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Multi-dimensional transport modelling of corrosive agents through a bentonite buffer in a Canadian deep geological repository

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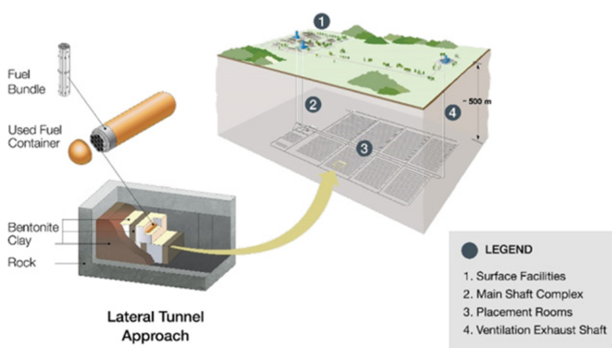
HIGHLIGHTS

- A 3D sulphide transport model of a deep geological repository was developed.
- The model assumed diffusion dominated transport through the repository and bentonite buffer.
- A non-uniform diffusive flux resulted due to the unique geometry of the used fuel container.

GRAPHICAL ABSTRACT

Three-dimensional modelling results suggest sulphide transport through a bentonite buffer to be non-uniform due to the unique geometry of the used fuel container (UFC). The UFC and the engineered barrier are parts of a proposed system for the long-term disposal of used nuclear fuel by Canada's Nuclear Waste Management Organization.



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ABSTRACT

The use of a deep geological repository (DGR) for the long-term disposal of used nuclear fuel is an approach currently being investigated by several agencies worldwide, including Canada's Nuclear Waste Management Organization (NWMO). Within the DGR, used nuclear fuel will be placed in copper-coated steel containers and surrounded by a bentonite clay buffer. While copper is generally thermodynamically stable, corrosion can occur due to the presence of sulphide under anaerobic conditions. As such, understanding transport of sulphide through the engineered barrier system to the used fuel container is an important consideration in DGR design. In this study, a three-dimensional (3D) model of sulphide transport in a DGR was developed. The numerical model is implemented using COMSOL Multiphysics, a commercial finite element software package. Previous sulphide transport models of the NWMO repository used a simplified one-dimensional system. This work illustrates the importance of 3D modelling to capture non-uniform effects, as results showed locations of maximum sulphide flux are 1.7 times higher than the average flux to the used fuel container.

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1. Introduction and background

Long-term solutions for the disposal of high-level used nuclear fuel are being studied by several countries including Canada, Finland, Switzerland, and the United States. Current designs include the use of deep geological repositories (DGRs) which would be located several hundred meters below ground level (Feiveson et al., 2011). In Canada, the Nuclear Waste Management Organization (NWMO) is responsible for the design and implementation of the DGR. NWMO is currently undertaking an extensive site selection process to find a willing and informed community to host Canada's DGR. It is expected that the site will be situated within either a crystalline or a sedimentary environment 500 m below ground surface. The DGR design involves an engineered barrier system (EBS) within a low permeability host rock that serves as a natural barrier. In addition, the EBS includes copper-coated steel used fuel containers (UFCs) within a bentonite clay buffer (Fig. 1).

While copper is naturally corrosion resistant, there are some scenarios under which corrosion could take place, including microbiologically influenced corrosion (MIC). Microbial activity can produce corrosion agents (e.g., ammonia, acetate and nitrite) and sulphide, which could potentially promote both stress corrosion cracking (SCC) corrosion and general corrosion (King and Kolar, 2006). Previous microbial modelling in the Canadian context indicated that the concentration of SCC agents reaching the container surface would be low and unlikely to cause SCC (King and Kolar, 2006; Ikeda and Litke, 2007, Ikeda and Litke, 2008; Litke and Ikeda, 2008, Litke and Ikeda, 2011). The expected absence of SSC is supported by conceptual stress models, as the tensile stresses required for SCC are absent in a DGR (Kwong, 2011). Therefore, SCC of the UFC is not expected and this work is primarily focused on understanding general corrosion due to MIC.

General corrosion of copper, in the form of Cu_2S formation, can occur in the presence of HS^-, as a result of sulphate reducing bacteria (SRB):

$$2Cu(s) + HS^- + H^+ \rightleftarrows Cu_2S(s) + H_2 \quad (1)$$

This reaction is thermodynamically favourable but the corrosion of copper via sulphide is limited by the presence of sulphide species. Since the bentonite buffer is designed to suppress microbial activity, no sulphide production is expected within the bentonite (Stroes-Gascoyne et al., 2010; Bengtsson and Pedersen, 2017). The more likely scenario is that sulphide is produced in the host rock surrounding the bentonite (King, 2007). Therefore, the rate at which sulphide can reach the container surface, causing corrosion, is likely to be controlled by the transport of the sulphide through the bentonite clay barrier.

Bentonite is a swelling clay that has a high sorption capacity resulting in a very low hydraulic conductivity. Therefore, species transport through bentonite is usually diffusion dominated (Alt-Epping et al., 2015; Bourg et al., 2008). Diffusion through porous media (D) is typically modelled considering porosity (θ), tortuosity (τ) of the medium and the free water diffusion coefficient D_0 (Apello, 2013) ($D = \theta\tau D_0$ Eq. (2)).

$$D = \theta\tau D_0 \quad (2)$$

Many studies have been conducted examining how species are transported through swelling clays, and while beyond the scope of the current research, most theories revolve around the double layer theory (Bradbury and Baeyens, 2003; Van Loon et al., 2007; Appelo et al., 2010; Glaus et al., 2011; Apello, 2013; Alt-Epping et al., 2015). In addition, sulphide moving through a bentonite system is subject to geochemical reactions and is sensitive to the porewater of both the bentonite and the host rock (Van Loon et al., 2007; Bourg et al., 2008; Idemitsu et al., 2016), temperature (Martın et al., 2000; Idemitsu et al., 2016), and bentonite density (Van Loon et al., 2007). Furthermore, the density of the bentonite can suppress microbial sulphide-producing activity (Bengtsson and Pedersen, 2017).

The rate of uniform anaerobic sulphide-induced corrosion of copper containers has been conservatively estimated in a Canadian DGR to be less than 1 nm per year for a continuous source concentration of 3 ppm of hydrogen sulphide (King, 1996; Kwong, 2011; Scully and Edwards, 2013). Actual concentrations of sulphide in Canadian groundwater are expected to be considerably lower (Gascoyne, 1996; Kremer, 2016). In the absence of site-specific sulphide measurements, 3 ppm was conservatively used so that initial modelling of the Fickian process could be conducted. Using this value, a corrosion rate of 1 nm/year corresponding to a total depth of corrosion of 1 mm after 1 million years was calculated. This estimate was based on one-dimensional (1D) geometry, a bentonite thickness of 40 cm, and an assumed diffusion coefficient of 1×10^{-11} m^2/s for sulphide. The estimated MIC corrosion rates compares similarly to the value calculated by other nuclear storage organizations (SKB, 2010; King et al., 2013) of less than 1 mm over 1 million years. Given that the NWMO UFC design has a unique cylindrical shaped body with hemi-spherical end caps, there is a need to refine the MIC estimate using a 3D modelling approach that accounts for the more complex UFC geometry and repository layout.

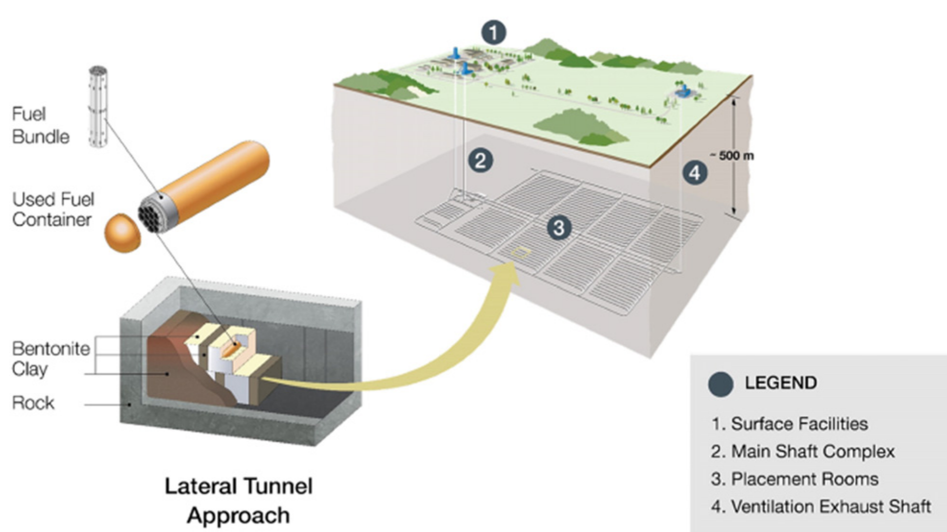

Fig. 1. Schematic of the Canadian DGR design for the long-term disposal of used nuclear fuel. Used nuclear fuel is contained within copper-coated steel containers placed within a bentonite buffer (Figure courtesy of NWMO).