Cigar Lake (Canada)

Description: Cigar Lake is one of the large unconformity-type uranium deposits of Proterozoic age which characterise the Athabasca Basin uranium province in northern Saskatchewan, Canada. It is notable from the other deposits in the region in that it is located entirely below the surface at approx. 450 m depth. There is no surface expression of its existence and it was discovered by a systematic drilling campaign through promising geological strata (Bruneton, 1987). Natural analogue studies were initiated by Atomic Energy of Canada Limited (AECL) in 1892 and continued through various phases until 1993.

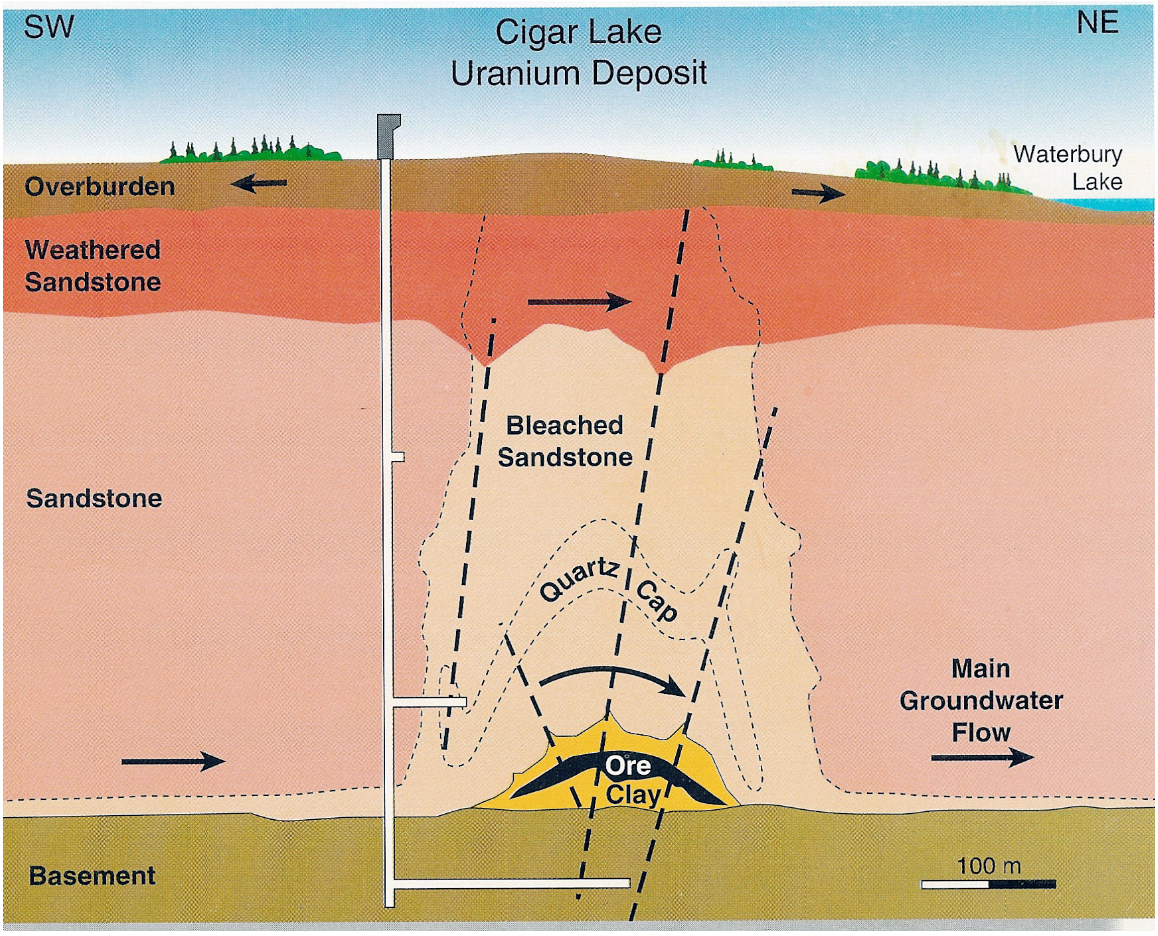

Figure 1: Schematic cross section through the Cigar Lake uranium deposit showing major lithological types, extent of the hydrothermal halo and groundwater flow pathways (after Cramer and Smellie, 1994).

The deposit is hydrothermal in origin and consists of an irregularly-shaped lens about 2150 m long, 25 to 105 m wide and 1 to 20 m in thickness. The average grade of the ore is around 12 wt% uranium with maximum concentrations up to 55 wt%. The ore reserve has been calculated to contain 1.5×10^5 Mg U. The hydrothermal fluids which were responsible for the alteration of the basement and the sandstone host rocks, thereby producing the uranium mineralisation, were reducing when discharged from the graphite-rich basement metasediments into the sandstone along regional fractures. These fluids interacted with downward percolating, oxidising, uranium-rich meteoric groundwaters in the sandstone, resulting in the mass precipitation of uraninite and pitchblende close to the unconformable contact with the Archean basement. Alteration of the host sandstone involved the dissolution of haematite (colour change from red to white) and also quartz, resulting in clay (illite) formation which increases markedly towards the ore zone. Reprecipitation of the dissolved quartz forms an arch-like zone, or cap rock, higher up in the sandstone formation (Fig. A). In addition to surrounding the ore body, the formation of the clay (5-30 m in thickness) has also effectively sealed the pre-existing fractures in the underlying basement and overlying sandstone which facilitated the passage of the hydrothermal solutions. The continued presence of illite indicates an absence of water-rock interaction since its formation.

The ore at Cigar Lake is typically polymetallic in type, the uranium (mainly as uraninite, pitchblende and coffinite) being accompanied by a broad suite of elements including Ni, Co, Mo, Pb (radiogenic), Zn, Mn, Fe and V. The presence of iron and magnesium carbonate (siderite and dolomite) is also characteristic. The ore body itself shows no mineralogical evidence of having been subject to major uranium dissolution or remobilisation processes since emplacement.

Hydrogeologically, three major sub-horizontal flow regimes (local, intermediate and deep) have been recognised in the overlying sandstone host rocks; some hydraulic interconnection occurs between each regime by means of minor fracture percolation and flow along major conducting sub-vertical fracture zones (Cramer and Smellie (Eds.),1994). The clay-rich rocks of the basement and surrounding the ore body direct the main volume of this groundwater through the basal sandstone and over and around the mineralised clay zone. Other than by diffusion, groundwater only penetrates the ore body via small fractures through the surrounding clay-rich rock.

Very low particle velocities were predicted in and around the deposit and the underlying basement, compared with the surrounding rock units; in fact, the permeability of the clay-rich zone is 1000 times lower than that of the overlying sandstone. This is reflected in the calculated residence times in the ore/clay zone for particles released within the ore; these vary between 18 000 and 85 000 years. and correspond closely with groundwater residence times calculated from ^{36}Cl data.

From the subsurface, to the vicinity of the deposit, the groundwaters show an increase in pH (5.6 to 7.8), increasing TDS (60 to 240 mg/L) and increased negative measured redox potential (+250 to -250 mV). Marked increases in ^{3}H, ^{14}C and ^{36}Cl also occur as the deposit is approached; this is a result of the production *in situ* of nuclear reaction products caused by neutron capture from the ore.

From its initiation in 1982, the Cigar Lake Study has addressed two major issues of importance to HLW repository performance assessment (PA): 1) the stability of the uranium ore body (as an analogue of spent fuel) since its formation over one billion years ago, and 2) the role of the surrounding clay zone (as an analogue of bentonite buffer material and/or a clay-based repository host rock) in helping to preserve the ore body, both chemically and hydraulically, over this timescale (Cramer, 1986; Cramer et al., 1987; Goodwin et al. (1989).

Later international collaboration (1989-1993), together with SKB (Sweden), and to a lesser extent Los Alamos National Laboratory (LANL, USA), widened the scope to include the role of colloids, organics and microbes in the vicinity of the ore body. PA-related models related to, for example, near-field (e.g. radiolysis) and far-field (e.g. mass transport) uranium mobility, were also tested, as were equilibrium thermodynamic codes and databases. These latter were the so-called ‘Blind Predictions’ (Vilks et al., 1991; Cramer and Smellie (Eds.), 1994; Karlsson et al., 1994; Liu and Neretnieks, 1996; Pericival et al., 1996; Smellie and Karlsson, 1996).

The main conclusions reached in the Cigar Lake Study were:

<u>Radiolysis</u>

PA models developed to calculate oxidation by water radiolysis were found to be realistic to conservative, and were considered to represent a step forward relative to earlier calculations. Radiolysis was demonstrated to be low at Cigar Lake, which means that the groundwater conditions could remain reducing as, in simple terms, the produced oxidants are being consumed close to the ore itself. Drawing on an analogy with exposed spent fuel, a similar situation would prevent the build-up of a redox front and probably simplify the chemical processes to be considered for radionuclide release.

Role of colloids, organics and microbes

Suspended particle concentrations, present in very small amounts, reflected the local contact rock rather than the geological formation and were found to be fairly uniform throughout the deposit. When recorded, the particles (>50 nm) consisted primarily of clay minerals (illite, chlorite and kaolinite), amorphous Fe-Si precipitates, quartz and organic material. The uranium concentrations of particles down-flow from the deposit were no greater than those in the surrounding sandstones, showing that there is no 'uranium plume' emanating from the ore body, i.e. the clay halo provides an efficient filter to colloid transport.

The study of organic substances in the Cigar Lake groundwaters, abstracted from the general sedimentary host rocks, showed 15-25% humics (mostly low molecular weight fulvic acid); this contrasted with those from the the ore body which recorded <2% humics. The extracted humic fraction from the ore body was dated as >15 000 years (by ^{14}C).

Although microbe studies were limited, several bacteria were identified in the vicinity of the ore body; those confirmed include sulphate-reducing, denitrifying, Fe-related and possibly methanogenic varieties.

Near- to far-field mass transport

Because of the generally reducing groundwater conditions at depth, the shielding effect of the low permeability clay zone, and its redox buffering capacity (pyrite and/or marcasite), the uranium-bearing minerals in the ore body appear to have remained stable since their formation and uranium mobilisation and transport have been insignificant. Consequently, it was decided to develop a series of iterative, generic models for potential mass transport scenarios of relevance for PA. The basic assumptions made were that due to a chemical concentration gradient, radionuclide migration would take place from the ore body, through the clay zone and out into the altered sandstone aquifer above the ore body. The models considered a range of scenarios covering the source term and transport processes, including solubility-controlled and oxidative dissolution of UO_2, diffusion and advection, and coupling mass transport and geochemical processes. The modelling showed that:

- the extent of a uranium plume breaking through the clay/altered sandstone interface would not exceed 0.5 m into the altered sandstone
- the 1-D modelling results are in good agreement with most measured parameters
- at the dissolved O_2 levels measured in the ore body waters, the modelling predicts that oxidation of the clay/ore contact over a 1 m distance is reasonable
- the low permeability clay zone surrounding the mineralisation is the most important parameter in limiting radionuclide mass transport

Such conclusions provide added confidence in repository design features using bentonite buffer material, in concepts planning to utilise a clay-based host rock, and in the ability to successfully model the system.

Testing of equilibrium thermodynamic codes and databases

There are two main uncertainties in the use of thermodynamic codes and databases in PA: a) the thermodynamic equilibrium constants required in the calculations are poorly defined, and in some cases missing, and b) the models assume that the groundwater chemical system is at equilibrium. The former can lead to an overestimation of trace element solubilities. Modelling of the Cigar Lake groundwaters indicated that the solubility tests increased confidence in the thermodynamic modelling of radionuclide solubilities and highlighted inadequacies in the current thermodynamic databases. The need for better mineralogical identification of the solid phases that actually control trace element solubility was also underlined.

Relevance: The Cigar Lake Study was focussed mainly on the radioactive waste disposal concepts in Canada and Sweden and their shared goal of dealing with spent nuclear fuel. Much of what has been learnt, however, has applications to other national disposal concepts, in particular the considerable amount of work carried out on the massive clay zone surrounding the ore body. This demonstrates its long-term stability and has far-reaching implications to clay-based repository host rock environments presently being considered by, for example, Switzerland, France, Spain, Belgium and Japan.

Position(s) in the matrix tables: The Cigar Lake Study is relevant to both the near-field (spent fuel stability; radiolysis and radionuclide mobility/transport; bentonite/clay stability) and far-field (uranium mass transport; role of colloids, microbes and organics) matrix tables. Common to both is the testing of equilibrium thermodynamic codes and databases.

Limitations: The Cigar Lake host rock sandstones are highly porous, saturated and hydraulically transmissive and would be considered unsuitable for a HLW repository. In addition, although naturally occurring uraninite is closely analogous to spent nuclear fuel, both being dominantly UO_2 in composition, uraninite does not contain any concentrations of fission products and the maximum temperatures reached in a modern reactor far exceed those experienced by the uraninite at Cigar Lake. Also, uraninite may contain substantial amounts of radiogenic lead not present in spent nuclear fuel. Furthermore, whilst the illite clay zone enveloping most of the ore body may be somewhat analogous to a clay-based repository host rock, it differs significantly from the preferred near-field bentonite (i.e. montmorillonite) barrier materials. Whilst the relevance of these dissimilarities are critical if close scientific analogies are to made, there is no denial that despite them the Cigar Lake ore body has survived virtually intact since its formation 1.3 Ga ago. This is a strong argument in support of present disposal concepts which would be expected to be even more robust over suitable geological timescales.

Quantitative information: Most information which has resulted from this study has been semi-quantitative or qualitative and very useful in respect to 'confidence building' in several FEPs. One exception has been the issue of radiolysis and spent fuel dissolution where considerable progress was made in testing and developing current radiolysis models used in PA methodology. Furthermore, support from laboratory experiments studies also showed that radiolysis is, in any case, not such an important process as initially predicted. This, plus the fact that the SKB (and Posiva) disposal concept now includes an inner steel liner to the copper canister, thus effectively buffering any radiolysis effects *in-situ*, means that radiolysis is no longer an issue of concern and this study is a good example of where analogue studies have been used to exclude a process previously thought important.

Uncertainties: On a scale of low-medium-high, the uncertainties in both the semi-quantitative and qualitative information are assessed as being medium to high, because of the difficulties in establishing boundary conditions to the various processes studied.

Time-scale: The time-scales addressed by the study are geological (Quaternary <2 Ma and >2Ma).

PA/safety case applications: Examples of their use in published PA include:

- SKB-91: Demonstrated conservatism in estimating radiolytic oxidation
- TILA-99: Support for conservatism in assumptions regarding spent fuel dissolution rate
- SR-97: Support for insignificant colloid concentrations at repository depths

With respect to PA-related modelling:

- SKB-91: Radionuclide solubility model testing (BPM)
- TVO: Testing of UO_2 spent fuel dissolution models
- AECL EIS: Radionuclide solubility model testing

Testing models and databases for colloid formation and organic complexation

- SR-97 Justification of model for radiolytic oxidation of UO_2
  Reference to matrix diffusion data for model testing

Communication applications: Previous uses of the analogue in communication and dialogue material for different audiences include official SKB and AECL brochures, the SKB mobile exhibition and its inclusion in the CEC-coordinated natural analogue video ('Traces of the Future').

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Added value comments: The Cigar Lake Study profited greatly from a concerted international effort representing different types of disposal ideas and concepts and scientific experience. Of prime importance was the derivation of a good conceptualisation of the site based on a rigorous geological, hydrogeological and hydrogeochemical characterisation and high quality data. This allowed the development of potential PA-related scenarios which led to the testing of mathematical mass transport models and geochemical thermodynamic databases and codes. Moreover, since so many integrated processes were being studied, the Cigar Lake site allowed an effective FEP screening process to be carried out. Finally, in terms of communication, Cigar Lake has proved to be one of the most successful analogues for presenting to the general public the potential robustness of an eventual repository system over long periods of geological time.

Potential follow-up work: With respect to further field activities, it is not known to what extent open gallery conditions have resulted in disturbance of the groundwater flow and hydrochemical systems, and whether widespread grouting has caused significant contamination in the vicinity of the project boreholes. Further studies of the ore body and surrounding clay halo from underground galleries would be of interest, to provide new data and/or confirm data already obtained, in order to complete the picture (e.g. is there a location where there is no clay protection, and what does the mineralogy look like?). I*n-situ* trace element speciation is still an open question; blind prediction modelling at Cigar Lake could benefit greatly from such measurements. Also, can coffinite formation from uraninite alteration be a low temperature (<100^{o}C) process in nature under reducing conditions? If so, this could be important in quantifying long-term spent fuel stability.

Otherwise, new and alternative modelling approaches could be tried out using the available database and the site could be more carefully evaluated with respect to the Glacial Scenario (e.g. depth of permafrost; penetration of oxidising(?) glacial melt waters etc.).

Keywords: Uranium mineralisation, uraninite, radiolysis, radionuclides, colloids, model testing.

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