Testing Coupled Microbial and Reactive Solute Transport Models with In Situ Experiments: REX and Redox Zone Experiments at Äspö (Sweden)

J. Samper[1], J. Molinero[1], Ch. B. Yang[1] and G. Zhang[2]
[1]*Escuela de Caminos, Universidad de Coruña, Coruña, Spain, jsc@iccp.udc.es*
[2]*Earth Sciences Division. Lawrence Berkeley Nat. Lab. Berkeley, CA-94720*

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 Äspö 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 performed in fratured granites. The Äspö HRL is located in the southeast part of Sweden, 400 km south of Stockholm. The underground facility consists of a 3,600 m long tunnel which starts with an access ramp and runs into two turns down to a depth of 450 m under the Äspö island (Molinero and Samper 2003). Here we present coupled reactive and microbial transport models for the REX and Redox Zone Experiments. The REX experiment was performed at a packed section of a borehole drilled from the access tunnel and aimed at studying and evaluating the mechanisms of oxygen depletion near the access tunnel. The Redox Zone Experiment aimed at evaluating the large-scale effects of the construction of the access tunnel on the hydrochemical conditions of a fracture zone.

2. MICROBIAL PROCESSES AT THE ÄSPÖ SITE AND THE REX EXPERIMENT

Safety assessment of the Swedish concept for high-level radioactive waste disposal considers the stability of redox conditions in the engineered barriers (copper, bentonite and backfill) and the surrounding rock as a key factor for the long-term performance of the repository. If a cooper canister corrodes, radionuclides may be released. Biochemical reactions are catalyzed by microorganisms, so it is needed to define these biochemical processes in relation to electron-donor availability and microbial diversity in deep groundwaters.

The Microbe-REX Project of the Äspö site (Kotelnikova & Pedersen, 1999) investigated microbial nutrients, organism diversity, microbial activity and O_2 reduction potential. This project studied O_2 depletion processes by creating 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 re-circulating system placed in borehole KA2861A. The circulation loop had a total volume of about 1 L, and before each O_2 pulse it was filled with fresh O_2-free groundwater from the adjacent borehole KA2862A. O_2 injection pulses started by replacing part of the volume by groundwater samples that had been previously saturated with either air or $O_2(g)$ (Figure 1).

The results of this experiment indicate that groundwaters contain abundant and diverse microbial populations, including not only anaerobic microorganisms, but also facultative aerobic and microaerophilic organisms. Several microbially-catalyzed reactions may contribute to biomass production in accordance to available nutrients for biomass production.

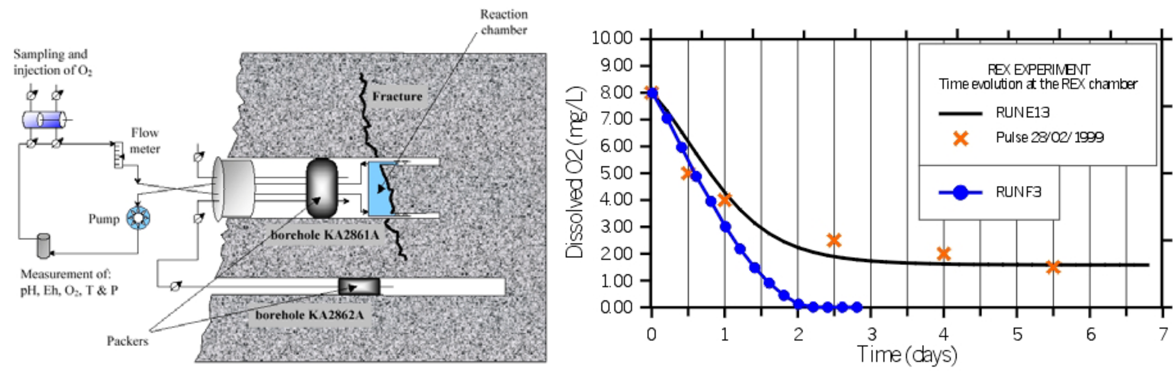

Figure 1. Setup of the REX in situ experiment (left) and time evolution of measured and computed dissolved oxygen (right). Computed values are shown for models considerin: DOC respiration (Run E13) and the combined effect of DOC respiration and methane oxidation (Run F3).

Both attached and dissolved microorganisms may be responsible for the observed oxygen depletion under in situ conditions. Although oxygen injection may inhibit iron- and sulfate-reducing bacteria, these microorganisms still appear with pronounced numbers. The percentage of iron-reducing organisms increases from 1.2-2.3% in the borehole to 0.2-55% in the REX chamber after a low oxygen pulse. The viable counts of sulfate- and iron-reducing bacteria are negatively correlated with increasing redox potential (Kotelnikova & Pedersen, 1999).

A wide variety of microorganisms, including fungi, facultatively anaerobic bacteria, and strict anaerobes are capable of reducing iron. Some of these organisms are capable of oxidizing organic compounds to CO_2, while others are incomplete oxidizers. Iron-reducing bacteria are known to catalyze the oxidation of aromatic compounds. Microbiological investigations in the Microbe-REX Project indicate that fermentation is a common microbial process taking place in the fracture zone which is carried out by anaerobic *rods* and *cocci*. Deep in the fracture zone, HS^- is unstable and is oxidized by Fe(III) or Mn(IV).

A coupled biogeochemical reactive transport model was performed for the REX experiment. The model reproduces the trends of measured oxygen consumption (Figure 1). Aerobic respiration of organic matter seems to be the main responsible process of oxygen uptake. The model indicates that abiotic consumption of dissolved oxygen in air-saturated water would take place on about 500 years. However, this time is reduced to 0.5 year when microibally-driven DOC respiration and methane oxidation are considered in the model.

3. NUMERICAL MODEL OF THE REDOX ZONE EXPERIMENT

On March 13th, 1991 the access tunnel of the Äspö HRL intersected a vertical fracture zone at a depth of 70 m below sea level. This vertical fracture zone is known within the context of the Äspö HRL as the Redox Fracture Zone because it was used to perform a long-term experiment for evaluating the effects of the construction of the access tunnel on the hydrogeological, hydrochemical and redox conditions prevailing at this fracture zone (Banwart et al., 1996). The fracture zone is approximately vertical with a thickness of 1 m. Prevailing hydraulic conditions changed drastically first when the access tunnel intersected the fracture zone and later when borehole KR0013B was open to flow. At the vicinity of the tunnel, dilution of the initially saline groundwaters by fresh recharge water was the dominant process controlling the hydrochemistry evolution during the experiment. Most chemical species showed dilution trends except for dissolved bicarbonate and sulfate which increased with time (Molinero & Samper 2003).

Molinero & Samper (2003) presented a hydrogeological groundwater flow and solute transport model of the experiment which is consistent with head and salinity data collected prior to tunnel construction and reproduced simultaneously the observed drawdowns and dilution curves induced by the construction of the tunnel. The revised hydrogeological model provided the approriate framework to perform simulations of coupled groundwater flow, reactive solute transport and microbial processes of the Redox Zone Experiment.

The reactive transport numerical model accounts for more than 60 homogeneous reactions, including aqueous complexation, acid-base, gas dissolution and redox processes, as well as 5 heterogeneous reactions including mineral dissolution / precipitation and cation exchange. A reactive solute transport model without microbial processes reproduces observed concentrations of most dissolved species both before and after tunnel construction, but fails to match measured bicarbonate and sulfate data.

The microbial model accounts for the fermentation of particulate organic carbon (POC) and the oxidation of dissolved organic carbon (DOC). POC containing sulphur compounds ferments in the shallow anaerobic zone releasing DOC and sulphur compounds. POC has a low mobility due to its large molecular weight. The low mobility of POC in the model is achieved by assigning a large distribution coefficient of 100 L/g to this species. Then, DOC is transported to deeper parts of the fracture zone and is further degraded by heterotropic bacteria which employ Fe^{3+} as electron acceptor. The model uses yeast as the microbial species representing all the fermentation-workers. POC acts as a substrate for yeast. A single-Monod kinetics is employed to describe POC fermentation. DOC and HS^- are products of POC fermentation. The microbial model assumes that SO_4^{-2} is a direct product of fermentation. As a product of fermentation, DOC is further transported into the deep parts of the fracture zone and degraded by heterotrophic bacteria such as iron-reducing bacteria. Fe^{3+} is considered to be the only electron acceptor to account for the biodegradation of DOC. Taking DOC as a substrate and Fe (III) minerals as electron acceptors, iron-reducing bacteria grow yielding HCO_3. Fe (III) minerals are abundant in the granitic fracture zones of the Äspö site, so it is assumed that the growth of the iron reducer is not limited by Fe^{3+}.

The numerical model is solved with BIO-CORE (Zhang & Samper, 2001), a general-purpose finite element solver developed at the University of A Coruña. BIO-CORE copes with both thermodinamically-controlled abiotic geochemical reactions and subsurface microbial processes in 2-D nonisothermal partly or fully saturated porous media and considers the availability of substrates for attached microorganisms by coupling a diffusion layer model to account for biofilm resistance.

Specific growth rates, half-saturation constants, yield and proportionality coefficients, and POC and DOC initial and boundary concentrations were estimated by trial-and-error matching of breakthrough curves of bicarbonate, sulfate, DOC and pH at a depth of 70 m. Computed HCO_3^- and SO_4^{2-} concentrations are extremely sensitive to small changes in microbial parameters. Calibration of microbial parameters required a lot of effort and presented numerous convergence problems. Calibrated microbial parameters are reported by Molinero et al., 2004).

Figure 2 shows the comparison of measured and computed concentrations of bicarbonate and sulfate near the tunnel location at a depth of 70 m. This figure also includes the values obtained with a reactive transport model that ignores microbial processes. Such model predicts a mild increase in bicarbonate and a decrease in sulfate. It clearly fails to reproduce the observed trend of increasing concentrations. On the other hand, the model accounting for microbial processes reproduces the observed increase in bicarbonate and sulfate during the experiment. In adition, values of DOC computed with this model are consistent with measured DOC concentrations. Computed pH values remain near neutral which are also consistent with field observations of pH remaining stable. Model sensitivity was evaluated for specific growth rates, half-saturation constants, proportionality and yield coefficients as well as initial and boundary POC and DOC concentrations (Samper et al., 2004). Results of the sensitivity analysis indicate that computed concentrations of bicarbonate and sulfate are sensitive to changes in the initial concentration of POC and the boundary concentration of DOC, but they lack sensitivity to the initial concentration of DOC and the boundary concentration of POC (Molinero et al., 2004).

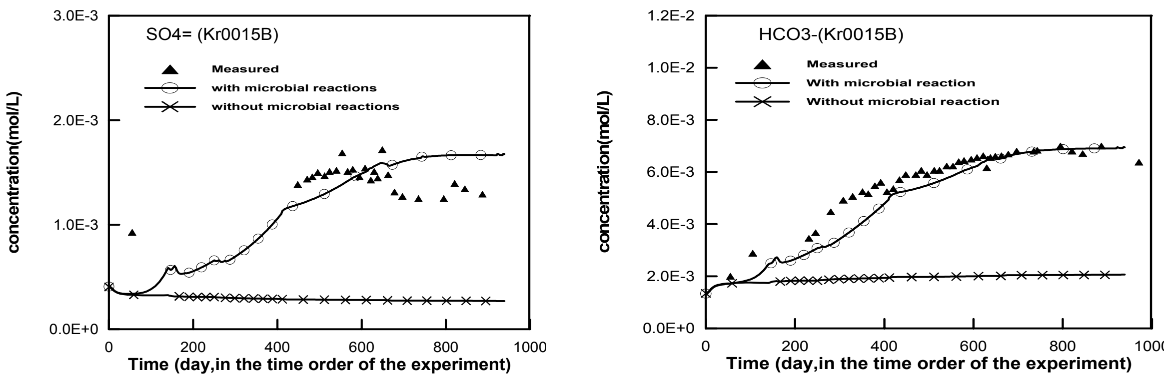

Figure 2. Comparison of measured and computed evolution of bicarbonates (left) and sulfates (right) with a model accounting for microbial processes (solid lines) and a model ignoring these processes (dashed lines).

4 CONCLUSIONS

Coupled hydrobiogeochemical models of REX and Redox Zone Experiments have been presented. The model of the Redox Zone experimen accounts for water flow, reactive solute transport, fermentation of POC by yeast and oxidation of DOC by iron-reducing bacteria. This model provides a plausible quantitatively-based explanation for the unexpected trends of bicarbonate and sulfate at the Redox Zone Experiment. Coupled hydrobiogeochemical modeling proves to be a powerful tool to improve our understanding of the hydrochemical evolution of a geological formation affected by the construction of an underground facility similar to that planned for a future deep geological repository.

5 REFERENCES

Banwart, S.; Tullborg, E.-L.; Pedersen, K.; Gustafsson, E.; Laaksoharju, M.; Nilsson, A.-C.; Wallin, B. and Wikberg, P. (1996): Organic carbon oxidation induced by large-scale shallow water intrusion into a vertical fracture zone at the Äspö Hard Rock Laboratory (Sweden). Journal of Contaminant Hydrology, 21, 115-125.

Kotelnikoba, S. & Pedersen, K. (1999): The microbe REX Project: Microbial O_2 comsumption in the Äspö tunnel. SKB Technical Report. Stockholm.

Molinero, J. & Samper, J. (2003): Modeling groundwater flow and solute transport in fracture zones: conceptual and numerical models of the redox zone experiment at Äspö (Sweden). Journal of Hydraulic Research. Special Issue for Groundwater. In press.

Molinero, J.; J. Samper, C.B. Yang & G. Zhang (2004): Biogeochemical reactive transport model of the Redox zone experiment of the Äspö hard rock laboratory (Sweden), Nuclear Tecnology (accepted, in press).

Samper, J., J. Molinero, C.B. Yang and G. Zhang, (2004) "Redox Zone II. Coupled Modeling of Groundwater Flow, Solute Transport, Chemical reactions and Microbial Proceses in the Äspö island", SKB Technical Report, Stockholm, In press.

Zhang, G. & Samper, J. (2001): BIO-CORE$^{2D©}$. A code for nonisothermal hydrobiogeochemical reactive transport. Users Manual V 0. Technical Report. University of A Coruña, Spain. 244 pp.