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SEALING MATERIALS PROPOSED FOR USE IN A DEEP GEOLOGIC REPOSITORY FOR USED NUCLEAR FUEL: TESTS CONDUCTED USING BENTONITE-BASED MATERIALS AT AECL'S URL (1988-2004)

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ABSTRACT

The development of a robust concept for deep geologic isolation of used nuclear fuel includes the selection and application of sealing systems that will ensure that the containers holding the used fuel are effectively isolated from the biosphere. The emplacement-room sealing system is required to provide a number of isolating functions as well as a means of transmitting the heat generated by the used fuel to the surrounding geosphere. While laboratory simulations to assess sealing system performance provide a starting point, ultimately their performance needs to be demonstrated and assessed at, or near full-scale and in a real geologic environment. Such large-scale simulations also provide a means of testing the capabilities of numerical simulation tools being developed for use in predicting longer-term repository performance.

Atomic Energy of Canada Limited (AECL) constructed and operated an Underground Research Laboratory (URL) in a previously undisturbed granitic rock mass near its Whiteshell Laboratories in Eastern Manitoba between 1983 and 2005. During the Operating Phase of the URL, a wide range of valuable experiments and demonstrations were conducted on two main testing levels (240-m and 420-m depth), to simulate the performance of sealing materials for potential application in a deep geologic repository. These included the following in situ engineered barriers simulations.

- The Buffer/Container Experiment (1990-1993)
- The Isothermal Test (1992-1996)
- The Tunnel Sealing Experiment (1998-2005)
- The Composite Seal Experiment (2001-2005)

Each of these studies is briefly described and the key results are presented in this paper.

I. INTRODUCTION

Canadian concepts for disposal of used nuclear fuel in a deep geologic repository[1, 2] have consistently included compacted bentonite-based materials to surround the long-lived, corrosion resistant containers as shown in Figure 1 for the emplacement geometries currently under consideration.

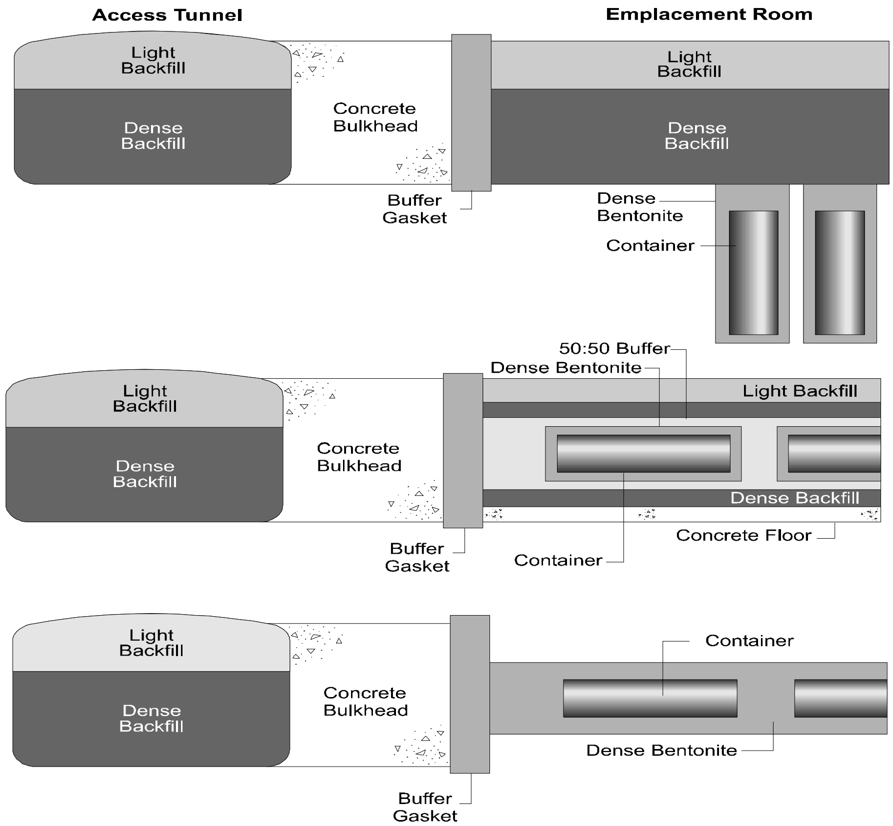

Figure 1: Generic emplacement geometries[2].

The development of a robust concept for deep geologic isolation of used nuclear fuel needs to include a sealing system that effectively isolates the containers holding the used fuel. The materials surrounding the emplacement containers should have low hydraulic conductivity, the ability to swell to fill any gaps or voids when water is available and effectively conduct heat away from the containers. Development of suitable materials has occurred over 25 years of study that has included investigations of thermal, hydraulic, mechanical, chemical and microbial parameters as well as engineering-scale simulations to evaluate how these parameters interact to control sealing system performance. Of the range of materials examined for these purposes, smectite clays (commonly referred to as bentonite) were determined to have the best potential for repository application. Smectite clay and mixtures of smectite clay and other components such as sand or crushed rock are referred to as dense bentonite, buffer, dense backfill, light backfill or gap fill.

II. SEALING EXPERIMENTS IN THE CANADIAN URL

While laboratory simulations to assess sealing system performance provide a good starting point, ultimately their performance needs to be demonstrated and assessed in a real geologic environment at, or near, full-scale. AECL operated an Underground Research Laboratory (URL) near Lac du Bonnet, Manitoba from the early 1980’s until 2005. Various aspects of constructing and operating a deep geologic repository in the granitic rocks of the Canadian Shield and the application of sealing technologies in that environment. Within the URL environment, natural processes that could not be simulated (or were not previously recognized) can progress and their influence(s) can be assessed. Experiments performed prior to 1997 were primarily focussed on demonstrating the safety and feasibility of AECL's deep geologic disposal concept. After 1997 work was directed towards addressing gaps identified in technologies required to licence and construct a deep geologic repository. Much of the work performed at the URL between 1997 and the announced closure of the URL in 2003 was conducted as part of Ontario Power Generation's (OPG)'s Deep Geologic Repository Technology Program, which addressed issues in geologic characterization, safety assessment and repository engineering.[3]

There were four major underground experiments at the URL that focused on sealing materials proposed for use in a repository. These were: the Buffer/Container Experiment (BCE), (1990-1993)[4, 5]; the Isothermal Test (ITT), (1992-1996)[5, 6]; the Tunnel Sealing Experiment (TSX), (1998-2005)[7, 8, 9, 10] and the Composite Seal Experiment (CSE), (2001-2005).[3] The locations of these experiments within the URL are shown in Figure 2.

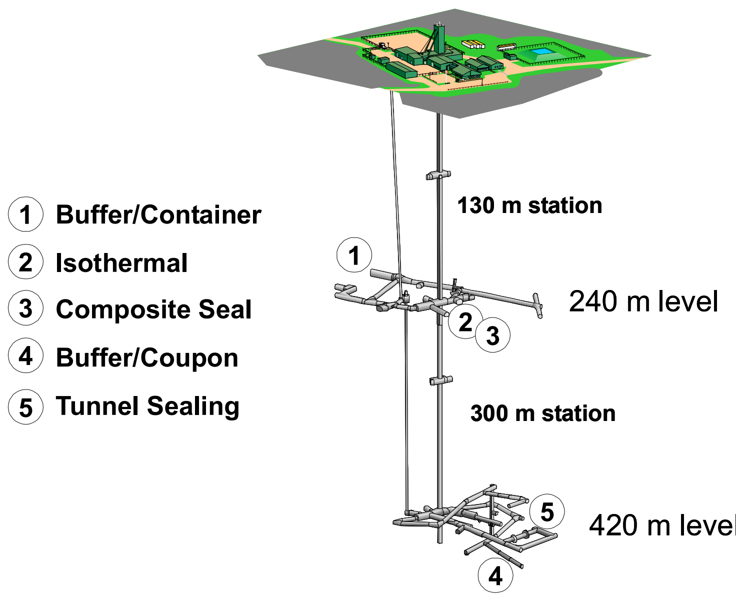

Figure 2: Location of Sealing Experiments at the URL.

The Buffer/Container Experiment (BCE) and Isothermal Test (ITT)

The BCE and ITT were the first and second large-scale experiments utilizing sealing materials conducted at the URL. These experiments are summarized in a number of papers.[3, 4, 5, 6]

The BCE and the ITT had many common goals but focussed on different details of sealing material evolution and performance.[5] The common goals of these experiments included measurement and examination of:

1. rate of water uptake from the rock and its distribution in compacted sand-bentonite materials,
2. swelling and self-healing characteristics of the buffer (a compacted blend of equal proportions of sand and bentonite clay),
3. swelling and swelling-induced pressures within the buffer mass and on the surrounding rock and concrete,
4. rates of change of pore water pressures in the buffer and the surrounding rock,
5. chemical changes as the result of thermal and hydraulic processes, and

6. possible buffer alterations and/or cementation due to heating or interaction with cementitious materials, and their effects on the buffer.

The BCE, which operated under heat for 2.5 years, was a full-scale simulation of a single emplacement borehole in the in-floor emplacement configuration. It was primarily intended to examine water movement around a heat-generating container in a natural geologic environment.[4, 5] The fuel waste container was simulated by a 2-m-high by 0.6-m-diameter electric heater that operated at a temperature of 85°C on the heater surface for 29 months. The heater was placed within a 5-m deep by 1.24-m diameter borehole and surrounded by compacted sand-bentonite buffer material (Figure 3). Early in the conduct of the BCE it was recognized that analysing the coupled thermal and hydraulic processes would be difficult unless it were possible to separate their effects. It was therefore decided to install a second mass of buffer in a similar borehole without a heater. This second experiment, called the ITT (Figure 3), operated for 6.5 years and represented a simpler test without thermal influences on saturation.[5, 6]

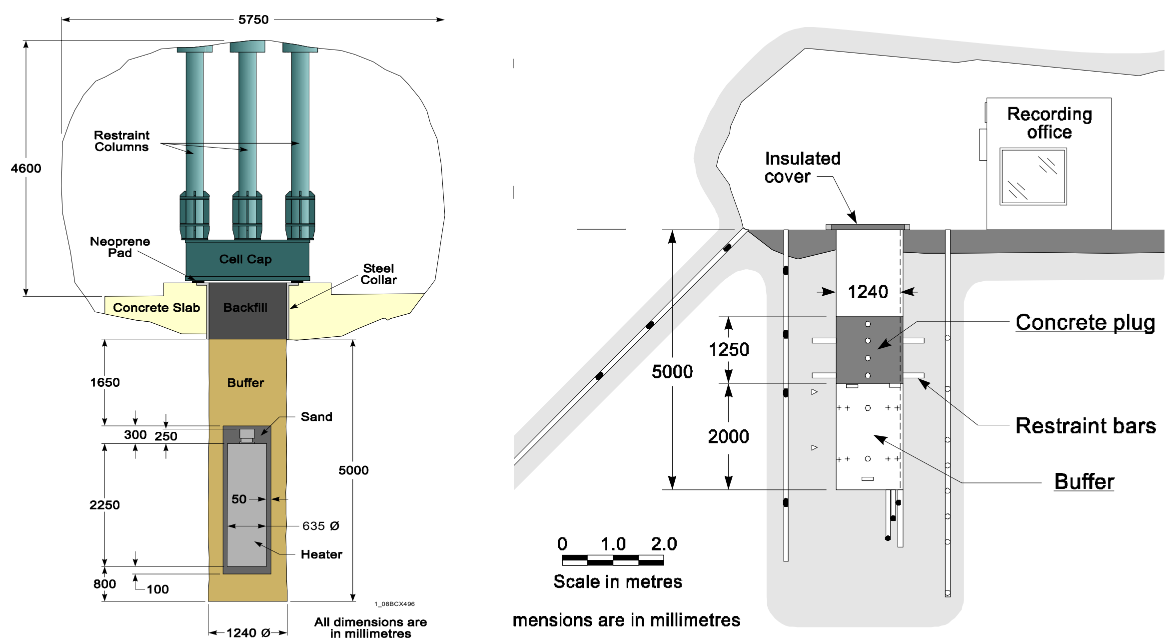

Figure 3: General layout of the BCE (left) the ITT (right)[4, 5].

Figure 4 shows the strong effect the heater had on water content distribution. Areas close to the heater had moisture driven outwards towards the edge of the borehole. As thermally-induced water movement was occurring, water continued to seep in from the rock. The combination of seepage from the rock and thermal drying near the heater created a strong water content gradient in the annulus of buffer between the heater and the surrounding rock mass. For comparison, the water content distribution observed at the time of ITT decommissioning is also shown in Figure 4. In the BCE, the buffer immediately adjacent to the rock surface and portions of the buffer below the heater exhibited water contents greater than that needed to achieve full saturation (20.8 %) of the as-placed buffer. This can only occur if the material has expanded, thereby increasing porosity. Since the buffer was constrained both vertically and radially, expansion must have been the result of drying and shrinkage of material adjacent to the heater.[4] Visual examination of the buffer prior to excavation identified cracks within the buffer that extended radially away from the heater, indicative of drying shrinkage. These cracks did not extend into the region of saturated buffer nearer the rock. On saturation in subsequent laboratory tests, these cracked samples of buffer exhibited the same low hydraulic conductivity as those that had never experienced thermal drying, illustrating buffer's self-sealing capability.

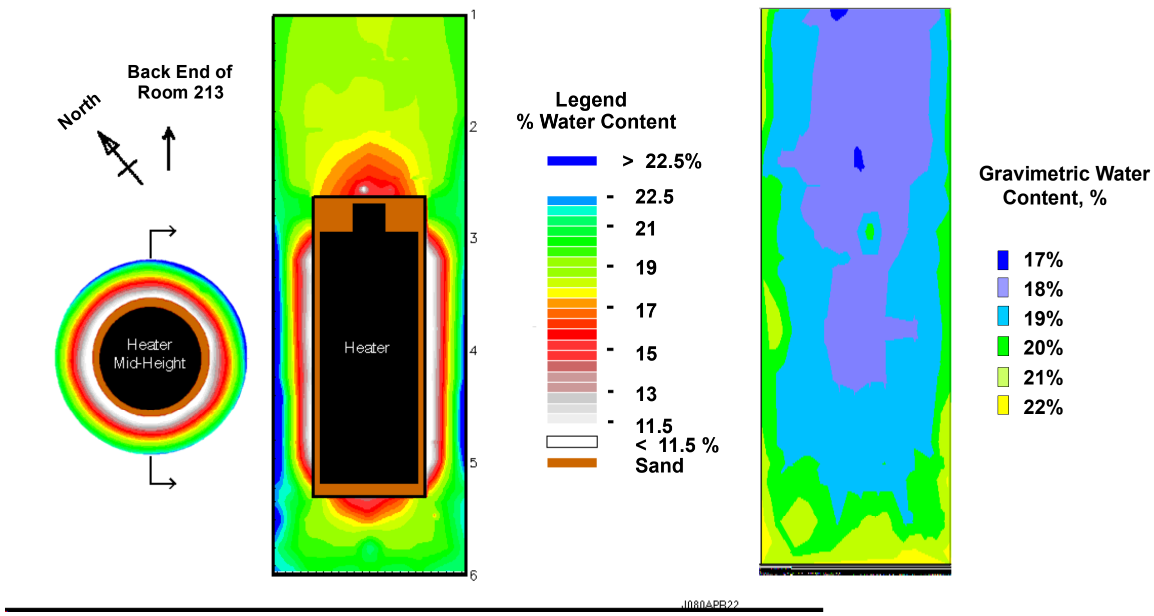

Figure 4: End-of-test water distribution within the BCE (left) and ITT (right).[4, 6]

The ITT examined moisture transfer from the surrounding rock to the buffer. The water content of the buffer at the time of decommissioning is presented in Figure 4. The highest water content occurred at the outer perimeter and decreasing to values close to the as-placed water content in the centre. The lower region of the borehole had a slightly reduced average density at the time of decommissioning, as indicated by the gravimetric water content contours above 20% (Figure 4). As with the BCE, 20.8% gravimetric water content represented full saturation, based on as-placed densities. Approximately 80 litres of groundwater seeped into buffer during the 6.5 years of ITT operation, resulting in a saturated outer skin and an inner core that remained essentially at its as-placed moisture content. In performing a numerical back-analysis of the ITT, a nearly impermeable hydraulic conductivity had to be assigned to the outer saturated skin of buffer in order for the model results to match the end-of-test data. This low permeability is believed to be a consequence of the constraint caused by both the *in situ* compaction method and the rigidity of the borehole rock boundary.

The BCE and ITT monitored the physical, thermal and hydraulic changes in and around emplacement borehole simulations for approximately 2.5 and 6.5 years respectively, generating a comprehensive database for use in developing and testing numerical models to describe barrier performance and repository saturation rates. These experiments both identified unanticipated interactions between the buffer and the rock mass that could significantly influence the rate of water uptake by the buffer. Rapid heating of the rock mass in the BCE by the simulated container resulted in considerable disturbance to the local porewater pressure regime. In the BCE a zone of high water potential developed approximately 1 m into the rock (Figure 5) and induced unexpected ground water pressures and flow in the vicinity of the borehole. This elevated potential can be explained as being a thermoporoelastic effect caused by thermal expansion of water in the pores of the rock.[11] Due to the low permeability of the rock these pressures had not completely dissipated at the time of decommissioning of the BCE two years later.[4] The result of this elevated pore water pressure was a temporary increase in the rate of flow towards the borehole. The elevated pore pressures would not be expected to continue indefinitely and would gradually decrease as the water moved from the region of high potential into the surrounding regions. Although not at equilibrium at the time of decommissioning, the BCE had begun to see some dissipation of locally elevated pore pressures.

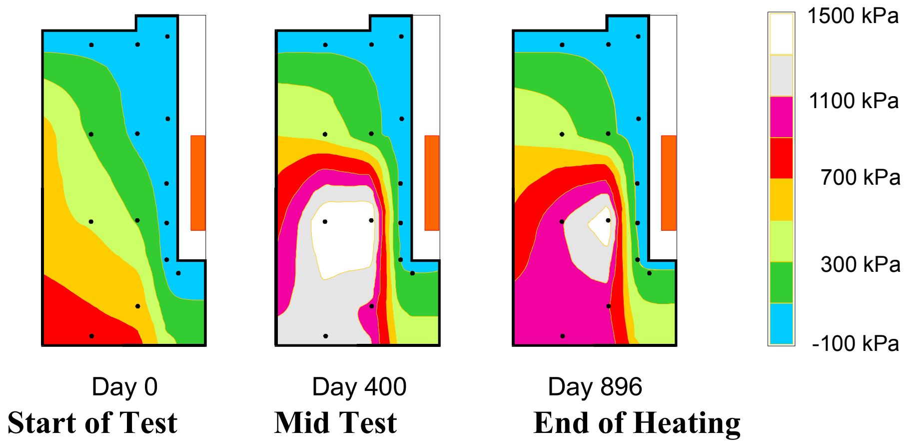

Figure 5: Magnitudes of porewater pressures generated in the rock surrounding the BCE.[4]

The result of such groundwater disturbance might be a longer than anticipated period that the buffer surrounding a container would remain unsaturated. The BCE and ITT both highlighted the importance of the rock immediately adjacent to the installations as well as interface regions between the various components of the sealing system, as important regions where saturation rate and other key performance parameters are likely to be controlled or at least significantly influenced.

These tests also identified a number of factors that had not been recognized as being important to the resaturation process, thereby assisting in developing more representative models for water uptake. Further long-term multi-component simulations of the long-term isolation were recommended in order to validate numerical models for repository resaturation and to extend the range of experience into more complex systems containing other types of barriers such as backfill and tunnel seals.

The Tunnel Sealing Experiment (TSX)

Bulkheads constructed at the entrances to backfilled emplacement rooms locations in a repository play an integral role in ensuring the overall safety of the disposal system. These bulkheads provide restraint for the swelling clay materials that are incorporated within the backfill. They serve to isolate backfilled and closed emplacement rooms from access tunnels that may remain open during the preclosure phase of the repository. The emplacement room sealing systems must minimize seepage, and hence, the potential for advective transport of radionuclides along the room. The bulkhead needs to interrupt axial flowpaths through the near-field rock or the backfilled tunnels, and provide additional sealing capacity. The TSX studied aspects of this system by examining the sealing potential of two bulkheads from the perspectives of both engineering performance and safety assessment.[7,8,9,10]

The TSX, shown in Figure 6, was conducted jointly by JNC of Japan, ANDRA of France, the USDoE (through the science advisor for WIPP) and AECL. One bulkhead was composed of highly compacted sand-bentonite blocks, while the second was constructed using Low-Heat High-Performance Concrete (LHHPC). The region between the bulkheads was filled with sand and water, which was pressurized to 4 MPa, using a static water head. The entire construction was taken through a series of heating stages by circulation of hot water within the sand-filled central chamber. The TSX assessed the constructability of practicable concrete and bentonite bulkheads and evaluated their short-term performance. In the TSX, bulkhead performance was assessed by the ability of the bulkheads to restrict the flow of water in the axial direction of the tunnel. Transport through and around the bulkheads was assessed using tracers injected into the water being circulated through the sand-filled chamber.[10] It should be noted that the TSX bulkheads were not optimized seals but rather two of the components that are most likely to be used in a composite bulkhead.

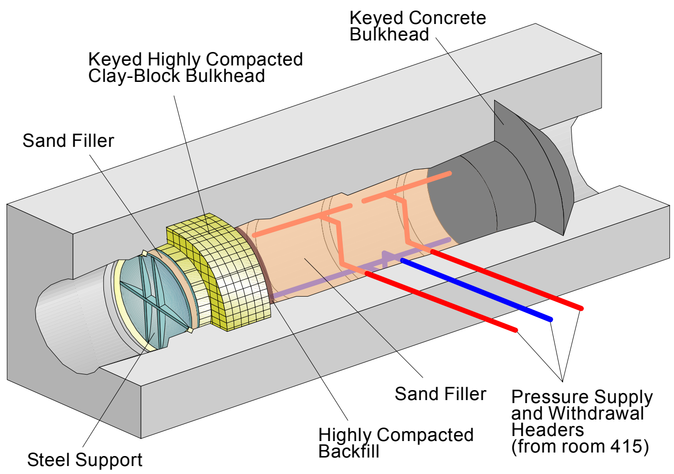

Figure 6: Configuration of the Tunnel Sealing Experiment.

An Excavation Damaged Zone (EDZ) was known to exist in the tunnel walls but beyond a distance of 0.5 m there was little discernible effect on the measured hydraulic conductivity.[7] Based on this information, a 1-m-deep excavation key was determined to be suitable for interrupting virtually all the stress-induced and blast-induced fractures around the tunnel. Therefore, the keys for both bulkheads were excavated by mechanically breaking the rock out from between lines of radial boreholes using hydraulic rock splitters. The excavated surface of the rock on the keys was rough (Figure 7). To facilitate clay block placement and minimize any construction voids, a shot-clay material was applied to this surface creating a smooth contact between the blocks and the rock. The blocks installed in the clay bulkhead had nominal dimensions of 0.1 m by 0.36 m by 0.17 m. Approximately 9000 blocks of 1.9 Mg/m^3 dry density were used in the construction of the bulkhead. At this density, the pressure exerted by the bentonite when it became saturated and swelled was predicted to be about 1 MPa, and the hydraulic conductivity of the main part of the clay bulkhead was expected to be about 10^{-12} m/s.

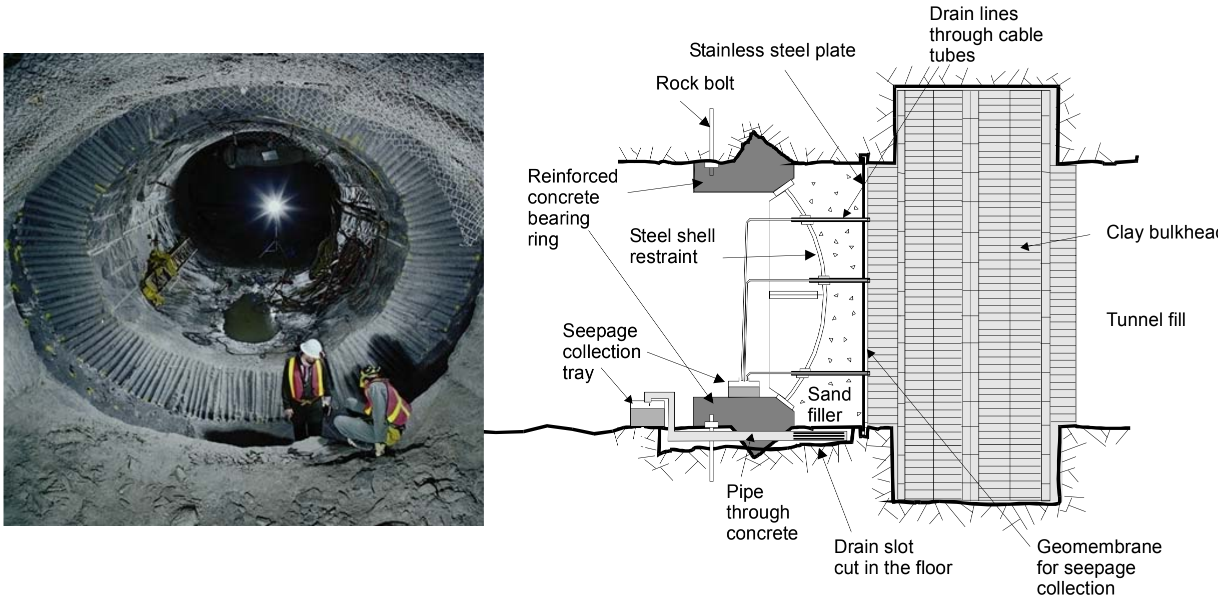

Figure 7: Excavated key and cross-sectional schematic of clay bulkhead.

The clay bulkhead was constrained against volume expansion on one end by a water-saturated sand filler. The other (downstream) end constraint was provided by a rigid steel restraint system (Figure 7) designed to resist the combined stress of 4 MPa of hydraulic pressure and 1 MPa of swelling pressure by transferring the resulting load through the steel shell and onto a high strength concrete bearing pad that was recessed into the rock and secured by rock bolts. A stainless steel plate and sand fill was used to transfer the load uniformly from the clay bulkhead to the steel shell.

Water seeping past the clay bulkhead was collected from 18 separate water collection zones at the downstream end of the clay bulkhead (Figure 7). In addition to these collection points, 235 sensors monitoring pore pressure, clay water content, swelling pressure, temperature and displacement were installed within the clay bulkhead.

The tunnel was filled with water in 1998 September and the pressure was slowly increased ultimately reaching to 4000 kPa. In the early stage of TSX pressurization the central mass of the clay blocks were relatively dry (close to as-placed moisture contents) and joints were still present between the individual blocks. Additionally, the shot-clay placed between the blocks and the rock was not uniformly moist or dense. As a result when the pressure was increased, water would flow along these channels from the sand-filled chamber to the downstream end of the clay bulkhead. During the first six months of TSX operation, eight separate large flow events occurred. After each leak and pressure loss, the tunnel pressure was again slowly increased over several weeks. With time, saturation of the clay blocks adjacent to the flow paths was achieved and swelling pressures were generated, closing the flow paths through the shot-clay. No further flow events occurred after the first six months of TSX operation and seepage rates rapidly decreased.

The second phase of clay bulkhead hydration occurred as water moved slowly through the saturated materials (shotclay) along the perimeter of the clay bulkhead. The result was a radially-inward movement of water together with seepage occurring from the upstream face of the clay bulkhead. This wetting process was captured by a variety of sensors installed within the clay bulkhead. Most of these sensors monitored changes in the total suction within the clay and since saturation is directly related to suction (the higher the degree of saturation, the lower is the suction), they provided a measure of the pattern of water uptake within the clay bulkhead. The suction conditions within the clay bulkhead at four times during TSX operation are shown in Figure 8.

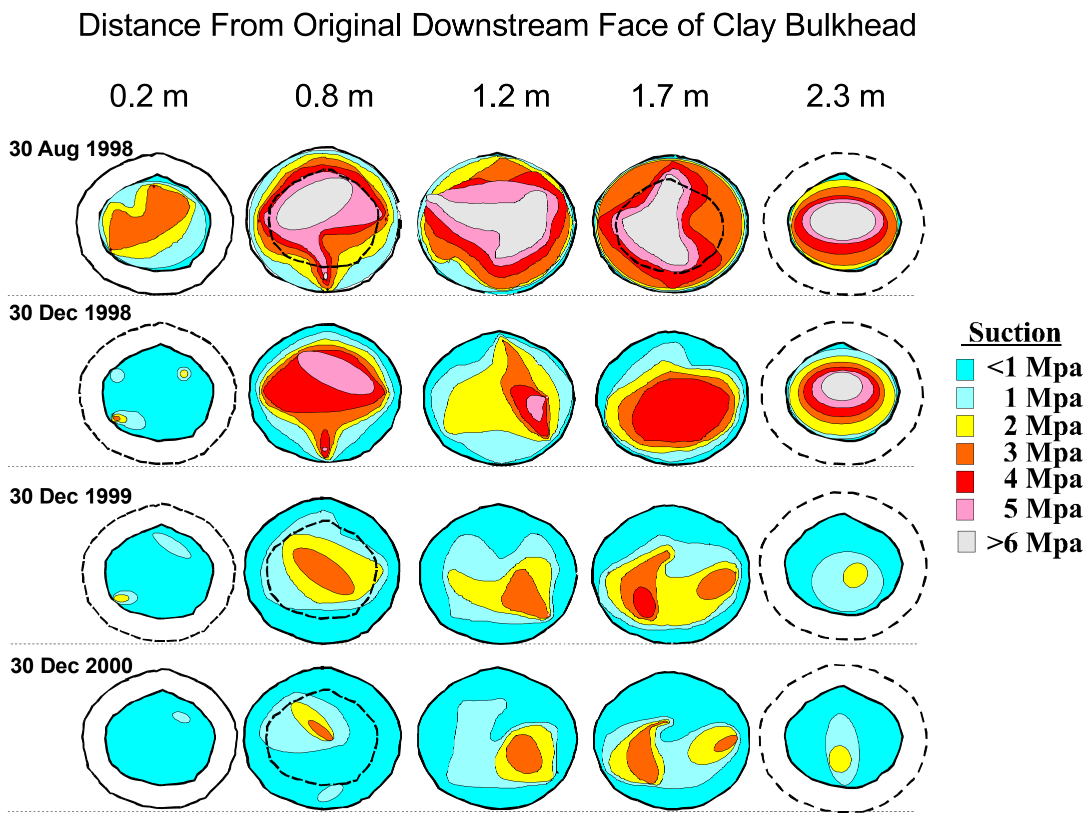

Figure 8. Suction (water content) within clay bulkhead of TSX at various distances from downstream face of backfill wall.

Once the period of equilibration and hydration at 800 kPa hydraulic pressure was completed and a tracer test conducted on the system, it was stepwise pressurized at ~100 kPa per week until it reached 2 MPa, where the system was maintained for approximately 8 months before being incrementally increased to its final operating pressure of 4 MPa. Throughout its pressurization the TSX was carefully monitored to record its hydraulic, mechanical and thermal response.[10] Figures 8 and 9 illustrate the patterns of water influx and distribution measured within the clay bulkhead during TSX operation and at its completion. Once the TSX had reached its maximum operating pressure of 4 MPa, it was allowed to "rest" at this pressure under ambient temperature for approximately 6 months. During this period a second tracer test was conducted before the thermal phase of the TSX began. Two subsequent tracer tests were conducted during the thermal phase.[10]

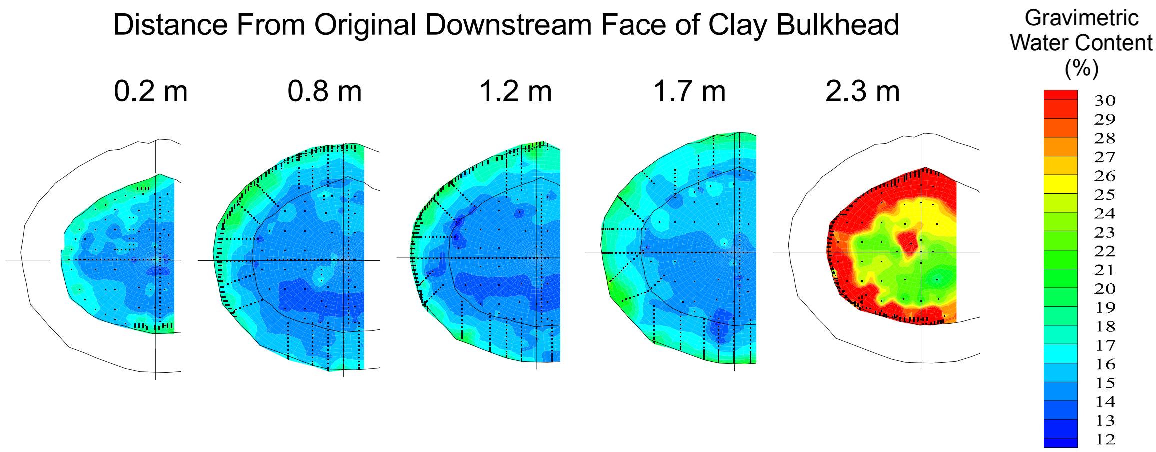

Figure 9: End-of-test water content distribution in TSX clay bulkhead.

The thermal phase of the TSX involved circulating hot water within the sand-filled chamber between the clay and concrete bulkheads consisted of two stages of heating. The first involved heating the upstream face of each bulkhead to 50°C, while monitoring the responses to thermal disturbance. The second stage of heating saw the temperature of the system increased as rapidly as possible using the available heaters. The upstream face of the clay bulkhead ultimately reached a maximum temperature of approximately 60°C before the heaters were turned off and cooling began in preparation for decommissioning of the TSX. In general, temperature had only a minor effect on the clay bulkhead.

In early 2004, decommissioning of the TSX was initiated with drainage of water from the sand-filled pressure chamber and partial removal of the clay bulkhead's restraint system. A comprehensive sampling plan was implemented that allowed the end-of-test water content, density, physical state, and homogeneity to be clearly captured. Figure 9 presents the end-of-test water content and by association, density conditions captured by the detailed sampling of the clay bulkhead during decommissioning.

The TSX has demonstrated how clay and concrete seals could be constructed in a repository using existing technology for placement of concrete and bentonite-based seal material components. The TSX has also provided data that can be used to identify certain performance requirements (e.g., effective hydraulic conductivity of the seal) that can be achieved by bulkheads constructed at the entrances to backfilled emplacement rooms.

Composite Seal Experiment (CSE)

The use of bulkheads having both concrete and swelling clay components was proposed in AECL's EIS[1], and has been incorporated into more recent sealing concepts.[2] The CSE was a logical extension of the technologies and concepts demonstrated in the TSX and examined a seal that incorporated the same components as the TSX (compacted bentonite clay/sand blocks and concrete). It was intended to provide information, at a representative scale, on compacted sand-bentonite material placed adjacent to high performance concrete within a single seal. AECL's Low-Heat High-Performance Concrete (LHHPC)[12] was used in both the CSE and the TSX as it has little or no free lime to react with other components of the sealing system. The first test (CSE-1) (Figure 10), was constructed and pressurized to 2 MPa, and has operated successfully for 4 years. A second test (CSE-2) was under construction when the closure of the URL was announced in 2003 and was never completed. It was originally planned as a series of tests to study the performance of seals with and without keys in low stress, low salinity and high stress, high salinity environments.[3] These experiments were also to have been used to test the longer-term performance of a variety of instrumentation in harsh environments.

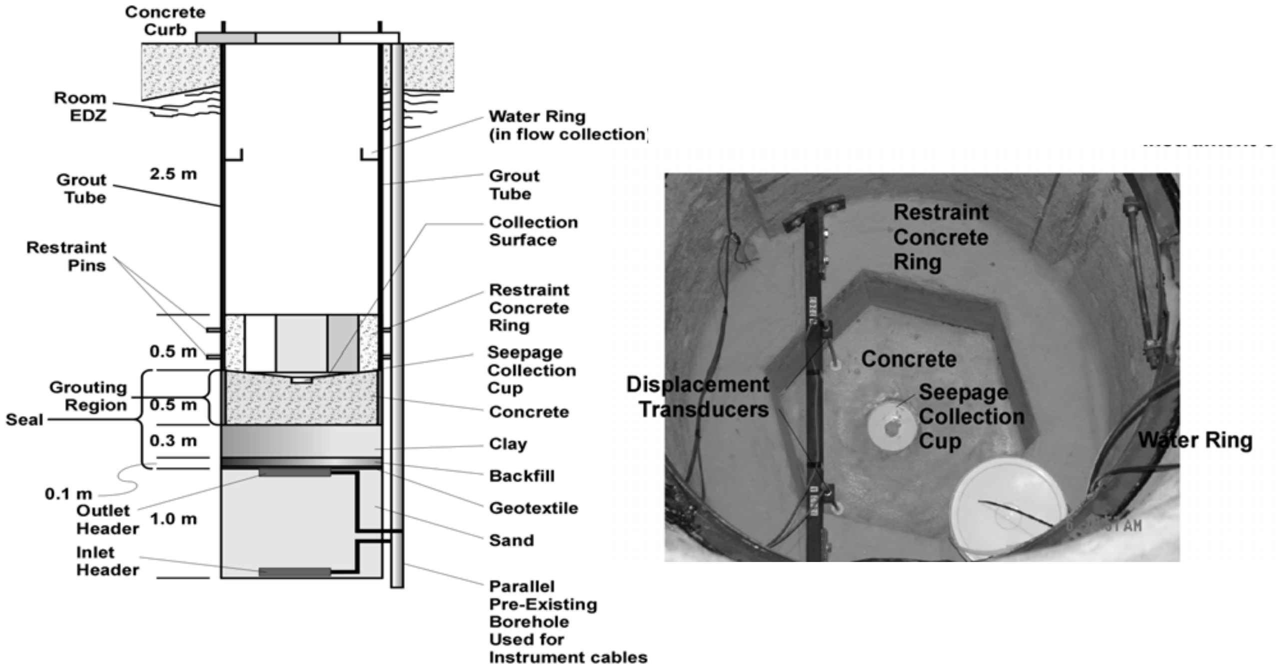

Figure 10: Layout of CSE-1 *(left)* and view of top of concrete *(right)*.[3]

CSE-1 was installed in a pre-existing 1.24-m-diameter borehole excavated using a water-jet drill, on the 240 Level of the URL. The borehole is 5 m deep, which allowed the seal to be placed below the depth of room-induced excavation damage. In CSE-1, a 0.3-m-thick section of TSX-type clay blocks was capped by 0.5 m of LHHPC. The clay block section consists of three layers of 0.105 m-thick blocks, oriented with a 60° offset to each other to minimize the possibility of a vertical joint remaining open between the blocks. Any gaps were filled with crushed block particles. The top of the concrete layer has a collection cup to measure seepage past the bulkhead. A second concrete layer acted as a restraint to the first but played no role in the seepage barrier. A suite of 45 instruments measured pressure in the sand zone under the seal, suction (moisture) and swelling pressure in the clay, strain and displacement of the concrete plug and temperature.[3]

As in the TSX the primary measure of seal performance in the CSE was the rate of water seepage past the seal. Seepage past the composite seal was not observed until the system was pressurized to 1640 kPa and was not measurable until the pressure reached 1840 kPa. Movement of water upwards through clay block assemblage was monitored and indicated that the wetting front moved most rapidly and quite uniformly along the outside perimeter of the clay with the core showing a more gradual saturation. This was a similar pattern to the ITT and the TSX clay bulkhead, where the interface provided the preferred flow path for seepage.

At a constant hydraulic head of 2 MPa, seepage decreased from a peak of approximately 0.04 mL/min to approximately 0.025 mL/min (hydraulic conductivity of 4.7×10^{-12} m/s). This was likely due to the hydrating clay exerting increased pressure on the interface and to the concrete interface accumulating fine clay or colloidal particles and thereby decreasing flow. The seepage rate continued to decrease and at the end of 2004 was immeasurable (<0.001 mL/min or a hydraulic conductivity of ~ 2×10^{-13} m/s). The hydraulic conductivity past the seal in the CSE-1 was two orders of magnitude less than the seepage rate past the TSX clay bulkhead alone, although the scale of the test must be considered together with the lack of a more permeable shotclay component. At 2 MPa the seepage at the TSX clay bulkhead was approximately 0.7 mL/min or a hydraulic conductivity of 1.3×10^{-11} m/s.

In CSE-1 the seepage past the seal indicates that the hydraulic conductivity of the seal is very close to that of the intact rock mass, providing evidence that in an intact rock mass, the excavations can be effectively sealed. The other planned CSE experiments would have provided a measure of the influences of keying, groundwater chemistry and other environmental considerations.

III. SUMMARY

Repository sealing experiments conducted at the URL have contributed greatly to our understanding of both technical and practical issues related to deep geological disposal of nuclear fuel waste. The operation of large-scale *in situ* tests in the crystalline rock of the Canadian Shield during the 14-year operation phase of the URL has generated a wealth of information and experience in the fields of solute transport, rock mass characterization, excavation design and engineered barriers. Large-scale *in situ* experiments provide the most complete assessment of the behaviour of whole systems and for modeling they provide challenging tests. The observed results incorporate the complexities of real, *in situ* system behaviour which must be incorporated by modellers to represent field behaviour in models and interpret *in situ* experiment results.

ACKNOWLEDGMENTS

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