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Influence of fracture networks on radionuclide transport from solidified waste forms

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HIGHLIGHTS

- Magnitude of peak radionuclide fluxes is less sensitive to the fracture network geometry.
- Time of peak radionuclide fluxes is sensitive to the fracture networks.
- Uniform flow model mimics a limiting case of a porous medium with large number of fine fractures.
- Effect of fracture width on radionuclide flux depends on the ratio of fracture to matrix conductivity.
- Effect of increased dispersivity in fractured media does not always result in a lower peak flux for specific fracture networks due to higher concentrations adjacent to the fracture plane.

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ABSTRACT

Analysis of the effect of fractures in porous media on fluid flow and mass transport is of great interest in many fields including geotechnical, petroleum, hydrogeology and waste management. This paper presents sensitivity analyses examining the effect of various hypothetical fracture networks on the performance of a planned near surface disposal facility in terms of radionuclide transport behaviour. As it is impossible to predict the initiation and evolution of fracture networks and their characteristics in concrete structures over time scales of interest, several fracture networks have been postulated to test the sensitivity of radionuclide release from a disposal facility. Fluid flow through concrete matrix and fracture networks are modelled via Darcy's law. A single species radionuclide transport equation is employed for both matrix and fracture networks, which include the processes advection, diffusion, dispersion, sorption/desorption and radioactive decay. The sensitivity study evaluates variations in fracture network configuration and fracture width together with different sorption/desorption characteristics of radionuclides in a cement matrix, radioactive decay constants and matrix dispersivity. The effect of the fractures is illustrated via radionuclide breakthrough curves, magnitude and time of peak mass flux, cumulative mass flux and concentration profiles. For the investigated system, radionuclide properties and the imposed water flow boundary conditions, results demonstrate that: (i) magnitude of peak radionuclide fluxes is less sensitive to the fracture network geometry, (ii) timing of the peak radionuclide fluxes is possibly sensitive to the fracture networks, (iii) a uniform flow model represents a limiting case of a porous medium with large number of fine fractures, (iv) the effect of fracture width on the radionuclide flux depends on the ratio of fracture to matrix conductivity and is less sensitive if the ratio is large and the width is higher than a certain critical size, and (v) increased dispersivity in fractured media influences the transport behaviour differently compared to non-fractured media; in particular, the behaviour depends on the nature of fracture network.

1. Introduction

Micro and macro cracks or fractures develop in any concrete structures as a result of interactive physical, chemical and mechanical processes (Neville, 1995; Page and Page, 2007). Structural fractures are commonly the result of flexural, tensile, compressive or bending loads. Typical non-structural fractures are due to

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S	storage coefficient (Pa^{-1})
p	total pressure (Pa)
t	time (s)
ρ_l	density of water (kg/m^3)
K_s	saturated hydraulic conductivity of concrete matrix (m/s)
g	acceleration due to gravity (m/s^2)
d_f	fracture width (m)
∇_T	tangential gradient (m^{-1})
K_f	hydraulic conductivity of a single fracture (m/s)
μ	dynamic viscosity of water (kg/m/s)
c	radionuclide concentration (Bq/m^3)
ρ_b	bulk density of concrete (kg/m^3)
η	porosity of concrete matrix
η_f	porosity of a discrete fracture
K_d	distribution coefficient (l/kg)
R	retardation factor
$\mathbf{D}_L$	hydrodynamic dispersion tensor (m^2/s)
D_0	free solution diffusion coefficient (m^2/s)
$\mathbf{u}$	Darcy's velocity vector (m/s)
$\mathbf{u}_T$	tangential velocity vector (m/s)
λ	decay constant (s^{-1})

early age thermal contraction, drying shrinkage, rebar corrosion, sulphate attack, decalcification, or creep (Page and Page, 2007). Voids or discrete pathways may also exist in concrete structures; either they are initially present as a design component to achieve lighter concrete structures (e.g. in cast-in-place and pre-cast concrete) or may develop along specific engineered features as a result of the production and/or hardening process (e.g. surface voids that result from the migration of mainly entrapped air to the fresh concrete-form interface, Samuelson, 1970). The voids often have a well-defined location and geometry and their effect on concrete properties is more easily quantified than the disseminated pathways throughout a concrete matrix as is the case with fractures (Pabalan et al., 2009). An additional complication associated with fractures relates to the time of fracture initiation and their evolution. This is because the processes initiating fractures may occur at different times depending on environmental conditions (e.g. rebar corrosion and earthquakes) which are unpredictable in the long term. Therefore, with the current knowledge, there is no way to quantify a priori evolution of such fractures especially over the long term. This in turn introduces significant uncertainty in performance assessment calculations for cement-based near surface disposal structures. Hereafter, fractures and voids are simply referred to as fracture networks.

As the existence of fracture networks forms preferential pathways for water flow and contaminant transport (Berkowitz et al., 2001; Perko et al., 2012), such networks in concrete are of particular concern for the long-term containment of contaminants (e.g., heavy metals, radionuclides) in disposal facilities in which hazardous wastes are solidified and stabilised in cement-based materials. In addition, disposal facilities such as radioactive waste repositories may contain several structural cement-based components (e.g. walls, floors, etc.) which may lead to the development of a complicated fracture network.

The effect of fracture networks on the release of contaminant fluxes is not univocal and depends on interplay between fracture network characteristics, matrix properties, water flow density and location of encapsulated wastes. Assessment of contaminant leaching is complicated by several additional factors. If contaminants are already present in water-filled fractures, the consequence of transport in the fractures is considered to be generally negative because of bypassing the matrix with high sorption capacity. On the other hand, when contaminants are still in the encapsulated waste, it may have a positive effect because fractures divert percolating water away from the waste zones. Different flow patterns develop depending on different water flow boundary conditions; if the water flux is low relative to the hydraulic conductivity of the fracture the fracture will not act as a preferential pathway and flow will be primarily through the matrix domain. On the other hand, a water flux high enough to saturate the fractures will result in fractures becoming effective pathways for rapid migration of radionuclides. Linked to this complex water flow pattern is the long term evolution of fracture network characteristics such as location, density, orientation and widths. Also the evolution of matrix properties and boundary conditions due to environmental conditions including chemical and physical degradation of cementitious materials affect the flow pattern (e.g., Breysse and Gerard, 1997).

Matrix properties that affect contaminant transport include porosity, bulk density, conductivity, diffusion, dispersivity, sorption and other geochemical reactions (Bodin et al., 2003). While it is reasonable to assume rock matrix to have negligible conductivity as in many hydrogeological conditions, this assumption will not be valid if a concrete structure is assumed to be fully physically degraded, for instance, due to severe micro or macrocrakcing. This means in addition to diffusion, also advection and dispersion should be considered within the concrete matrix. The relative importance of transport in porous matrix and in cracks can be examined by evaluating travel times, breakthrough curves or time of peak flux for various fracture networks and matrix properties.

Several modelling approaches both in the hydrogeological and concrete mechanics community have been proposed to model flow and transport through fractured porous media. The hydrogeological mathematical models for flow and transport through fractured porous media fall into one of three broad classes: (a) equivalent (or single) continuum models and derivatives as dual-porosity and dual-permeability (or multiple) continuum models (e.g. Šimůnek and van Genuchten, 2008), (b) discrete network simulation models either as single fracture or fracture network models (e.g. Sudicky and McLaren, 1992), and (c) hybrid models (e.g. Oda et al., 1987). The equivalent continuum models require volume-averaged hydraulic properties that reflect the large-scale average effects of fractures whilst the discrete models need mainly information about the geometric characteristics of fractures. The hybrid models need both sets of information.

When considering the long term containment of radioactive waste components in a cementitious-based disposal system, the considered time frames can easily extend up to hundreds if not thousands of years. However, there hardly exist historic data on fractures and their evolution in concrete components over such timescales.

The effect of various assumed fracture networks and characteristics on the flow and transport behaviour has already been studied in the past. For instance, Walton et al. (1990) showed via analytical calculations that when cracks are small and closely spaced, resistance in passing through the cracks controls flow. However, when cracks are larger and widely spaced, resistance through the overlying or underlying porous medium (i.e. soil) governs flow rates through cracks. If calculations consider both the adjacent porous media and the cracks, maximum predicted flow rates do not occur for the largest or smallest crack sizes and spacing, but somewhere in between the extremes. In general, their calculations suggest concrete vaults will perform best in terms of flow if cracks are widely spaced and if a low permeability porous material (e.g. clay) is placed next to the concrete. A 2D coupled calcium leaching and radionuclide transport analyses of a section of a fractured concrete container wall by Bejaoui et al. (2007) concluded that fracture density and fracture depth are the two most