

In-situ spectroscopic investigation on arsenic sequestration mechanism by nanoscale zerovalent iron in the liquid-phase flow cell

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Abstract

While high efficiency of nanoscale zerovalent iron (nZVI) removing arsenic has been often reported, the chemical transformations of As(III) enabled by nZVI during the sequestration process are not well understood. It is important to investigate how arsenic complex interaction with redox in groundwater environment of Taiwan, which is a commonly occurring toxic substance in groundwater. Synchrotron-based X-ray absorption spectroscopic analysis was applied in situ using a reaction cell containing nZVI and aqueous As(III) solutions. This analysis enabled us to examine the dynamic changes in the arsenic chemical state over time in the reaction medium and to investigate in detail the bonding environments of arsenic at and within the nanoparticles under varying conditions. Results from this present study have direct implications for the potential use of nZVI for in situ groundwater remediation.

Keywords - nZVI; *in-situ* spectroscopic; flow cell.

Introduction

High levels of arsenic in groundwater influence millions of human health around the world. Nanoscale zero-valent iron (nZVI) is widely recognized as a material with a high potential agent for environmental friendly treatment of groundwater. Recent studies demonstrate that nZVI can retain a high level of As(III) and As(V) under laboratory and field conditions within a short time period. Identifying the nature of the embedded arsenic requires techniques that are capable of probing the core or bulk of the nanoparticles and generating data related to the local bonding environment of arsenic. To achieve this goal, synchrotron-based X-ray absorption spectroscopic (XAS) analysis was applied in-situ using a reaction cell containing nZVI and aqueous arsenic solutions. The penetrative nature of X-rays enables collection of information in-situ at liquid cell, while the chemical, local and electronic sensitivities of XAS allow the determination of the geometrical features of the metal complexes existing in solution, together with electronic information such as the oxidation state. It enabled us to examine the dynamic changes in the arsenic chemical state over time in the reaction medium and to investigate in detail the bonding environments of arsenic at and within the nanoparticles under varying conditions. In this study, we focused on identification and chemical description of mechanisms governing removal of arsenite and arsenate by nZVI particles in aqueous solution. Additionally, arsenic and iron species qualitative analysis was also investigated to clarify the role of nZVI during the reaction. Results from this present study have direct implications for the potential use of nZVI for in-situ groundwater remediation.

Experiments

nZVI particles were synthesized from ferric chloride using sodium borohydride reduction. As(III) and As(V) stock solutions were prepared from NaAsO_2 and $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$, respectively. Overall in-situ X-ray absorption measurements were performed at beamline TLS 07A1 and the rapid redox process was measured at TPS 44A of the National Synchrotron Radiation Research

Center (NSRRC). Scans were performed at room temperature in fluorescence mode at the arsenic K-edge and iron K-edge. Batch experiments of As(III) removal were performed in 100 mL serum bottles containing 50 mL of As(III) solution at 100 mg/L and connected with a micro pump with 0.05 ml/min flow rate.

Results

nZVI has high efficiency for removal arsenic in aqueous solution, over 99% of the As(III) in the solution was captured by 2 g/L of nZVI in 5 min. Dynamic transformation of arsenic in the presence of nZVI was directly monitored by collecting time-dependent XANES spectra of the mixture in the reaction cell. The overall reaction analysed result was shown in figure 1a. Quick XAS was used as an in situ technique to directly observed the initial change of arsenic state, the results were shown in figure 1b.

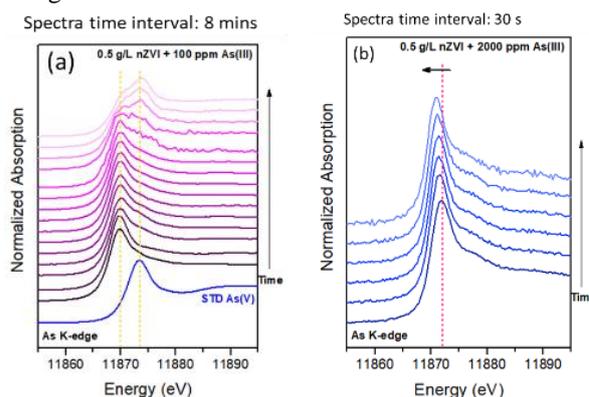


Figure 1. Time-dependent XANES data of As K-edge of (a) 2 hours and (b) initial 3 minutes reaction.

References

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