

Conceptual and numerical model of uranium(VI) reductive immobilization in fractured subsurface sediments

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Abstract

A conceptual model and numerical simulations of bacterial U(VI) reduction in fractured subsurface sediments were developed to assess the potential feasibility of biomineralization at the fracture/matrix interface as a mechanism for immobilization of uranium in structured subsurface media. The model envisions flow of anaerobic groundwater, with or without acetate as an electron donor for stimulation of U(VI) reduction by dissimilatory metal-reducing bacteria (DMRB), within mobile macropores along a one-dimensional flow path. As the groundwater moves along the flow path, U(VI) trapped in the immobile mesopore and micropore domains (the sediment matrix) becomes desorbed and transferred to the mobile macropores (fractures) via a first-order exchange mechanism. By allowing bacterial U(VI) reduction to occur in the mesopore domain (assumed to account for 12% of total sediment pore volume) according to experimentally-determined kinetic parameters and an assumed DMRB abundance of 10^7 cells per cm^3 bulk sediment (equivalent to 4 mg of cells per dm^3 bulk sediment), the concentration of U(VI) in the macropore domain was reduced ca. 10-fold compared to that predicted in the absence of mesopore DMRB activity after a 6-month simulation period. The results suggest that input of soluble electron donors over a period of years could lead to a major redistribution of uranium in fractured subsurface sediments, converting potentially mobile sorbed U(VI) to an insoluble reduced phase (i.e. uraninite) in the mesopore domain that is expected to be permanently immobile under sustained anaerobic conditions.

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1. Introduction

Uranium is an important radionuclide contaminant in soils and subsurface sediments at nuclear weapons manufacturing and uranium mining sites in the US and abroad (Landa and Gray, 1995; Bradley et al., 1996). The long half-life of ^{238}U (4.5×10^9 yr) and its decay to a suite of radioactive daughter products makes subsurface uranium contamination a significant environmental

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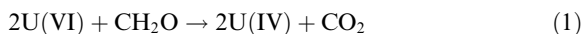
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concern. For example, the risk posed by groundwater uranium contamination in the vicinity of uranium mill tailings exceeds that posed by high level nuclear waste subsequent to the decay of relatively short-lived (e.g., <600 years) isotopes (Landa and Gray, 1995).

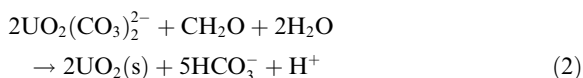
Although aqueous U(VI) species are subject to surface complexation by particles in soils and sediments (Ticknor, 1994), in particular Fe(III) oxide surfaces (Hsi and Langmuir, 1985; Kohler et al., 1992; Casas et al., 1994; Ticknor, 1994; Waite et al., 1994; Morrison et al., 1995; Arnold et al., 1998), aqueous complexation of U(VI) by carbonate ions decreases the tendency of U(VI) to bind to mineral surfaces (Waite et al., 1994) and thereby enhances subsurface uranium migration. Since many subsurface environments contain substantial amounts of dissolved inorganic carbon (DIC), uranium can be relatively mobile in groundwater, and U(VI) is classified as a high mobility contaminant in the subsurface at several DOE sites (DOE, 1991). As a result, there is substantial interest in development of technologies for retarding uranium migration in subsurface sediments (NationalResearchCouncil, 2000).

1.1. Uranium bioremediation via bacterial reductive immobilization

A promising approach for immobilization of uranium in the subsurface involves harnessing the activity of dissimilatory metal-reducing bacteria (DMRB) (Lovley et al., 1991; Gorby and Lovley, 1992; Lovley and Phillips, 1992; Lovley, 1995), which under anaerobic conditions catalyze enzymatic reduction of U(VI) to U(IV) via reactions such as



where CH₂O represents a generic unit of organic carbon. The significance of this reaction in terms of uranium mobility is that uranium in the +4 oxidation state tends to precipitate rapidly as the insoluble mineral uraninite, UO₂(s) (Langmuir, 1978). As a result, microbial uranium reduction can provide a mechanism for immobilization of uranium in subsurface environments via reactions such as



where UO₂(CO₃)₂²⁻ represents the dominant aqueous U(VI)–carbonate complex in DIC-bearing solutions at circumneutral pH (Langmuir, 1997). Anderson et al. (2003) recently provided an example of an in situ U(VI) remediation operation in unconfined (sandy, porous) aquifer sediments at a former uranium ore processing facility in Rifle, CO: addition of acetate to groundwater stimulated the activity of DMRB within the family *Geobacteraceae*, resulting in significant

declines in aqueous U(VI) concentration during the period of organic carbon addition. A prerequisite for this type of process to operate effectively is the existence of both anaerobic and nitrate-free conditions in the aquifer sediments, because net U(VI) reduction by DMRB occurs only in the absence of O₂ and NO₃⁻ (Finneran et al., 2002a; Holmes et al., 2002). Thus, unless a uranium-contaminated aquifer is *already* anaerobic and nitrate-free (as in the case of the Rifle, CO site), it would be necessary to induce anaerobic conditions and consume groundwater nitrate (through addition of organic carbon) in order to set the stage for effective U(VI) bioremediation. Recent studies suggest that poly lactate ester compounds (e.g. Hydrogen Release Compound (HRCTM)) can provide consistent release of electron equivalents to maintain anaerobic conditions and stimulate reductive biotransformations in contaminated aquifer sediments (Haas and Trego, 2001). For the purpose of the conceptual development and numerical simulations presented in this paper, we assume that the subsurface environment is already anaerobic, and that DMRB have the potential to carry out U(VI) reduction when provided with suitable organic substrates.

An advantage of the above bioremediation strategy is that the DMRB that reduce and precipitate uranium can also utilize Fe(III) oxides as electron acceptors for anaerobic respiration (Lovley, 1995). Many subsurface sediments contain substantial quantities of Fe(III) oxides. In such environments, addition of electron donors and nutrients would stimulate utilization of the large supply of endogenous electron acceptor provided by the Fe(III) oxides to generate and maintain DMRB biomass, which could then reduce and immobilize U(VI) moving through the treatment zone. This “in situ biogenic redox barrier” concept is analogous to processes involved in the formation of roll-front geological uranium ore deposits (Lovley and Anderson, 2000), in which U(VI) is reduced and precipitated along an advancing front localized at the interface between oxidized and reduced zones (Langmuir, 1997). Recent studies have demonstrated the feasibility of stimulating the growth and activity of DMRB for Fe(III)/U(VI) reduction in previously well-oxidized subsurface sediments (Abdelouas et al., 1998; Snoeyenbos-West et al., 2000; Holmes et al., 2002). The biogenic redox barrier concept may offer a far less costly and ultimately more effective approach to U(VI) remediation compared to protracted pump-and-treat operations for groundwater contaminant plumes which are dispersed over large areas and located 10's to 100's of meters below ground.

1.2. Potential complications in structured subsurface media

Although the U(VI) reductive immobilization strategy described above is conceptually sound, complica-

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