MODELING MICROBIALLY-MEDIATED CONSUMPTION OF OXYGEN TRAPPED IN VOIDS OF A POTENTIAL REPOSITORY AFTER BACKFILLING AT ÄSPÖ SITE

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ABSTRACT. The presence of molecular oxygen left in the voids of a deep geological repository of high level radioactive waste (HLW) after backfilling could affect the corrosion of canisters and the migration of radionuclides eventually released from a damaged canister. Knowing how O₂ is consumed is therefore important for safety analyses. Available data from laboratory and in situ experiments indicate that microbes play a substantial role to restore redox condition near the HLW repository. The REX in situ experiment was conducted at the Åspö Hard Rock Laboratory by the Swedish Nuclear Waste Management Company to investigate microbial nutrients, organism diversity, microbial activity and O₂ reduction. This paper presents a hydro-bio-geochemical model to evaluate how O₂ is consumed after backfilling of a HLW repository planned according to the Swedish reference concept. The microbial model accounts for dissolved organic carbon (DOC) respiration and methane oxidation. Parameters for these processes were calibrated with measured oxygen in the REX experiment. Computed concentrations of oxygen match measured data in the chamber of the REX experiment. Calibration results indicate that the microbial model can be used to simulate the processes of O₂ reduction trapped in the voids of the bentonite buffer. The role of microbes in the consumption of O₂ is evaluated for several cases corresponding to various hypotheses. Numerical results show that microbial processes are relevant for O₂ consumption in the repository. The time needed to consume the O₂ trapped in the buffer decreases from over 50 for only geochemical processes to a few months when the catalytic effect of microbes in DOC respiration and methane oxidation is taken into account.

RESUMEN. El oxígeno molecular disuelto en los poros del material de relleno y sellado de un almacenamiento geológico profundo (AGP) de residuos radiactivos después de su clausura podría afectar negativamente en la corrosión de los contenedores y en la migración de los radionucleidos que se pueden liberar tras el colapso del contenedor. Para la evaluación de la seguridad de un AGP es importante saber cómo se consume el O₂. Los datos disponibles procedentes tanto de ensayos de laboratorio como ensayos in situ en laboratorios subterráneos indican que los microorganismos desempeñan un papel relevante en el restablecimiento de las condiciones redox en el entorno de un AGP. El experimento in situ REX fue realizado en el laboratorio subterráneo de Åspó (Suecia) por SKB (compañía sueca para la gestión de residuos radiactivos) para estudiar los nutrientes microbiológicos, la diversidad de microorganismos y su actividad. Este trabajo presenta un modelo acoplado hidro-bio-geoquímico para evaluar la reducción del O₂ disuelto presente en la barrera de bentonita de un AGP de las características contempladas por SKB. El modelo microbiológico tiene en cuenta la respiración del carbono orgánico disuelto (DOC) y la oxidación del metano. Los parámetros para estos procesos han sido calibrados con los valores de oxígeno medidos en el experimento REX. Los valores calculados de la concentración de oxígeno reproducen los datos medidos en la cámara del citado experimento. Los resultados de la calibración indican que el modelo microbiológico puede ser utilizado para simular los procesos de reducción del O₂. El papel de los microorganismos en la reducción del O₂ se analiza con varias hipótesis. Los resultados numéricos muestran que los procesos microbiológicos son relevantes para la desaparición del O₂ en el AGP. La reducción del oxígeno mediante procesos exclusivamente químicos requiere casi 50 años para agotar el O₂ atrapado en la bentonita. Este tiempo se reduce a unos pocos meses cuando se considera además de los procesos químicos el papel catalizador de los microorganismos de los procesos de respiración del DOC y la oxidación del metano.

1. Introduction

Underground facilities are being operated by several countries around the world for performing research and providing demonstration of the safety of deep radioactive waste repositories. The Aspö Hard Rock Laboratory is one of such facilities launched and operated by the Swedish Nuclear Waste Management Company where various in situ experiments have been carried out in fractured granites.
One of the main tasks of those experiments is to evaluate the stability of redox conditions in the engineered barriers and the surrounding rock. Redox conditions in deep hard rock environments are usually stable with redox potentials between -100 and -400 mV (Haveman et al. 1998). When the repository is constructed, oxygen could be introduced and the redox conditions could be affected. After the tunnels have been sealed, oxygen will still remain trapped in backfilling and buffer materials. Therefore, evaluating consumption of O₂ is very important for the performance assessment of a HLW repository.

Several researchers reported that microbes could catalyze redox reactions in the repository environment and take a significant role in the stability of redox condition in the area adjacent to and within nuclear waste canisters (Pedersen 1995, 1999, 2000a, 2000b, Jolley et al. 2003). Available data from REX in situ experiment at Äspö site conducted by SKB confirm that microbes play a substantial role in O₂ consumption. This experiment studied O₂ depletion processes by imposing a controlled oxidizing perturbation in a deep rock environment at the Äspö site. Oxygen was injected into a fracture zone and water samples were collected for microbiological analyses. The REX niche is located 380 m below ground surface. The experiment was performed in a closed recirculating system placed in a borehole.

The circulation loop had a total volume of about 1 L, and before each O₂ pulse it was filled with fresh O₂-free groundwater from an adjacent borehole. O₂ injection pulses started by replacing part of the volume by groundwater samples that had been previously saturated with either air or O₂(g) (Puigdomenech et al. 2001). The main objective of this paper is to present a hydrobiogeochemical model to evaluate the role of microbes in the consumption of O₂ trapped in the voids after backfilling a potential repository. BIOCORE2D is used to solve the coupled model.

BIOCORE2D is a general purpose code, presented by Zhang (2001) and improved by Samper et al. (2005a). BIOCORE2D incorporates groundwater flow, thermal and multicomponent solute transport in variably saturated porous media with a complete suite of abiotic, such as aqueous complexation, adsorption, ion-exchange, redox, precipitation-dissolution, acid-base reactions, and microbial growth transformation processes accounting for metabolic competition, decay, metabolism and endogenous respiration and the resistance of the biofilm to the substrates.

BIOCORE2D has been verified with some other codes, such as BIOLOG3D (Engesgaard, 2000) and FEREACT (Tebes et al., 1998). BIOCORE2D has been applied to several real cases, such as the biotransformation of Fe(III) mineral (goethite) to Fe(II) mineral (pyrite) in the near field of Boom clay (Zhang, 2001) and the interpretation of the increase in concentrations of bicarbonate and sulfate measured at the in situ Redox Experiment (Samper et al. 2004, Molinero et al. 2004).

2. Microbial models

The in situ REX experiment shows that different microbial groups could theoretically contribute to the reduction of oxygen at the Äspö site and two microbial processes control oxygen consumption. One is DOC respiration and the other is methane oxidation (Samper et al. 2005a).

2.1. DOC Respiration

Dissolved Organic Carbon (DOC) is a complex mixture of substances derived from biotic and abiotic reactions. It is both consumed and produced by microorganisms. One of the substances, glucose, is an easily respirable substrate, but it is normally not found in groundwaters. Acetate is a key intermediate product in anaerobic metabolism of organic matter both in marine and groundwater environments, being the main product of anaerobic DOC degradation. Formate is also an important intermediate compound in anaerobic reactions. Formate may be present as a transit molecule in metabolic pathways. Both processes have been lumped into a single process “DOC-respiration” in this study. Microbial aerobic respiration of DOC is a very efficient process for oxygen consumption in groundwaters. This process has been extensively studied at the REX project of the Äspö HRL (Samper et al. 2005). Aerobic heterotrophic bacteria are expected to play a relevant role in the post-closure stage of a repository. DOC respiration is simplified to proceed according to:

\[ CH_2O + O_2 \xrightarrow{\text{heterotrophs}} HCO_3^- + H^+ + \text{Biomass} \]

Microorganisms performing the catalysis of this process are aerobic heterotrophs, probably of the bacterial group pseudomonas. The growth rate of heterotrophic microbes, \( r_{\text{het}} \), is assumed to follow a dual Monod kinetics:

\[
r_{\text{het}} = \mu_{\text{het}} C_{\text{DOC}} \frac{C_{\text{O}_2}}{(K_{\text{DOC}} + C_{\text{DOC}})(K_{\text{O}_2} + C_{\text{O}_2})} \tag{1}
\]

where \( C_{\text{het}} \) is the concentration of aerobic heterotrophs, \( \mu_{\text{het}} \) is the specific growth rate, subindexes DOC and O₂ refer to dissolved organic matter (substrate) and oxygen (electron acceptor), respectively and, \( K \) denotes half-saturation constants.

The rate of dissolve organic matter consumption, \( r_{\text{DOC}} \), is related to the rate of microbial growth through:

\[
r_{\text{DOC}} = \frac{1}{Y_{\text{DOC}}} r_{\text{het}} \tag{2}
\]

The consumption rate of electron acceptor, \( r_{\text{O}_2} \), is related to the rate of microbial growth through:

\[
r_{\text{O}_2} = \frac{f_{\text{O}_2}}{Y_{\text{DOC}}} r_{\text{het}} \tag{3}
\]

where \( Y_{\text{DOC}} \) is the yield coefficient, \( f_{\text{O}_2} \) is the proportionality coefficient between consumed substrate and electron acceptor, which is equal to 1.
Available values of DOC respiration kinetic parameters have been reported by several researchers (Qu and Bhattacharya, 1996; Kotelnikova & Pedersen 1999). Vogelaar et al. (2003) also reported kinetic parameters describing the growth and decay of mesophilic (30 °C) aerobic biomass, a kind of bacteria involved in the degradation of DOC. The parameters were determined in continuous and batch experiments from measured oxygen uptake rates. The intrinsic maximum growth rate was 0.48 ± 0.11 h⁻¹ while biomass decay rate was 0.004 h⁻¹. Biomass yield coefficient takes a value of 0.5 g biomass per gram of acetate.

2.2. Methane oxidation

The REX project revealed that methane oxidizers are very common in the Äspö HRL tunnel (Kotelnikova & Pedersen, 1999). CH₄ and H₂ have been detected in all investigated boreholes in Äspö with concentrations which exceed the half-saturation constants of competent microorganisms able to oxidize those substrates. This indicates that concentrations of these compounds are large enough to support significant microbial growth. Measured concentrations of dissolved methane in Äspö groundwaters range from 0.9 up to 980 µM. Active methanotrophic bacteria have been found in Äspö groundwaters. In fact a new species, Methylomonas Scandinavica, was isolated from Äspö groundwaters and characterized for the first time (Kalyuznaya et al., 1999). Physiological characterization of Methylomonas Scandinavica revealed that this species is able to grow in the range of 5-30 °C and pH = 5.0-9.0, with optimal growth at 17 °C and pH =6.8-7.8. It was also found that it corresponds to a halophilic species which is able to grow in a wide range of salinity. This species is clearly adapted to the environmental conditions prevailing at the Äspö site.

Methanotrophic bacteria are relevant microbial species in the consumption of oxygen. In this case, methanotrophs are expected to be one important oxygen consumers in the geosphere. Microbially-driven methane oxidation is assumed to proceed according to the following reaction:

\[
\text{CH}_4 + 2\text{O}_2 \rightarrow \text{HCO}_3^- + \text{H}^+ + \text{H}_2\text{O} + \text{Biomass}
\]

which is catalyzed by *Methylomonas Scandinavica*. The growth rate of methanotrophic bacteria, \( r_{\text{growth}}^{\text{met}} \), is governed by dual Monod kinetics,

\[
r_{\text{growth}}^{\text{met}} = \mu_{\text{max}}^{\text{met}} \frac{C_{\text{CH}_4}}{K_{\text{CH}_4} + C_{\text{CH}_4}} \frac{C_{\text{O}_2}}{K_{\text{O}_2} + C_{\text{O}_2}}
\]

where \( C_{\text{met}} \) is the concentration of aerobic methanotrophs, \( \mu_{\text{max}}^{\text{met}} \) is the maximum growth rate, and subindexes \( \text{CH}_4 \) and \( \text{O}_2 \) refer to dissolved methane (substrate) and oxygen (electron acceptor), respectively. The rate of substrate consumption (methane) is related to microbial growth rate through

\[
r_{\text{CH}_4}^{\text{met}} = \frac{1}{Y_{\text{CH}_4}^{\text{met}}} r_{\text{growth}}^{\text{met}}
\]

Similarly, the rate of electron acceptor, \( r_{\text{O}_2}^{\text{met}} \), is related to microbial growth rate through

\[
r_{\text{O}_2}^{\text{met}} = \frac{f_{\text{O}_2}}{Y_{\text{CH}_4}^{\text{met}}} r_{\text{growth}}^{\text{met}}
\]

where \( Y_{\text{CH}_4} \) is yield coefficient, and \( f_{\text{O}_2} \) is the proportionality coefficient between consumed substrate and consumed electron acceptor.

Pedersen (1992) reported the following values for maximum growth rate, decay rate and yield coefficient of methane oxidizer: 12.9 d⁻¹, 0.96 d⁻¹ and 0.57 g biomass/g CH₄, respectively. A detailed survey of microbial kinetic parameters is presented by Samper et al. (2005a).

2.3. Model calibration

Both microbial processes take O₂ as an electron acceptor and compete to consume O₂. The consumption rate of O₂ depends on the values of microbial kinetic parameters in 1 to 6. Therefore, it is crucial to choose proper values of kinetic parameters to simulate the consumption of oxygen trapped in voids of the repository.

Using the reported values of microbial kinetic parameters, the microbial model was calibrated by trial and error to fit the data of oxygen measured in the REX in situ experiment (Samper et al., 2005a).

The calibrated kinetic parameters are listed in Table 1. Fig. 1 shows the computed and measured time evolutions of oxygen concentration. It can be seen that computed concentrations of oxygen match well measured data in the REX Experiment.

### Table 1. Calibrated kinetic parameters for two microbial processes at the Äspö site

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Calibrated Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum growth rate</td>
<td>0.215 d⁻¹</td>
</tr>
<tr>
<td>Yield coefficient</td>
<td>0.41 gB / gCH₄</td>
</tr>
<tr>
<td>Half-saturation methane</td>
<td>32.5 µM</td>
</tr>
<tr>
<td>Half-saturation oxygen</td>
<td>1.5 µM</td>
</tr>
<tr>
<td>Maximum growth rate</td>
<td>0.48 h⁻¹</td>
</tr>
<tr>
<td>Decay rate</td>
<td>0.04 h⁻¹</td>
</tr>
<tr>
<td>Yield coefficient</td>
<td>0.5 gBiomass / gDOC</td>
</tr>
<tr>
<td>Half saturation DOC</td>
<td>125 µM</td>
</tr>
</tbody>
</table>

Fig. 1. Computed (line) and measured dissolved oxygen concentration
3. Evaluating consumption of O\textsubscript{2} left in the bentonite after the backfilling

3.1. Hydrobiogeochemical model

The consumption of O\textsubscript{2} trapped in the voids of a potential repository is evaluated with a multicomponent reactive model which accounts for 18 primary species, more than 60 aqueous complexes, 7 minerals (two of them are kinetically controlled) and 2 microbes (see Table 2) (Samper et al. 2005b). The model domain includes a 2D axisymmetric domain (Fig. 2), which consists of two material zones: granite and bentonite. Solute transport takes place only by diffusion.

The inner boundary is assumed impervious while at the outer boundary the concentration is prescribed. The model considers the Åspö granite and the bentonite buffer made of MX-80 bentonite. The bentonite buffer is assumed to be water saturated. Chemical compositions of granite and bentonite porewater as well as boundary water are listed in Table 3. Chemical composition of granite was selected from water samples collected on February 6th 1998 at borehole KA2861A (Samper et al., 2005a and b). The composition of initial water in bentonite considered in the numerical model of the LOT experiment by Arcos et al. (2003) was adopted in the current model except for oxygen. The initial concentration of O\textsubscript{2} in bentonite considered in the numerical model of the potential repository is evaluated with O\textsubscript{2} in the atmosphere before the repository is closed. Granitic water is initially assumed to be equilibrated with O\textsubscript{2}(gas) in the atmosphere. The model domain includes a 2D axisymmetric domain (Fig. 2), which consists of two material zones: granite and bentonite. Solute transport takes place only by diffusion.

Pyrite and chlorite are important inorganic reductants. Their dissolution is kinetically controlled dissolution in the near field of HLW repository. Brandt et al. (2003) reported a value of steady-state dissolution rate of chlorite (at 25ºC and pH=5) of 10\textsuperscript{-13} mol/m\textsuperscript{2}s. Similar results were obtained by Gustafsson and Puigdomenech (2003) for Swedish chlorites. These authors reported dissolution rates within the range of 4x10\textsuperscript{-13} mol/m\textsuperscript{2}s for pH = 7, and 3x10\textsuperscript{-12} mol/m\textsuperscript{2}s for pH = 12. According to these values, a constant reaction rate of 10\textsuperscript{-12} mol/m\textsuperscript{2}s was adopted for chlorite. Kinetics of pyrite oxidation has been studied extensively (Evangelou et al., 1998; Williamson and Rimstidt, 1994). According to these authors, the kinetic oxidation of pyrite has been simulated as a function of pH and dissolved oxygen concentration, according to:

\[ r_m = \kappa_e C_{O2}^{0.5} C_{H^+}^{-0.11} \]  

where \( \kappa_e \) is the effective kinetic rate constant which is equal to 6.5·10\textsuperscript{-9} mol/m\textsuperscript{2}s.

<table>
<thead>
<tr>
<th>Components</th>
<th>Mineral species</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na\textsuperscript{+}, K\textsuperscript{+}, Mg\textsuperscript{2+}, Cl\textsuperscript{-}, SO\textsubscript{4}\textsuperscript{2-}, H\textsuperscript{+}, HCO\textsubscript{3}\textsuperscript{-}, H\textsubscript{2}CO\textsubscript{3}, OH\textsuperscript{-}, Si\textsubscript{4}O\textsubscript{4}\textsuperscript{4-}</td>
<td>Calcite, Siderite, Gypsum, Ca(H\textsubscript{3}SiO\textsubscript{4})\textsubscript{2}</td>
</tr>
<tr>
<td>Na\textsuperscript{+}, K\textsuperscript{+}, Li\textsuperscript{+}, Mg\textsuperscript{2+}, Mn\textsuperscript{2+}, Cl\textsuperscript{-}, Br\textsuperscript{-}, SO\textsubscript{4}\textsuperscript{2-}, H\textsuperscript{+}, HCO\textsubscript{3}\textsuperscript{-}, H\textsubscript{2}CO\textsubscript{3}, OH\textsuperscript{-}, Si\textsubscript{4}O\textsubscript{4}\textsuperscript{4-}</td>
<td>Na\textsuperscript{+}, K\textsuperscript{+}, Mg\textsuperscript{2+}, Cl\textsuperscript{-}, Br\textsuperscript{-}, OH\textsuperscript{-}, Si\textsubscript{4}O\textsubscript{4}\textsuperscript{4-}</td>
</tr>
<tr>
<td>Na\textsuperscript{+}, K\textsuperscript{+}, Mg\textsuperscript{2+}, Cl\textsuperscript{-}, SO\textsubscript{4}\textsuperscript{2-}, H\textsuperscript{+}, HCO\textsubscript{3}\textsuperscript{-}, H\textsubscript{2}CO\textsubscript{3}, OH\textsuperscript{-}, Si\textsubscript{4}O\textsubscript{4}\textsuperscript{4-}</td>
<td>Na\textsuperscript{+}, K\textsuperscript{+}, Mg\textsuperscript{2+}, Cl\textsuperscript{-}, Br\textsuperscript{-}, OH\textsuperscript{-}, Si\textsubscript{4}O\textsubscript{4}\textsuperscript{4-}</td>
</tr>
</tbody>
</table>

3.2. Results

Six cases have been analyzed numerically in order to evaluate the effects of abiotic or biotic processes in the consumption of O\textsubscript{2} trapped in the bentonite.

Case 1 is designed to simulate the evolution of O\textsubscript{2} in the bentonite considering only diffusion into the granite. Case 2 is based on case 1, but accounts for kinetically-controlled dissolution of chlorite in granite. One of the products of the dissolution of chlorite, Fe(II), is oxidized by O\textsubscript{2} in bentonite. The objective of this case is to evaluate the role of the dissolution of chlorite. Based on Case 2, Case 3 also accounts for kinetically-controlled dissolution of pyrite both in bentonite and granite. Case 4, which is based on Case 3, takes into account the two microbial processes which take place only in the bentonite. It should be noticed that the concentration of CH\textsubscript{4} is the same in both bentonite and granite in Case 4. Compared with Case 4, Case 5 assumes that there is no organic matter in the bentonite. It is worth noting that Cases 4 and 5 assume that no microbial processes in the bentonite. In Case 6 which is similar to Case 4,
it is assumed that the two microbial processes can take place in the bentonite with a concentration of organic matter equal to that of granite. This case assumes the unlikely hypothesis that microbes can survive in fully saturated compacted bentonite.

Table 3. Concentrations (M) of initial and boundary waters

<table>
<thead>
<tr>
<th>Species</th>
<th>Initial waters</th>
<th>Boundary water</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>bentonite</td>
<td>Granite</td>
</tr>
<tr>
<td>Sr</td>
<td>1.0x10^-3</td>
<td>7.92x10^-1</td>
</tr>
<tr>
<td>Mn</td>
<td>1.0x10^-3</td>
<td>7.83x10^-1</td>
</tr>
<tr>
<td>Fe</td>
<td>1.0x10^-3</td>
<td>7.83x10^-1</td>
</tr>
<tr>
<td>Ca</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>Mg</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>SO4</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>FeO</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>HCO3</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>pH</td>
<td>7.2</td>
<td>7.2</td>
</tr>
<tr>
<td>CH4</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>DOC</td>
<td>1.0x10^-3</td>
<td>1.2x10^-3</td>
</tr>
<tr>
<td>Heterotrophs</td>
<td>4.0x10^-8</td>
<td>4.0</td>
</tr>
<tr>
<td>CH4</td>
<td>1.0x10^-3</td>
<td>1.0x10^-4</td>
</tr>
<tr>
<td>Methylomonas</td>
<td>1.0x10^-3</td>
<td>1.0</td>
</tr>
</tbody>
</table>

* that component is not included in the numerical model of LOT experiment by Arcos et al., 2003.

Fig. 3 shows the results of the time evolutions of O2 in the bentonite computed in Cases 1, 2 and 3. One can see that the concentration of O2 in the bentonite decreases to 1.21x10^-5 mol/L after about 1000 years in Case 1, 558 years in Case 2 and 52 years in Case 3. Clearly, the consumption of O2 by chlorite is much smaller than that of pyrite. For the assumed kinetic rate, pyrite will deplete O2 in 52 years. A sensitive run was performed in which the kinetic rate of pyrite was 10 times smaller than that in Case 3. The time needed to consumption O2 increases to 225 years. According to Pedersen (1995), the assumption of Case 6 that microbial processes can proceed in bentonite seems, however, unrealistic.

Table 4 lists a summary of the times needed to consume O2 for each one of the 6 cases.
4. Conclusions

The results of the 6 cases evaluated allow drawing the following conclusions:

1) Dissolution of chlorite in granite is smaller than that of pyrite.

2) The effect of pyrite dissolution in the bentonite is important. The time needed to consume O2 due to the dissolution of pyrite is about 50 years for the parameters and conditions used in the model.

3) The effect of microbial processes on O2 consumption is significant. If microbial reactions can proceed in the bentonite, the time needed to consume O2 is dramatically decreased to about 2 weeks.

4) The presence of dissolved organic matter in bentonite porewater can influence O2 consumption when microbial reactions can only take place in the granite. The time to consume O2 decreases from 28.4 without organic matter in the bentonite to 4.2 years when the same concentration of CH4 is assumed in both granite and bentonite.

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