbic acid can enhance the H_2O_2 conversion efficiency of real aquifer sediments by enhancing the Fe(III)/Fe(II) cycle for the ISCO remediation of contaminated groundwater.

Herein we first systematically investigate the effect of ascorbic acid on the activation of H_2O_2 with relatively low concentration (10 mmol/L) by aquifer sediments, and then study Fe(III)/Fe(II) cycle, ROS generation, and organic pollutants degradation efficiency of ascorbic acid/aquifer sediment Fenton system. Finally, we utilize packed column of aquifer sediment to catalyze the conversion of H_2O_2 to $\cdot\text{OH}$ for the removal of different organic contaminants in the presence of ascorbic acid, aiming to simulate the ISCO remediation of organic contaminated groundwater.

2. Experimental section

2.1. Chemicals and materials

Ascorbic acid (AA), sodium ascorbate (SA), rhodamine B, sodium hydroxide (NaOH), hydrochloric acid (HCl), gauze and quartz sand were obtained from Sinopharm Chemical Reagent Co., Ltd. *p*-chloronitrobenzene, chloramphenicol, bipyridine and 5,5-dimethyl-1-pyrroline-N-oxide (DMPO) and alachlor were obtained from Alfa Aesar. Montmorillonite, muscovite and chlorite were purchased from the American Clay Minerals Society. Phlogopite was achieved from Shijiazhuang Huabang Mineral Products Co., Ltd. All the chemicals and materials were used as obtained without further purification. The aquifer sediment used in this study (SM Fig. S2) was sampled from an aquifer located within 1–6 m under the ground surface near Yangtze river in Wuhan city (113.678331°E, 30.175845°N), Central China with using a pocket earth drill (Australia, CHRISITE, SD-1). The groundwater table was 2 m below the ground at the time of sampling. The aquifer sediment was sealed and stored in the refrigerator after sampling. The physicochemical properties of aquifer sediment were summarized in SM Table S1.

2.2. H_2O_2 decomposition

The H_2O_2 decomposition experiments were conducted in aquifer sediment suspensions under typical anaerobic and dark groundwater environment at $17 \pm 2^\circ\text{C}$. Typically, suspensions were prepared with aquifer sediment concentration of 5 g/30 mL with using anaerobic deionized water or simulated groundwater (SM Table S2). In this process, the aquifer sediment was used as obtained without further treatment. Then, the experiments were initiated by adding 100 μL of 0.75 mol/L AA solution and 250 μL of 1.2 mol/L H_2O_2 solution into the aquifer sediment suspensions in sequence. The initial pH values of aquifer sediment suspensions were about 8.9. The initial pH values of aquifer sediment and AA solution were about 5.7. The concentration of H_2O_2 was analyzed by a modified *p*-hydroxyphenylacetic acid (POHPAA) fluorescence method [26]. Briefly, the fluorescence reagent was prepared by adding 1 mg of horseradish peroxidase and 2.7 mg of POHPAA into the 10 mL of 8.2 g/L potassium acid phthalate aqueous solution. Then, 2 mL of reaction solution was taken with subsequently adding 50 μL of the fluorescence reagent. After reaction for 10 min, 1 mL of 0.1 mol/L NaOH solution was added into the resulting solution for the subsequent fluorescence analysis at 409 nm with using a Fluoro Max-P spectrophotometer (FL1008M018, Cary, USA) after excitation at 315 nm.

2.3. Organic contaminants degradation

The degradation procedures of organic contaminants, including *p*-chloronitrobenzene, alachlor, chloramphenicol and rhodamine B, were almost the same as the above conversion process of H_2O_2 with just adding different organic contaminants of 10 mg/L in concentration. The concentrations of *p*-chloronitrobenzene, alachlor and chloramphenicol were determined by a high performance liquid chromatograph (HPLC, Shimadzu, SB-C18) equipped with a UV-vis detector. The mobile phases for the analyses of *p*-chloronitrobenzene, alachlor, and chloramphenicol were acetonitrile/water (30/70, v:v), acetonitrile/water (30/70, v:v), and methanol/water (55:45, v:v), respectively [4]. The detection wavelengths of *p*-chloronitrobenzene, alachlor and chloramphenicol were 260 nm, 225 nm, and 275 nm, respectively. All the flow rates were 1.0 mL/min. The concentration of rhodamine B was analyzed with a UV-vis spectrophotometer (UV-2550, Shimadzu, Japan). Column experiments were also conducted to check the feasibility of H_2O_2 and AA injection for in situ chemical oxidation of contaminated groundwater. The parameters of column apparatus were summarized in SM Table S3.

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