Journal of Hydrology 536 (2016) 133–146

Contents lists available at ScienceDirect

Journal of Hydrology

journal homepage: www.elsevier.com/locate/jhydrol

Solute transport through fractured rock: Radial diffusion into the rock matrix with several geological layers for an arbitrary length decay chain

Batoul Mahmoudzadeh *, Longcheng Liu, Luis Moreno, Ivars Neretnieks

Department of Chemical Engineering and Technology, Royal Institute of Technology, S-100 44 Stockholm, Sweden

A R T I C L E   I N F O

Article history:
Received 16 October 2015
Received in revised form 7 January 2016
Accepted 22 February 2016
Available online 3 March 2016
This manuscript was handled by Peter K. Kitanidis, Editor-in-Chief, with the assistance of Jian Luo, Associate Editor

Keywords:
Solute transport model
Semi-analytical solution
Radial matrix diffusion
Geological rock layers
Fractured rock
Radionuclide decay chain

S U M M A R Y

The paper presents a model development to derive a semi-analytical solution to describe reactive solute transport through a single channel in a fracture with cylindrical geometry. The model accounts for advection through the channel, radial diffusion into the adjacent heterogeneous rock matrix comprising different geological layers, adsorption on both the channel surface, and the geological layers of the rock matrix and radioactive decay chain. Not only an arbitrary-length decay chain, but also as many number of the rock matrix layers with different properties as observed in the field can be handled. The solution, which is analytical in the Laplace domain, is transformed back to the time domain numerically e.g. by use of de Hoog algorithm. The solution is verified against experimental data and analytical solutions of limiting cases of solute transport through porous media. More importantly, the relative importance and contribution of different processes on solute transport retardation in fractured rocks are investigated by simulating several cases of varying complexity. The simulation results are compared with those obtained from rectangular model with linear matrix diffusion. It is found that the impact of channel geometry on breakthrough curves increases markedly as the transport distance along the flow channel and away into the rock matrix increase. The effect of geometry is more pronounced for transport of a decay chain when the rock matrix consists of a porous altered layer.

1. Introduction

In many countries, high level radioactive wastes will be deposited in deep repositories in crystalline rocks because of their stability and low permeability (Li and Chiou, 1993; Gylling et al., 1998). It is then very important to understand the main mechanisms and processes, which govern transport of radionuclides in fractured rocks, and consequently to develop methods and models to be used to efficiently simulate these phenomena for the safety and performance assessment of deep repositories for spent nuclear fuel and other radioactive wastes.

It has been observed in many large field experiments, as well as in drifts and tunnels on fractured rocks, that groundwater flows mainly in open parts of fractures, called channels. There is negligible flow through the rock matrix (Birgersson et al., 1992, 1993; Abelin et al., 1991, 1994; Tsang and Neretnieks, 1998; Stober and Bucher, 2006). Three-dimensional networks of channels connect to form pathways for solute transport from the repository to the biosphere. The low permeability porous rock matrix plays a dominant role on retarding solute transport. The solutes, which are passed by the flowing water in the channel, can also diffuse in and out of the immobile groundwater in the rock matrix, through "matrix diffusion" process that makes a much larger water volume accessible for the nuclides to reside in (Neretnieks, 1980; Sudicky and Frind, 1984). Sorbing nuclides find more mineral surfaces in the matrix to attach to. Many nuclides can be retarded sufficiently to decay to insignificant concentrations.

Over the recent years, many different models have been studied in order to account for two processes of rock matrix diffusion and advection in the fracture. The comparison of different modeling approaches for this practical matter has been done by Selroos et al. (2002). Discrete fracture network, DFN, models are based on the assumption that some fractures are fully open over their entire size, and they form a conductive network by intersecting each other (Hartley and Roberts, 2012; Dershowitz et al., 1999). However, the premise that rock fractures are either fully open or closed is not supported by observations. Numerous field observations suggest that fracture surfaces are uneven, and groundwater flows only in a narrow part of the whole fracture. The narrow conductive parts will form "flow channels" (Tsang and Neretnieks, 1998). These flow channels form a network for water flow and solute transport (Neretnieks, 1993). The channel network model, CNM, has been used for solute transport simulations in fractured

---

* Corresponding author. Tel.: +46 8 790 6414.
E-mail address: mbatoul@kth.se (B. Mahmoudzadeh).

http://dx.doi.org/10.1016/j.jhydrol.2016.02.046
0022-1694/© 2016 Elsevier B.V. All rights reserved.

rocks (Gylling et al., 1999; Gylling, 1997; Moreno and Neretnieks, 1993a; Moreno et al., 1997). In such heterogeneous porous medium where the fluid velocity field varies, there are some regions in the fracture plane, in which advective transport is essentially negligible. The low velocity may be because the fracture has a small aperture there or because it contains more particle fragments forming a porous medium with low permeability. We call such regions with much less flow, stagnant water zones. The solutes carried by the flowing water in the channel diffuse into the stagnant water zones in the fracture plane and from there further diffuse into the porous rock matrix.

The stagnant water zone in the fracture plane has been ignored in many models including channel network model (Poteri and Laitinen, 1999; Hartley, 1998; Dershowitz et al., 1998), however, this effect has been recently studied for solute transport in fractured rocks (Neretnieks, 2006; Mahmoudzadeh et al., 2013). It was found that the diffusion into the stagnant water zone can significantly retard solute transport, when the zone is wide. This gives the solute access to a larger surface over which to diffuse into the rock matrix increasing retardation. The diffusion into the stagnant water zone in the fracture plane can be accounted for in the channel network model, but not in fracture network models because of its main assumption of fully open or closed fractures.

In most of the models for flow and solute transport through fractured rocks, it is assumed that the rock matrix is a homogenous medium with either infinite or finite thickness (Moreno and Neretnieks, 1993b; Moreno et al., 1997; Barten, 1996; Neretnieks, 1980; Grisak and Pickens, 1980; Tang et al., 1981; Sudicky and Frind, 1982; Davis and Johnston, 1984; van Genuchten, 1985; Zimmerman and Bodvarsson, 1995; Sharifi Haddad et al., 2013). However, the structure of the rock matrix, in reality, is more complex as the rock may be altered near the fracture (Löfgren and Sidborn, 2010; Cvetkovic, 2010; Piqué et al., 2010). The altered zones of the rock are significantly different from the intact wall rock in retardation properties (Selnert et al., 2009). It has been recently shown that the altered rock will intensely influence the access of solutes to the rock matrix and as a consequence may cause more retardation of the solute transport in fractured media (Moridis, 2002; Mahmoudzadeh et al., 2013, 2014). For this reason, some work has been done to account for matrix heterogeneities by devising effective values of diffusion and sorption parameters based on stochastic information (Dai et al., 2007, 2009, 2012; Deng et al., 2010). In the present model, we specifically account for the effect of different geological layers with different thickness, sorption and diffusion properties.

The diffusion in the rock matrix is assumed, in most models, to be one dimensional and orthogonal to the fracture surface where the fracture is conceptualized as a rectangular channel. However, since the flow channels in fractures are usually narrow, the matrix diffusion is more realistic to be modeled as radial diffusion from a cylindrical channel when the diffusion penetration depth grows larger than the width of the channel. On the other hand, although fully cylindrical channels are seldom found in nature, there are some cases where cylinder-like channels may be more adequately used for modeling fluid flow and solute transport. For instance, circular layered rock formations might be built up during the completion (rock cracking, mud contamination etc.) of a horizontal well used to inject the waste. Such cylinder-like channels can also be found at fracture intersections in fractured rocks. Around these channels it may be expected that over time, chemical changes will lead to development of circular matrix layers when the diffusion/reaction penetration depth becomes larger than the channels width. This can also be expected to occur around flat narrow channels over long times.

As a consequences, several numerical, analytical and experimental studies have been conducted to describe the radial diffusion of solutes into a soil or rock matrix from a cylindrical macropore or fracture for different boundary conditions (van Genuchten et al., 1984; Rasmuson and Neretnieks, 1986; Carrera et al., 1998; Rahman et al., 2004; Cihan and Tyner, 2011; Chopra et al., 2015). The radial diffusion has been shown to be more effective than linear diffusion in retarding solute transport through a channel with the same flow-wetted surface (Rasmuson and Neretnieks, 1986).

In the present paper we develop a model for reactive solute transport through a circular conductive fracture with the rock matrix composed of several geological layers with different properties, as schematically shown in Fig. 1. The definition and properties of these geological materials have been studied previously (Mahmoudzadeh et al., 2013). Our aim, in the present study, is to explore the relative impact and contribution of the altered rock matrix and the decay chain on transport of radionuclides through a cylindrical channel with radial matrix diffusion. However, the model ignored the presence of stagnant water zone in the fracture plane at present. The results are compared with those obtained from the "rectangular" model (Mahmoudzadeh et al., 2014) where nuclides are flowing in a channel in fracture with linear diffusion in the rock matrix layers. The analytical solution, in the Laplace domain, presented in this work is applicable for the cases involving an arbitrary-length decay chain, where the rock matrix consists of any number of different geological layers. These analytical solutions can be used for prediction of nuclide transport through complex networks of channels, and serve as verification tools for the numerical codes.

As a matter of fact, we conceptualize the flow and transport in a fractured rock mass to take place in a three-dimensional network of channels; the solute from one channel enters one or more channels at an intersection. This channel network model allows us not to account explicitly for "hydrodynamic dispersion" as the conventional advection–dispersion model does. Instead, as discussed by Neretnieks (1983) hydrodynamic dispersion may be more adequately considered to be the result of different residence times in different pathways. This eliminates the problem of the inconsistency of the advection–dispersion equation in which the observed dispersion coefficient increases with distance in field experiments, whereas the advection–dispersion equation requires a constant value of the "dispersion coefficient". In addition, we have also found in simulations with the channel network model that the large scale dispersion in a network is dominated by velocity dispersion and matrix diffusion; and that dispersion in individual channels has a small impact (Moreno and Neretnieks, 1991, 1993b; Gylling, 1997). For this reason and also because hydrodynamic dispersion in channels in fractures is not well studied (Molz, 2015; Tsang et al., 2015) we do not include this mechanism in the model for solute transport through individual channels.

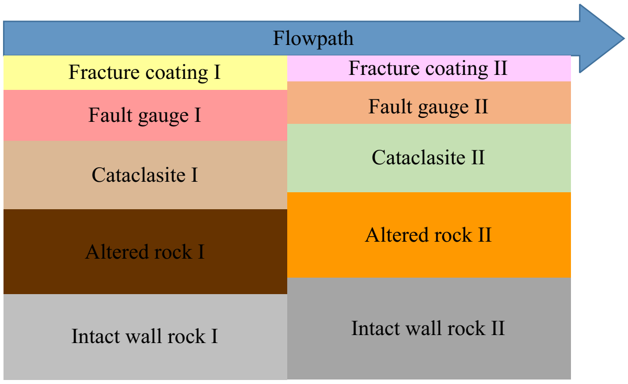

Fig. 1. Schematic conceptualization of two layered rock volumes.