



Limits to the use of highly compacted bentonite as a deterrent for microbiologically influenced corrosion in a nuclear fuel waste repository

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ABSTRACT

Recent studies have suggested that microbial activity in highly compacted bentonite ($\geq 1600 \text{ kg/m}^3$) is severely suppressed. Therefore, it appears that the dry density of emplaced bentonite barriers in a geological repository for nuclear waste may be tailored such that a microbiologically unfavorable environment can be created adjacent to used fuel containers. This would ensure that microbiologically influenced corrosion is a negligible contributor to the overall corrosion process. However, this premise is valid only as long as the emplaced bentonite maintains a uniform high dry density ($\geq 1600 \text{ kg/m}^3$) because it has been shown that high dry density only suppresses microbial activity but not necessarily eliminates the viable microbial population in bentonite. In a repository, a reduction in the dry density of highly compacted bentonite may occur at a number of interface locations, such as placement gaps, contact regions with materials of different densities and contact points with water-carrying fractures in the rock. Experiments were carried out in our laboratory to examine the effects of a reduction in dry density (from 1600 kg/m^3 to about 1000 kg/m^3) on the recovery of microbial culturability in compacted bentonite. Results showed that upon expansion of compacted bentonite into a void, the resulting reduction in dry density stimulated or restored culturability of indigenous microbes. In a repository this would increase the possibility of *in situ* activity, which might be detrimental for the longevity of waste containers. Reductions in dry density, therefore, should be minimized or eliminated by adequate design and placement methods of compacted bentonite. Materials compliance models can be used to determine the required as-placed dry densities of bentonite buffer and gap fillings to achieve specific targets for long-term equilibrium dry densities for various container placement room designs. Locations where flowing fractures could be in contact with highly compacted bentonite should either be avoided or grouted adequately to limit contact between bentonite and flowing water. Even if localized enhanced microbial activity at interfaces remains of concern, despite adequate engineering and placement methods, the potential consequences can be assessed and quantified adequately by a combination of *in situ* activity measurements and modeling calculations.

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1. Introduction

Highly compacted bentonite-based sealing materials are being developed for use in future deep geological repositories for high-level nuclear (fuel) waste. Such materials would protect the waste containers physically and chemically, would ensure a diffusion-controlled transport of solutes, and would form a sorption barrier against radionuclide migration after container breach and waste form dissolution.

The possibility of microbial activity in bentonite-based sealing materials in a deep geological repository for high-level nuclear waste is of concern for a number of reasons (e.g. Pedersen, 1999,

2000; Stroes-Gascoyne and West, 1996, 1997). A main concern is that such activity may result in microbiologically influenced corrosion (MIC), which could affect the longevity of the containers, through the formation of corrosion-inducing aggressive environments under biofilms or through the production of corrosive metabolites. For the latter, sulfate-reducing bacteria (SRB) that produce sulfides are of specific concern. Another concern is that mobile microbes may absorb radionuclides (released from breached containers) on or inside their cells and act as colloids, potentially increasing the migration of radionuclides through these engineered barriers. Microbial gas production also needs to be considered because this may cause a build-up of a gas phase in a repository, potentially reducing the effectiveness of the bentonite-based barriers. Fortunately, because of the inherent and specific physical characteristics of highly compacted bentonite, such as small pores, low water activity (a_w) and high swelling pressure,

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an additional role of highly compacted bentonite may be the elimination of significant microbial activity in such sealing materials. A suppression of microbial activity in sealing materials around waste containers would reduce the contribution of MIC to the overall corrosion process to insignificant levels.

As-received commercial bentonite is not sterile. Masurat et al. (2010a) concluded from enrichment cultures in SRB-selective medium with Wyoming MX-80 bentonite inoculum that the SRB species *Desulfovibrio africanus* was present in this commercial bentonite. This species was able to survive in a state of desiccation at high temperatures (100 °C) and high salt concentration (4%) and could be induced to sulfate-reducing activity under suitable conditions. Stroes-Gascoyne et al. (2006) cultured heterotrophic aerobic bacteria (HAB) ($(2.07 \pm 0.23) \times 10^2$ Colony Forming Units (CFU)/g), heterotrophic anaerobic bacteria (HANB) ($(5.51 \pm 1.91) \times 10^1$ CFU/g), and SRB (1.0×10^1 most probable number (MPN)/g) from as-received MX-80 bentonite powder that had a water content of 9.3%, a_w value of 0.471 and had been stored under oxic conditions. Subsequent repeat analyses of the as-received bentonite showed slowly decreasing numbers for HAB (Stroes-Gascoyne and Hamon, 2008). Fig. 1 shows that the Phospholipid Fatty Acid (PLFA)-based biomass in the as-received MX-80 bentonite suggested a viable population of about 10^6 cells/g. This figure illustrates that few of the viable cells present in this as-received bentonite were culturable (Stroes-Gascoyne et al., 2010).

Highly compacted saturated bentonite is also not sterile. Fru and Athar (2008) studied the bacterial population of compacted Wyoming MX-80 bentonite (at saturated conditions for a dry density of 1600 kg/m³ (wet density of 2000 kg/m³) and elevated temperatures) in contact for 5 years with granitic groundwater that contained indigenous microbes. Results suggested that the bacterial population in highly compacted bentonite consisted largely of gram-positive spore-formers, contrary to the largely gram-negative bacterial population dominant in the groundwater, which, therefore, did not appear to have colonized the compacted bentonite over the period of 5 years. Recently, HAB isolates on R2A medium (Reasoner and Geldreich, 1985) from both low-compacted and highly compacted saturated Wyoming MX-80 bentonite plugs (with dry densities of 800 and 1600 kg/m³) were identified by the BIOLOG™ system in our laboratory. Results indicated the almost exclusive presence of gram-positive spore-forming bacteria (Stroes-Gascoyne et al., 2009, unpublished data), corroborating

the results of Fru and Athar (2008). These results illustrate that commercial bentonites contain a viable microbial population that is quite hardy, not easy to eliminate and possibly revivable to *in situ* activity.

Stroes-Gascoyne et al. (2010) investigated the effects of the physical properties of highly compacted MX-80 saturated bentonite on the culturability of its indigenous microbial population. Fig. 1 shows that at a measured dry density of about 750 kg/m³ (with a corresponding swelling pressure of about 0.2 MPa and a_w value of 0.998, as measured in our laboratory), aerobic culturability in saturated compacted MX-80 bentonite increased by three orders of magnitude compared to the aerobic culturability in the as-received bentonite powder (from 1.4×10^2 to 1.4×10^5 CFU HAB/g). At a dry density of 1300 kg/m³ (with a corresponding swelling pressure of 1.9 MPa and a_w value of 0.980, as measured in our laboratory) aerobic culturability dropped to the level in the as-received bentonite. At dry densities of 1600 and 1800 kg/m³, aerobic culturability dropped below that of dry as-received bentonite, while the PLFA-based biomass remained stable at $(1.5 \pm 0.1) \times 10^6$ cells/g. The large increase in culturability at low dry density was not accompanied by a large increase in PLFA-based viability. This implies an increase from 0.01% HAB culturability in as-received bentonite to 6% HAB culturability in saturated compacted bentonite with a low dry density of 750 kg/m³. The culturability at dry densities of 1300, 1600 and 1800 kg/m³ declined steadily from about 0.008–0.004% of the PLFA-indicated viable population. This suggests that the indigenous cells in the as-received MX-80 bentonite powder could regain considerable culturability (and hence potential *in situ* activity) in saturated compacted bentonite, but only at low dry densities.

Stroes-Gascoyne et al. (2010) confirmed that a swelling pressure of about 2 MPa with a corresponding low a_w value of about 0.96 appears to keep aerobic culturability at or below background levels in the as-received MX-80 bentonite. Most of the pore diameters in the highly compacted bentonite were measured around 0.02 µm (with a range from ~0.005 to 0.1 µm), with a very small population of macropores in the range of 5–100 µm (Stroes-Gascoyne et al., 2010), using mercury intrusion porosimetry. Dixon et al. (1999) reported a large component of micropores at about 0.02 µm in highly compacted Wyoming bentonite with a much smaller population of macropores in the range of 10–200 µm. In comparison, the size range for most vegetative microbes is 0.5–10 µm,

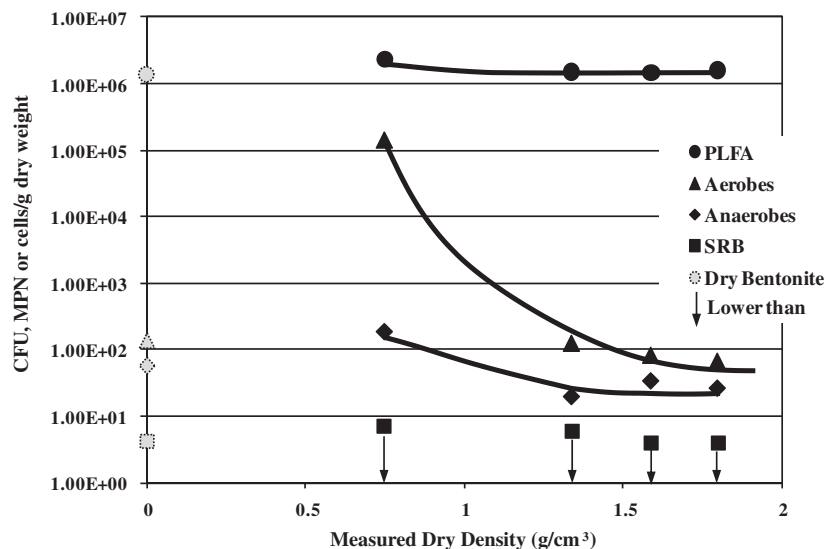


Fig. 1. Comparison of culturable and PLFA-indicated viable biomass in saturated Wyoming MX-80 compacted to target dry densities ranging from 800 to 1800 kg/m³ and in as-received bentonite powder (Stroes-Gascoyne et al. 2010).

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