

Whiteshell underground laboratory (Canada)

Description: The Lac du Bonnet granite batholith, dated to 2665 ± 20 Ma, forms part of the Archean Superior Structural Province of the Canadian Shield. The granite batholith, which is elongate in shape, intrudes a suite of earlier gneisses and metavolcanic rocks to the east and is overlain by Phanerozoic sedimentary rocks to the west (Brown et al., 1989). During emplacement, chemical differentiation of the magma led to an increase in both $f(\text{O}_2)$ and $f(\text{H}_2\text{O})$, resulting in transgression of the magnetite/haematite boundary within the rock body. Consequently, the upper, altered and pink (haematite-bearing) granite extends to 200-250 m, and the lower, unaltered and medium-grained grey (magnetite-bearing) granite extends to at least 1000 m depth.

A section of the batholith, the Whiteshell Research Area, has been studied extensively as part of the Canadian Nuclear Fuel Waste Programme, which was initiated in 1978. The Research Area hosts the Whiteshell Underground Research Laboratory (URL). Numerous boreholes have been drilled to depths of up to 1000 m. Sub-vertical fractures and at least three low-intermediate dip fracture zones have been identified at the investigation site. In terms of fracture mineralogy, early granite, aplite and pegmatite dykes are cut by late chlorite-filled fractures formed under lower greenschist facies at 100-200 MPa and at 250-350°C. The youngest low temperature fracture fillings are carbonates, iron oxides and clays (Stone et al., 1989).

The sub-vertical fractures form the main conduits for groundwater infiltration, at least to around 400 m. This comprises shallow (<100 m), low saline groundwaters (Ca-Na-HCO₃ type), intermediate (100-300 m) more saline groundwaters (Ca-Cl-SO₄ type), and deeper saline brines (Ca-Na-Cl-SO₄ type).

Studies, including those of the behaviour of U, Th and Ra isotopes and REEs, have been taking place since the late 1970s to the early 1990s. The objectives of these mainly U-decay series isotopic studies included:

- the general determination of actinide and daughter element mobilities in rock/water systems and estimation of the time interval during which mobilisation occurs (Gascoyne, 1985);
- combined measurements on rock and groundwater samples as a useful indicator of recent actinide migration patterns and processes in igneous rocks (Gascoyne and Cramer, 1987);
- the use of U-decay series disequilibria as indicators of geochemical disturbance on the granite (Ivanovich et al., 1987);
- the application of U-decay series measurements to understand the ability of the rock mass to retard radionuclide migration and examination of the properties of natural water-conducting fractures that might influence radionuclide migration rates (Griffault et al., 1993); and
- the determination of U, Ra (as ²²⁶Ra) and Rn (as ²²²Rn) concentrations in groundwaters to provide evidence for the mobility or retardation of these radionuclides based on concentration profiles (Gascoyne and Barber, 1992).

Relevance: The study is relevant to the understanding of far-field radionuclide mobility and retardation processes in repositories where a crystalline host rock is envisaged.

Position(s) in the matrix tables: This study falls within the long term radionuclide transport and retardation mechanism box in the far-field matrix table.

Limitations: This study is a reasonably good analogue for to the mobility and retardation processes expected to occur in the far-field in the vicinity of a leaking radionuclide source at depth under ambient temperatures. The limitations are largely logistical, i.e. sampling is restricted to only a few boreholes, so that the vertical and lateral isotopic and elemental spatial variation at a site scale can not be evaluated with certainty. Furthermore, although present-day hydraulic and hydrochemical boundary conditions can be estimated, little is known about past prevailing boundary conditions which have resulted in many of the elemental and isotopic changes that have been observed.

Quantitative information: The early work of Gascoyne (1985) demonstrated that the relatively unaltered Lac du Bonnet granite is in secular equilibrium, i.e. no rock/water interaction has influenced this rock mass for at least 2 Ma. The later work of Gascoyne and Cramer (1987) indicated a multi-stage history of alteration and concomitant element migration for the fractures in the granite. Adjacent to the fracture zones in the host granite, ancient hydrothermal interactive processes had resulted in an enrichment of Fe (as haematite) and REEs, countered by a loss of Na, Ca and U. Subsequent low temperature groundwater interaction at low Eh along these fractures has removed the haematite. Later, more oxidising groundwater incursions served to mobilise the uranium, resulting in isotopic disequilibrium, at least down to 260 m depth. The coincidence of this U-disequilibrium with REE enrichment at the fracture surfaces suggests that REE mobilisation, in common with that of uranium, is a recent phenomenon, probably within the last 100 000 years.

The study also indicated the pervasive leaching of uranium (up to 80%) in the rock mass at surface and shallow depths. The work of Ivanovich et al. (1987) also recognised several periods of geochemical alteration and confirmed that the latest period has been operating over the last million years as reflected by isotopic disequilibrium of the U, Th, Pa and Ra daughter radionuclides with their respective parents. Also, the main locations for the rock/water interaction and exchange processes are the fracture surfaces.

Griffault et al. (1993) focussed on two deep fracture-zone sites at around 1175 m depth. These were characterised mineralogically, chemically and isotopically; hydraulic and hydrochemical data were also available. In addition to confirming and quantifying the parageneses of the different generations of fracture-filling mineral phases indicated by earlier studies, detailed rock profiles were also selected, perpendicular through the fracture margins out into the surrounding rock matrix, and analysed. These clearly linked the mineralogical, chemical and isotopic variations to mobilisation and migration processes between the fracture and the host rock matrix.

Of importance was evidence of isotopic disequilibria within both fracture zones at these depths, one characterised mainly by a haematite coating and the other brecciated, with illite as an alteration phase at the fracture margin, formed by interaction of saline groundwater with the adjacent granite. These disequilibria are consistent with a model involving loss of ^{234}U , ^{230}Th and ^{226}Ra to groundwater by alpha-recoil from U deposited on the illite clay surfaces, and not from any oxidative bulk leaching. These radionuclides were subsequently deposited in the nearby brecciated zone.

Uncertainties: On a scale of low-medium-high, the uncertainties associated with this semi-quantitative study are assessed as medium; the sources of these uncertainties have been referred to in the above section on 'Limitations'.

Time-scale: The time-scale addressed by the study is geological, both Quaternary to recent (<2 Ma) and beyond (>2Ma).

PA/safety case applications: Reference is made in the *AECL EIS (Environmental Impact Study), 1994 (Canada)* to radionuclide retardation, including matrix diffusion bounding values.

Communication applications: There are no examples of its use in communication and dialogue material.

References:

Brown, A., Soonawala, N.M., Everitt, R.A. and Kamineni, C., 1989; Geology and geophysics of the Underground Research Laboratory site, Lac du Bonnet Batholith, Manitoba. *Can. J. Earth Sci.*, 26, 404-425.

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Ivanovich, M., Longworth, G., Wilkins, M.A. and Hasler, S.E., 1987; A study of uranium series disequilibrium in core profiles and mineral separates from the samples of Lac du Bonnet Granite from the URL Site, Pinawa, Manitoba, Canada. UK DOE Rep. No. DOE/RW/87.077.

Stone, D., Kamineni, C., Brown, A. and Everitt, R., 1989; A comparison of fracture styles in two granitic bodies of the Superior Province. Can. J. Earth Sci., 26, 387-403.

Added value comments: There are many isotopic data from detailed profile studies from fracture systems out into the adjacent host rock. These could have been discussed more critically in the context of possible evidence (or otherwise) of matrix diffusion mechanisms.

Generally, however, much of the data produced from the various studies are most useful in confidence building (with respect to certain FEPs) and also in scenario development. In addition, the conclusions to the various studies could be used more in communication.

Potential follow-up work: Much excellent data have been produced over the years in the Whiteshell area and in the URL facility itself. Some of these data have been interpreted and published in the open literature, but much more exists in AECL reports, some restricted. A reappraisal (and possible reinterpretation in some cases) of what has been carried out over the years could provide a very useful basis to identify areas where, for example, improvements (e.g. in sampling/analysis) can be made and where recent advances in palaeohydrogeochemical interpretation could be applied.

Keywords: Radionuclides, mobility, retardation, U-decay series.

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