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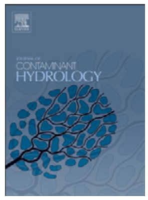

Reactive transport of uranium with bacteria in fractured rock: Model development and sensitivity analysis

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ABSTRACT

A numerical model for the reactive transport of uranium and bacteria in fractured rock was newly developed. The conceptual model consists of four phases (fracture, fracture surface, matrix pore, and matrix solid) and eight constituents (solutes in the fracture, on the fracture surface, on mobile bacteria, on immobile bacteria, in the rock matrix pores and on the rock matrix solids, and bacteria in the fracture and on the fracture surface). In addition to the kinetic sorption/desorption of uranium and bacteria, uranium reduction reaction accompanying with bacteria growth was considered in the reactive transport. The non-linear reactive transport equations were numerically solved using the symmetric sequential iterative scheme of the operator-splitting method. The transport and kinetic reaction modules in the developed model were separately verified, and the results were reasonably acceptable. From the sensitivity analysis, the uranium transport was generally more sensitive to the sorption rate rather than desorption rate of U(VI). Considering a uranium reduction reaction, bacteria could considerably retard the uranium transport no matter the uranium sorption/desorption rates. As the affinity of U(VI) onto the bacteria becomes higher than that onto a rock fracture surface, a biofilm effect, rather than a colloidal effect, of the bacteria becomes more influential on the uranium transport.

1. Introduction

Once high-level radioactive waste (HLW) is disposed of and closed in a bedrock, accessibility to the HLW becomes very limited. Therefore, a disposal system should be carefully prepared, and the future states of the system should be reasonably predicted considering all events possibly occurring in the bedrock for a long period of time. One possible phenomenon is the groundwater intrusion into the HLWs followed by the leakage of radionuclides into the groundwater flow. In a deep geological disposal system, the radionuclides generally leaked from the engineering barrier (usually compacted bentonite) can be transported and will eventually reach the biosphere through the rock fracture network, which is called a natural barrier, and/or major water conducting feature (MWCF) (Hodgkinson et al., 2009).

The transport of radionuclides through fractures can be retarded not only by diffusion into the rock matrix (Holtta et al., 2008; Neretnieks, 1980) but also by an interaction with the fracture surface, fracture filling materials, colloids, organics, bacteria, etc. (Baik et al., 2008). For many radionuclides, sorption is an important phenomenon, as their migration rates in groundwater are reduced in both an engineered barrier and a fractured rock matrix (Alexander and McKinley, 2007). The sorption of radionuclides is strongly dependent on the chemistry of the surrounding groundwater, such as the pH, Eh, and ionic strength by changing their valence states (e.g., Katsoyiannis et al., 2006). In addition, it is also known that some bacteria can change the mobility and speciation of a radionuclide in groundwater. Biological immobilization mechanisms of radionuclides include precipitation and transformation into less soluble forms (Gadd, 1996). On the other hand, bacteria can also play a role of a sorbent for radionuclides (Xie

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et al., 2008). Since bacteria can not only be mobile as a colloid but can also be immobile as a biofilm in the rock fracture, bacteria as the sorbent of radionuclides in groundwater can have both positive and negative effects on radionuclide migration. To improve the roles of natural barriers for retarding a radionuclide transport in a deep geological disposal system, the retardation factors mentioned above must be preferentially characterized and research strategically encouraged.

Research on the characterization of radionuclide transport (including retardation in the fractured rock) has suffered from limited accessibility and long-term of concerns since a repository is usually located in the ground several hundreds of meters deep, and the half-lives of most radionuclides are longer than thousands or even millions of years (Neretnieks, 1980). Owing to the restricted conditions of a deep geological disposal system, numerical modeling approaches have been relatively spotlighted in radionuclide migration. After an analytical solution of the contaminant transport in a single fractured porous medium was reported by Tang et al. (1981), Park et al. (2001) identified the effect of the limiting zone for the matrix diffusion by using full analytic solutions. For the numerical modeling approaches, Buckley and Loyalka (1993) developed an explicit finite difference numerical technique for a solution of radionuclide transport by advection–diffusion in a single fracture and diffusion in the surrounding rock-matrix. In the following year, Buckley and Loyalka (1994) studied the effect of lateral diffusion in the solute transport in a rock fracture. However, their studies were limited to the equilibrium condition as they considered only a linear sorption isotherm using a retardation factor without any kinetic reaction terms in the model. Recently, many researchers have tried to add the bio-geo-chemical effects on the radionuclide transports into existing transport models, which are usually called reactive transport models. Reactive transport models were finely summarized and reviewed by MacQuarrie and Mayer (2005).

In this context, an orthogonal one-dimensional numerical model for the reactive transport of radionuclides in a single fractured rock was developed in this study using an operator splitting method (OSM). The OSM is a numerical technique to efficiently compute non-linear advection–dispersion equations, including reactive kinetics. Several operator splitting techniques for a reactive transport were reviewed by Carrayrou et al. (2004). From a comparison of these techniques, Carrayrou et al. (2004) concluded that the Strang-splitting sequential non-iterative scheme, which was employed in this study, was rather more accurate than the other iterative and non-iterative schemes.

In this study, a numerical model for the reactive transport of bacteria and radionuclides in fractured rock was newly developed using the OSM to assess the effects of both the sorption/desorption of bacteria and radionuclides and a bacterial reaction on the transport of radionuclides. For the model verification, the transport module of the newly developed model was first verified by comparing it with an analytical solution under limited conditions. The reaction module was then verified by comparing it with experimental results from alluvial gravel aquifer media by Pang et al. (2005). Finally, the effects of bacteria sorption and reaction on the transport of radionuclide were investigated by varying the kinetic sorption/desorption rate coefficients of the radionuclide.

2. Model development

2.1. Conceptual model

The conceptual model of the reactive transports of bacteria and solutes (e.g., nutrients, radionuclides, etc.) in the fractured rock is depicted in Fig. 1. The system includes four phases, fracture ('*F*'), fracture surface ('*S*'), matrix pore ('*P*'), and matrix solid ('*M*'), and eight constituents such as solute in the fracture ('*Fc*'), solute on the fracture surface ('*Sc*'), bacteria in the fracture ('*Fb*'), bacteria on the fracture surface ('*Sb*'), solute on the mobile bacteria surface in the fracture ('*FBc*'), solute on the immobile bacteria surface on the fracture surface ('*SBc*'), solute in the matrix pores ('*Pc*'), and solute on the matrix solids ('*Mc*'). The definitions of the abbreviations used in this study are listed in Table 1.

The transport of bacteria was regarded as a colloid type considering the size of the bacteria (several μm; Prescott et al., 2002). Some properties of colloid transport are different from those of solute transport. First, the pore water velocity of the colloid is affected by various forces, such as the hydrodynamic lateral force, colloidal forces (repulsive double-layer force and attractive van der Waals force), gravitational force, and so on (Li et al., 2004). Secondly, bacteria have difficulty diffusing into the porous rock matrix since the bacteria size is bigger than a common rock matrix pore size. For instance, Inigo et al. (2000) reported that even weathered rock has less than a 10 nm pore size. Thus, it was assumed that bacteria can transport only in a fracture, and solute in both a fracture and matrix. Finally, bacteria can play a role of adsorbent for the solute, and thus their colloidal characteristics can facilitate the solute transport (e.g., Pang et al., 2005), and the immobilized bacteria (biofilm) can enhance the retardation of the solute transport (e.g., Anderson et al., 2007). Accordingly, it was also assumed that bacteria are not only transporting materials but also a medium that can accommodate solutes in a fracture and on a fracture surface.

In addition to the bacteria's colloidal properties, the bacterial metabolism also influences the concentration and/or mobility of solutes in the groundwater. In terms of radioactive waste disposal, bacterial reduction of radionuclides in deep groundwater is one of the significant mechanisms for safety. For instance, Wilkins et al. (2006) reported that a bacterial reduction can retard the radionuclide transport by an increase in the sorption rate and precipitation by mineralization.

The major processes considered in the model development for the reactive transports of bacteria and solutes in the fractured rock are listed in Table 2. Our main concern in the model development is the reactive transport of radionuclides in fractured rock. For simplicity, uranium was considered a single radionuclide in this study. Since it is known that the iron reducing bacteria (IRB) reduces the uranyl ions (U(VI); UO_2^{2+}) (Lovley et al., 1991), IRB was considered a single bacteria species in this study. For the bacterial metabolism or redox reaction, lactate ($C_3H_5O_3^-$) was assumed as a single carbon source. The redox reactions considered in this study are as follows:

$$C_3H_5O_3^- + 2UO_2^{2+} + 2H_2O \Leftrightarrow C_2H_3O_2^- + 2UO_2 + \chi\left(HCO_3^- + 5H^+\right) + \text{Bacteria growth}.$$