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Radionuclide migration through fractured rock for arbitrary-length decay chain: Analytical solution and global sensitivity analysis

Pirouz Shahkarami *, Longcheng Liu, Luis Moreno, Ivars Neretnieks

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

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SUMMARY

This study presents an analytical approach to simulate nuclide migration through a channel in a fracture accounting for an arbitrary-length decay chain. The nuclides are retarded as they diffuse in the porous rock matrix and stagnant zones in the fracture. The Laplace transform and similarity transform techniques are applied to solve the model. The analytical solution to the nuclide concentrations at the fracture outlet is governed by nine parameters representing different mechanisms acting on nuclide transport through a fracture, including diffusion into the rock matrices, diffusion into the stagnant water zone, chain decay and hydrodynamic dispersion. Furthermore, to assess how sensitive the results are to parameter uncertainties, the Sobol method is applied in variance-based global sensitivity analyses of the model output. The Sobol indices show how uncertainty in the model output is apportioned to the uncertainty in the model input. This method takes into account both direct effects and interaction effects between input parameters. The simulation results suggest that in the case of pulse injections, ignoring the effect of a stagnant water zone can lead to significant errors in the time of first arrival and the peak value of the nuclides. Likewise, neglecting the parent and modeling its daughter as a single stable species can result in a significant overestimation of the peak value of the daughter nuclide. It is also found that as the dispersion increases, the early arrival time and the peak time of the daughter decrease while the peak value increases. More importantly, the global sensitivity analysis reveals that for time periods greater than a few thousand years, the uncertainty of the model output is more sensitive to the values of the individual parameters than to the interaction between them. Moreover, if one tries to evaluate the true values of the input parameters at the same cost and effort, the determination of priorities should follow a certain sequence.

1. Introduction

In response to a keen interest in the performance and safety assessment of repositories for spent nuclear fuel and other radioactive waste, recent decades have seen rapid development of new methods used to simulate radionuclide transport in fractured rocks. Analytical models have gained much attention, as they can provide valuable insights into fundamental physical phenomena and are computationally efficient over a wide range of time scales required for performance assessments of radioactive waste repositories. The analytical models, although limited to simplified geometries, are also useful for validating more complex numerical codes.

Should nuclides leak from the repository, the question of prime interest is at what rate the nuclides might reach the biosphere. In fractured crystalline rocks, the nuclides are carried by seeping water through interconnected networks of fractures (Moreno and Neretnieks, 1993a; Gylling et al., 1998) and may be subject to different retarding mechanisms (Bodin et al., 2003a; Moreno et al., 1996; Neretnieks, 2005). Advection and hydrodynamic dispersion are the principal mechanisms governing the transport process along the fractures. Molecular diffusion from the flowing channels into the porous rock matrix, in which nuclides can sorb, is by far the most important retardation mechanism, (Neretnieks, 1980). The larger the contact surface between the contaminated water, the larger is the retardation. The nuclides may also diffuse into stagnant water zones in the fracture and from there into the matrix. This can considerably add to the retardation and allow them to decay. Bodin et al. (2003b) give an overview of models that address the transport in fractured rock.

For a simple case of a fracture surrounded by a porous homogeneous rock matrix, Neretnieks (1980) obtained an analytical

* Corresponding author at: Department of Chemical Engineering and Technology, Division of Chemical Engineering, KTH Royal Institute of Technology, Teknikringen 42, SE-100 44 Stockholm, Sweden. Tel.: +46 8 790 6416, +46 8 790 63 46; fax: +46 8 10 52 28.
E-mail address: pirouzs@kth.se (P. Shahkarami).
URL: http://www.kth.se (P. Shahkarami).

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Nomenclature

b_f	half aperture of the flowing channel (L)
b_s	half aperture of the stagnant water zone (L)
C_0	concentration at the inlet of the flowing channel (ML^{-3})
C_f	concentration in the flowing channel (ML^{-3})
C_{pf}	pore water concentration in the rock matrix adjacent to the flowing channel (ML^{-3})
C_{ps}	pore water concentration in the rock matrix adjacent to the stagnant water zone (ML^{-3})
C_s	concentration in the stagnant water zone (ML^{-3})
D_{af}	apparent diffusivity in the rock matrix adjacent to the flowing channel ($L^2\,T^{-1}$)
D_{as}	apparent diffusivity in the rock matrix adjacent to the stagnant water zone ($L^2\,T^{-1}$)
D_{ef}	effective diffusivity in the rock matrix adjacent to the flowing channel ($L^2\,T^{-1}$)
D_{es}	effective diffusivity in the rock matrix adjacent to the stagnant water zone ($L^2\,T^{-1}$)
D_s	diffusivity in the water in the stagnant water zone ($L^2\,T^{-1}$)
F_f	ratio of the flow-wetted surface of the flowing channel to the volumetric water flow rate (TL^{-1})
F_s	ratio of the stagnant-water-wetted surface to the diffusion conductance of the stagnant water zone (TL^{-1})
MPG_f	material property group of the rock matrix adjacent to the flowing channel ($LT^{-1/2}$)
MPG_s	material property group of the rock matrix adjacent to the stagnant water zone ($LT^{-1/2}$)
n	number of nuclides in a decay chain (–)
N	ratio between the diffusion rate into the stagnant water zone and the mass flow rate through the channel (–)
Pe	Peclet number (–)
S	first-order sensitivity index (–)
S_T	total sensitivity index (–)
R_f	surface retardation coefficient in the flowing channel (–)
R_{pf}	retardation coefficient of the rock matrix adjacent to the flowing channel (–)
R_{ps}	retardation coefficient of the rock matrix adjacent to the stagnant water zone (–)
R_s	surface retardation coefficient in the stagnant water zone (–)
s	Laplace transform variable (T^{-1})
t	time (T)
u	groundwater velocity (LT^{-1})
W_f	half width of the flowing channel (L)
W_s	half width of the stagnant water zone (L)
x	distance along the flow direction (L)
y	distance into the stagnant water zone (L)
z	distance into the rock matrix adjacent to the flowing channel
z_s	distance into the rock matrix adjacent to the stagnant water zone
δ_f	thickness of the rock matrix adjacent to the flowing channel (L)
δ_s	thickness of the rock matrix adjacent to the stagnant water zone (L)
ε_{pf}	porosity of the rock matrix adjacent to the flowing channel (–)
ε_{ps}	porosity of the rock matrix adjacent to the stagnant water zone (–)
λ	decay constant (T^{-1})
τ_f	characteristic time of advection (–)
τ_{Df}	characteristic time of diffusion through the rock matrix adjacent to the flowing channel (–)
τ_{Ds}	characteristic time of diffusion through the rock matrix adjacent to the stagnant water zone (–)
τ_s	characteristic time of diffusion through the stagnant water zone (–)

Subscripts

f	refers to the flowing channel
s	refers to the stagnant water zone
pf	refers to the rock matrix adjacent to the flowing channel
ps	refers to the rock matrix adjacent to the stagnant water zone

solution for transport along a discrete fracture in a porous rock matrix. It accounts for advection, diffusion into the rock matrix, adsorption on the fracture surface and within the matrix, as well as radioactive decay of a single nuclide. Tang et al. (1981) extended the model to include dispersion. Sudicky and Frind (1984) and Cormenzana (2000) presented an analytical solution for a two-member radionuclide decay chain transport through a fracture. Although the solution is restricted to a two-member decay chain, it represents a more realistic approach, and the simulations they made suggest that neglecting the parent-daughter interactions can lead to a significant error in the estimated time of arrival. In the late 1980s, Hodgkinson and Maul (1988) applied a semi-analytical approach to solve radionuclide transport equations for an arbitrary-length decay chain in fractured rocks. In this study, the governing equations are first transformed in the Laplace domain and analytically solved, then numerically inverted back to the time domain. Using the same approach, Neretnieks (2005) recently considered the influence of a stagnant water zone adjacent to the flowing channel. In this model, the nuclides may first diffuse into a stagnant water zone in the fracture plane and then further into the rock matrix adjacent to it. It was found that this process may considerably contribute to the retardation of nuclides because more rock matrix can be accessed from the stagnant zone. Based upon this idea, Mahmoudzadeh et al. (2013) further developed a more realistic model to describe the nuclide transport and retention in fractured rocks. It accounts for the presence of several geological layers adjacent to the fracture surface both in the flowing channel and in the stagnant part of the fracture. The results show that the transport properties within the fracture and other subsystems can be fully described by a few characteristic parameters. More importantly, the findings demonstrate that under certain conditions, it is necessary to account for both the stagnant water zone and the rock matrix comprising at least two geological layers in safety and performance assessments of repositories for spent nuclear fuel.

These studies help to give a better insight into the influences of the individual mechanisms acting on nuclide migration. However, to the best of our knowledge, none of the studies discussed so far have attempted to consider all of the important mechanisms together to present an analytical solution for nuclide transport in fractured rocks. The main contribution of the present work is to extend the model developed by Mahmoudzadeh et al. (2013) to include an arbitrary-length decay chain in the presence of a stagnant water zone. The model takes into account not only advection and hydrodynamic dispersion along the fracture but also linear equilibrium sorption, diffusion into the rock matrix from the flowing channel, diffusion into the stagnant water zone from which also diffusion into the rock matrix occurs, and radioactive chain