Isotopes and geochemistry of rock-water interaction, Chalk River, Ontario

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A five-year pre-project study under the Nuclear Legacy Liabilities Program was undertaken to assess the suitability of the bedrock at the Chalk River Laboratories (CRL) site to safely host a proposed Geologic Waste Management Facility (GWMF) for Atomic Energy of Canada Limited's low- and intermediate-level radioactive waste (LILW) at CRL. The site investigations involved the disciplines of geology, hydrogeology, geochemistry, geomechanics and microbiology.

The authors have investigated certain geochemical parameters in the groundwater (e.g. redox, pH, Fe, U, isotopic data) and the mineralogy of the host rock. The geochemical composition of the groundwater and rock pore water at repository depth may affect the performance of the repository barrier system and ideally should be favourable to retarding radionuclide movement. Characterization of the groundwater was also identified as important in determining its residence time ('age') and origin.

This talk will summarize isotopic data (^{3}H, ^{18}O, ^{13}C, ^{14}C, ^{36}Cl, ^{87}Sr/ ^{86}Sr ratio, U-series) from CRL crystalline rocks, porewaters, groundwaters and fracture minerals. The application of heavy element isotope systematics (Rb-Sr, U-Pb and U-series) was also studied in selected bulk-rock samples from CRL. Currently, information is being obtained from isotope systematics based on past fluid chemistry, isotopic composition, and pressure and temperature conditions during paleohydrogeological events (such as metamorphic and glacial episodes). Other data for examination include age dates of rocks and groundwater that may indicate recent U mobility and transitions from oxidizing to reducing groundwaters effects of rock-water interactions.

The geochemical composition of the groundwater and the rock pore water is consistent with low hydraulic conductivity of rocks. It further indicates limited fracture-matrix interaction and implies that solute movement in the rock matrix in this area is diffusion controlled. The Rb-Sr & U-Pb isotope systematics indicate no geologically recent U mobility (<100 Ma) and most U isotopic data show secular equilibrium, and, therefore, no U loss/gain in the last 1 Ma.

Ureolytic CaCO$_3$ Precipitation in the Presence of Non-ureolytic Bacteria

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Microbially induced CaCO$_3$ precipitation (MICP) is recently being studied due to its various potential applications, such as: sequestration of soil contaminants [1]; and mitigation of seismic liquefaction [2], among others. Some of these applications involve *in-situ* treatment of soils, and therefore require a better understanding of the interactions between microbial ecology and aquatic geochemistry.

Hydrolysis of urea, catalyzed by the enzyme *urease*, is one of the most common pathways for MICP [3]. Hydrolysis of urea produces ammonium and carbonate: $CO(NH_2)_2 + 2H_2O \rightarrow 2NH_4^+ + CO_3^{2-}$, thus increasing surrounding alkalinity. In the presence of dissolved calcium this process will result in the precipitation of CaCO$_3$: $Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3$.

In order to study the interactions between ureolytic and non-ureolytic bacteria we performed a MICP experiment in artificial groundwater medium, inoculated with two model bacteria: *S. pasteurii* (ureolytic bacteria) and *B. subtilis* (non-ureolytic bacteria), and supplemented with Nutrient Broth and urea. Control was inoculated with *S. pasteurii* alone. The experiment lasted 10 days, during which NH$_4^+$, Ca^{2+}, pH, dissolved inorganic carbon and optical density (OD) were measured.

Results are summarized in Table 1. Most of the CaCO$_3$ precipitation took place until the 80th hour of the experiment. During which time, pH values and carbonate ion concentrations in the presence of non-ureolytic bacteria were lower than in their absence. However, measured Ca^{2+} concentrations indicates that CaCO$_3$ precipitation was accelerated in the presence of non-ureolytic bacteria, despite less favorable chemical conditions. Bacterial growth, as infferred from OD measurements was significantly higher in samples containing non-ureolytic bacteria. Therefore, we suggest that CaCO$_3$ precipitation in the presence of non-urealytic bacteria was accelerated due to addition of nucleation sites in the form of bacterial cells.

	Control	Both Bacteria
pH	7.49-8.45	7.50-8.21
CO$_3^{2-}$ (mM)	0.005-0.132	0.005-0.087
OD	20 hr. lag phase	6 hr. lag phase
CaCO$_3$ precipitation	100% in 123 hours	100% in 80 hours

Table 1: Results summary.

[1] Fujita, Taylor, Gresham, Delwiche, Colwell, McLing, Petzke, Smith, (2008) *Enviro. Sci. Technol.* **42**, 3025-3032.
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