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Water Rock Interaction [WRI 14]

Assessment of microbiological development in nuclear waste geological disposal: a geochemical modeling approach

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Abstract

Deep geological environments are very often poor or devoid of biodegradable organic molecules, but hydrogen could be an efficient energetic source to replace organic matter and promote redox processes such as reduction of O₂, NO₃⁻, Fe³⁺, SO₄²⁻ and CO₂. Moreover, the accessibility and availability of H₂ and nutrients depend on gas/liquid permeability and their migration in the claystone porosity through the excavation damaged zone (EDZ). This study evaluates the spatial and temporal evolution of the geochemical conditions with regard to microbial development. The corrosion process in the argillite is investigated using numerical modeling over a period of 100,000 years. The development of bacterial biomass is estimated using potential redox reactions catalyzed by microorganisms and available nutrients. The simulations show that after the thermal peak (ca. 100-1000 years), physico-chemical conditions are favourable to support bacterial life. Relevant amounts of H₂ and nutrients are released and migrate over the first 2 m of the argillite. Most of the biological redox process are localised close to the container where a high amount of magnetite is produced, providing Fe(III) (electron acceptor) that favours the development of iron-reducing bacteria (IRB).

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Keywords: microbial development, claystone, disposal nuclear waste, geochemical modeling.

1. Introduction

During the last 20 years, numerous studies have shown the existence of a subsurface biosphere e.g. in granitic rocks, clay formations, underground basalts, geothermal water, and oil facilities. The availability of nutrients and energetic sources that can be used by microorganisms in deep clay environments is a key point to the understanding of microbial subsurface life and the impact on the safety of a future geological disposal of nuclear waste. Indeed, such disposal systems aim at preventing water circulation around different metallic packages, because corrosion is the main factor responsible for the

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alteration of the waste packages. Moreover, this phenomenon may be enhanced by microbiologically influenced corrosion [1, 2]. Deep geological environments are very often poor or devoid of biodegradable organic molecules but some authors indicate that hydrogen will be an efficient energetic source to replace organic matter [3]. Large amounts of H₂ are expected to be generated, originating from the corrosion processes of metallic components in anoxic conditions [4] and also from radiolysis [5]. Indeed, many authors have demonstrated a variety of waste disposal conditions in which some microorganisms are able to use hydrogen for redox processes such as reduction of O₂, NO₃⁻, Fe³⁺, SO₄²⁻ and CO₂ [3]. Moreover, porosity seems to be accessible to gas and H₂ could therefore migrate through the argillites and the connected fractured zone (CFZ). The main goal of this study is to evaluate the spatial and temporal evolution of the geochemical conditions with regard to microbial development, i.e. to look at the availability and the migration of nutrients and energetic substrates through the clay barrier, especially during the corrosion process.

2. Material and methods

2.1. Concept

The approach consists in simulating HLW packages corrosion in contact with claystone during anoxic period of nuclear waste disposal and assesses the migration of nutrients and energetic compounds through the CFZ. The initial system is defined in 3 parts: the interstitial water (based on [6]); the claystone primary minerals (based on [7]); and the HLW packages composed of carbon steel P235 [8].

2.2. Model

The corrosion process in the argillite is investigated using numerical modeling at 90°C over a period of 100,000 years with a purely diffusive transport in water saturated medium at fixed porosity. The stainless steel container is represented by a material containing metallic iron and trace elements (Fe, C, N, P, S) and the corrosion proceeds at a rate of 5 µm/year. Simulations were performed with the reactive transport code CRUNCHFLOW, using the Thermoddem 1v12 database (BRGM).

The development of bacterial biomass is estimated using potential redox reactions catalyzed by microorganisms and available nutrients. For this purpose, two parameters are used:

- Biomass formula C₅ H₇ O₂ N_{0,75} P_{0,05} S_{0,025} following average chemical [9],
- The energy required for the synthesis reaction of a gram of biomass in this study: ΔG°_w = 64 kJ.mol⁻¹ e⁻, standard value [10].

3. Results: spatio-temporal evolution

3.1. Corrosion product

With a corrosion rate of 5 µm.year⁻¹, the container is totally corroded in 30 000 years (Figure 1). In this scenario, the main corrosion products (magnetite) are formed directly in replacement of the container, or in the interface between container and claystone (siderite + Fe-rich clay minerals + sulphide minerals). Moreover, we can observe the formation of a phosphorus mineral, vivianite, both in the container zone and at the interface canister/claystone (Figure 1). Vivianite precipitation can reach up to 4 % of mineral volume. Thus, high amount of potential nutrients for microbial development are immobilized close to the container, preventing the nutrients from migrating through the connected fractured zone.

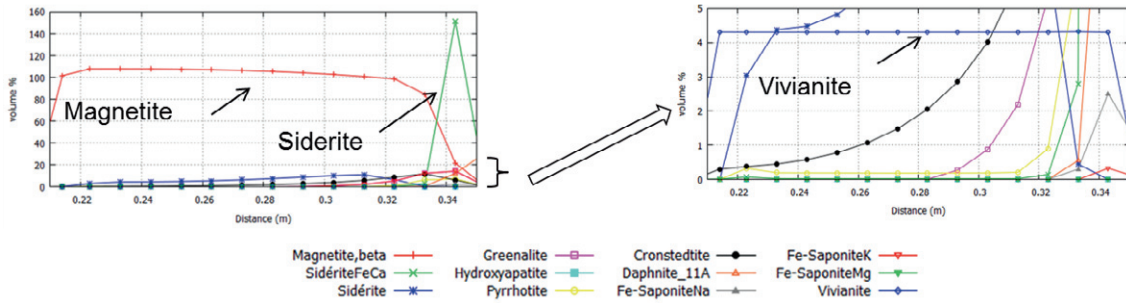


Fig. 1. Corrosion product volume percent at 30,000 years (end of corrosion) in the container zone.

3.2. Physicochemical conditions

Evolution of physicochemical parameters shows pH values in the range of 8 to 9, during all the corrosion process in CFZ (Figure 2, left). This represents acceptable conditions for microbial development. Dissolved H₂ evolution also shows a migration from waste package to claystone. H₂ appears available for microbial development in sufficient quantity after 100 years in all the CFZ (Figure 2, right).

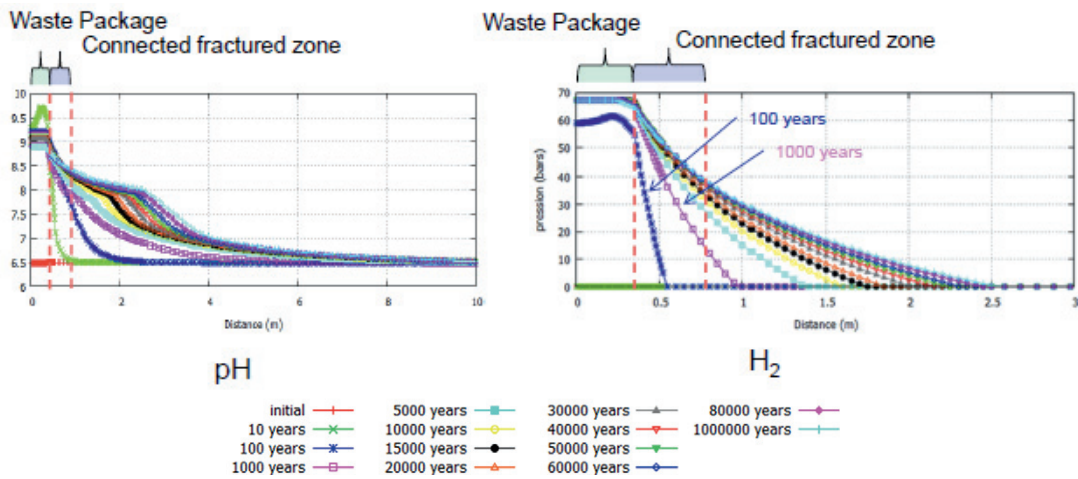


Fig. 2. Evolution of pH and H₂ in claystone during HLW packages corrosion process.

3.3. Nutrients

Although a fraction of the nutrients are trapped close to the containers, especially phosphate and sulphate, some of them, carbonates, phosphates and nitrogen, follow a diffusion pattern through the connected fractured zone and the sound claystone (Figure not shown).

4. Discussion and Conclusion

The simulations show that after the thermal peak (ca. 100-1000 years), physico-chemical conditions are favourable to support bacterial life. Relevant amounts of H₂ and nutrients are released and migrate by diffusion in the first meter in the argillite.

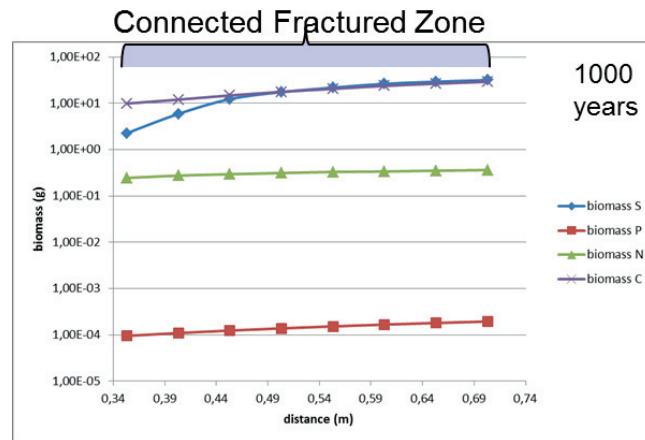


Fig. 3. Assessment of potential bacteria development in gram, according to calculated concentration of nutrients in the connected fractured zone of claystone, 1000 years after the beginning of HLW packages corrosion process.

Phosphorus has been determined as the main limiting nutrient for bacteria growth in this scenario (Figure 3). This simulation shows that most of the biological redox process could be localised close to the container where nutrients are trapped in mineral phases (in particular as a source for phosphate) and where a high amount of magnetite is produced, providing Fe(III) (electron acceptor) which can especially favour the development of iron-reducing bacteria (IRB).

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