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Water Rock Interaction [WRI 14]**Are underground clay disposal conditions favorable for microbial activity and biocorrosion?**M. Libert^{a,*}, M. Kerber-Schütz^a, L. Esnault^b, O. Bildstein^a^aCEA, DEN/DTN/SMTM/LMTE, bât. 307, 13108 Saint Paul Lez Durance Cedex, France^bECOGEOSAFE, Technopôle de l'Environnement Arbois- Méditerranée, BP 90027 Aix en Provence, France

Abstract

The French concept for geological disposal of high-level radioactive waste is based on a multi-barrier system including metallic containers confined in a claystone layer. The main alteration vector is water coming from the host rock and triggering corrosion of metallic components. Despite extreme conditions, microorganisms can adapt and survive in these environments. Anoxic corrosion of metallic containers and water radiolysis produce H₂, which potentially represents an abundant energetic source for microbial development, especially in this type of environment containing low amounts of biodegradable organic matter. Moreover, formation of Fe(III)-bearing corrosion products such as magnetite (Fe₃O₄) can provide electron acceptors for microbial development. Therefore, biocorrosion studies are needed in order to investigate the activity of hydrogenotrophic bacteria able to reduce sulphates or Fe(III) from iron oxides (passive layer). These studies help in evaluating such microbial impacts on the long-term stability of metallic components involved in radioactive waste disposal.

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Keywords: underground disposal, biocorrosion, iron-reducing bacteria, hydrogen, anoxic conditions, radioactive waste.

1. Introduction

Geological disposal of High Level Radioactive Waste (HLW) is investigated in several countries as a means to isolate them from the biosphere for long periods of time. According to current design in France [1], HLW will be stored in stainless steel containers and low-alloy steel overpacks, emplaced then directly in the clay host rock of the geological repository. This multi-barrier system is designed to delay the access of water to the waste and eventually to limit the migration of radionuclides. An underground research

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laboratory built at 500 m depth in the Callovo-Oxfordian clay formation in eastern France confirmed the feasibility of a nuclear waste disposal site in this rock formation. But with time, the main alteration vector will be the water coming from the geological formation and eventually reaching the waste matrix. Therefore, anoxic aqueous iron corrosion phenomena, especially over a long time, must be understood in order to evaluate the confinement capacity of the repository.

Despite the extreme conditions in the deep geological disposal (redox conditions, high pressure and temperature, irradiation), microorganisms can adapt and survive in these environments. The effect of microbial activity on the corrosion processes is a well-known mechanism that could increase the steel corrosion rate [2]. Bacteria can act directly on iron reduction [3] (catalysed by iron-reducing bacteria) or through their metabolism products, such as organic acids and corrosive hydrogen sulphide produced by sulphate-reducing bacteria [4]. However, remaining questions are still under debate: are bacteria able to grow in the extreme conditions of a geological disposal? Which species? Which kind of metabolism do they develop? Are they inducing biocorrosion phenomena, and if yes, is it possible to quantify their effects on the safe disposal?

This paper focuses on the potential microbial activity in an underground disposal, with the identification of the reservoirs of nutrients able to support such activity. Among them, corrosion products such as iron oxides and H_2 can promote the development of hydrogen-oxidizing bacteria (HOB) and iron-reducing bacteria (IRB). Their impact in repository conditions is under investigation.

2. Interactions

Figure 1 summarizes the different interactions prevailing in an underground disposal between materials, medium and microorganisms. This “3M approach” has been previously described for others biocorrosion situations [11].

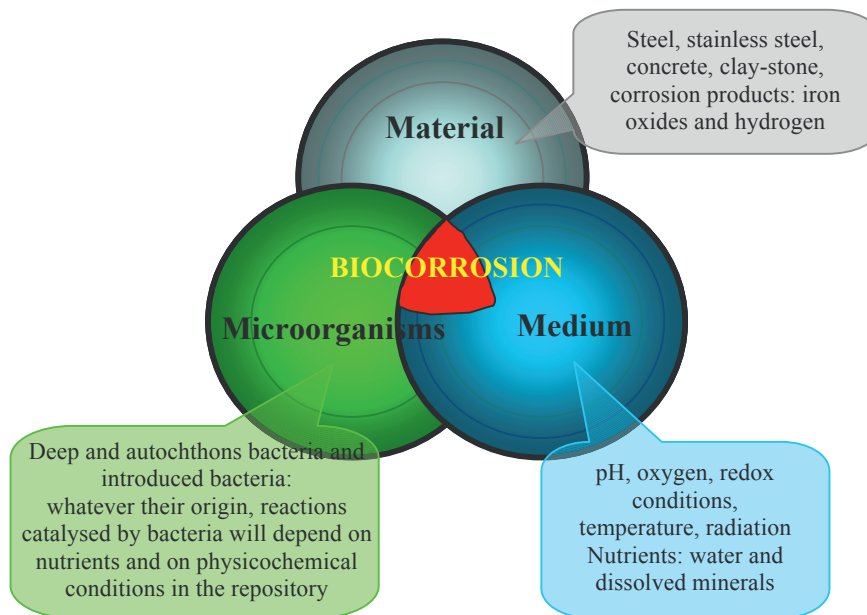


Fig 1. Interactions between metallic materials, porous medium (host rock), and microorganisms.

2.1 Extreme conditions

The environmental conditions within repository will change with time: after closure, the conditions will be fairly high temperature, oxidising, water unsaturated and with a high amount of radiation. However, after a period of about hundred years, the dose rate of radiation will decrease (to a value in the order of 50 Gy/h), the temperature will reach about 50°C, and the geochemical conditions will change to anoxic. The dynamics of resaturation in the repository by interstitial water coming from the geological formation is complex and depends on the production of H₂, but the saturation is thought to remain high, about 97%, during this time span [1].

Consequently, these conditions could lead to microbial activity due to microorganisms already present in the deep geological clay formations, which can survive even under extreme conditions, and to microorganisms introduced into disposal facilities as a consequence of the human activities during the construction and operational phases [5].

Despite a porosity dominated by pore sizes of a few tens of nanometers in the claystone, small cracks in the connected fractured zone (due to excavation operations) and remaining pore spaces can still be favourable for anaerobic microbial growth. These void spaces may remain open for thousands of years as a consequence of H₂ pressure build-up resulted from the anoxic metallic corrosion [6].

2.2 Nutrients

The availability of nutrients and energetic sources able to be used in deep geological environments is a key point to understand microbial subsurface life. An inventory of nutrients, energetic sources and thermodynamically possible reactions has been performed in the case of the French geological disposal [7,8,9].

Different reservoirs of nutrients able to support microbial activity have been identified. Among them, minerals and organic matter of the host rock, soluble nutrients present in the interstitial water and corrosion products. Metallic corrosion can also be considered as a source of nutritional and energetic substrates for microbial growth. High amount of H₂ gas will be produced as corrosion product within nuclear waste repository as well as from water radiolysis, and consequently will support microbial activity in this specific environment poor in organic matter. H₂ is known as one of the most energetic substrates for deep terrestrial subsurface environments [10].

We have previously demonstrated that microorganisms are able to use H₂ for redox processes (reduction of O₂, NO₃⁻, Fe⁺³) in several waste disposal conditions, such as basins containing irradiating waste, bituminous waste disposal and clay transformation by bioreduction of the structural Fe(III) [8]. It is noteworthy that such microbial activity can also modify iron (hydr)oxides (e.g. magnetite, Fe₃O₄) formed as corrosion products (as passive layers) by Fe(III) reduction coupled to H₂ oxidation. Therefore, the activities of HOB and IRB become parameters to be evaluated regarding the safety of the current disposal concept.

3. Conclusion

Large amounts of H₂ are expected to be generated, originating from the corrosion processes of metallic components in anoxic conditions and also from water radiolysis. It will favour the development of hydrogenotrophic bacteria, but high concentrations of H₂ may also inhibit some metabolisms. Microbial development in underground disposal can result in changes in the groundwater chemistry and the geochemical equilibrium, gas production or consumption, biocorrosion and potential modifications of confinement properties.

Bacteria have to develop strategies such as modification of chemical conditions (pH, Eh) to access energetic substrates, especially for Fe(III) reduction from clay or magnetite.

Complementary studies are now in progress (1) to clarify the biocorrosion mechanisms, (2) to determine the parameters governing the corrosion rate in order to assess the long-term behaviour of the system and (3) to quantify phenomena on a repository scale with integrated experiments. These microorganism-mediated reactions will then be integrated in reactive transport models.

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