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ZEOLITE PREVENTS DISCHARGE OF STRONTIUM -90-CONTAMINATED GROUNDWATER

David R. Lee, Dale S. Hartwig
Environmental Technologies Branch
Atomic Energy of Canada Limited
Chalk River Laboratories, Chalk River, Ontario

ABSTRACT

In December 1998 a vertical, permeable "barrier' of granular zeolite (clinoptilolite) was emplaced beneath the surface of the earth, in the path of groundwater that was transporting strontium-ninety (^{90}Sr), one of the most ubiquitous groundwater contaminants at nuclear installations, toward a wetland on the property of AECL's Chalk River Laboratories. This emplacement of zeolite, known as the Wall-and-Curtain, constituted the nuclear industry's first permeable reactive barrier. Through the process of ion exchange, the zeolite was intended to prevent the movement of ^{90}Sr into the wetland.

The Wall-and-Curtain featured 1) direct measurement of flow and contaminant concentration of the water exiting the barrier and 2) hydraulic adjustment of the groundwater capture zone. During the first 5 years of operation, this permeable reactive barrier saved $200,000 per year in operational costs as compared with a comparable "Pump and Treat" system and on that basis has paid for itself in less than two years.

BACKGROUND

Two alternatives exist for treatment of contaminated groundwater: 1). the pump and treat option in which extraction wells are installed in the contaminated aquifer and groundwater is pumped to surface where it is treated and 2). a passive system known as a permeable reactive barrier (PRB) where a suitable material is placed across the moving groundwater so that contaminants are attenuated while the water moves through (e.g. Benner, 1997). Natural attenuation processes include radioactive and microbial decay.

A permeable reactive barrier, known as the Wall-and-Curtain (WC), was installed in December of 1998 on the property of Atomic Energy of Canada Limited, Chalk River Laboratories, Chalk River Ontario. Its purpose was to prevent the discharge of ^{90}Sr-contaminated groundwater.

AQUIFER AND ITS CONTAMINANT PLUME

In the early 1950s, a pilot plant was operated at the Chalk River Laboratories for the purpose of decomposing and reducing the volumes of ammonium nitrate solutions containing mixed fission products. Some of these solutions were released into a pit lined with crushed limestone. The most important constituent of the release was ^{90}Sr. Medium to fine grained sands, derived from granitic gneiss, overlie the bedrock. The saturated thickness of the sandy aquifer ranges in thickness from 5 to 13 m. The hydraulic conductivity of the aquifer is in the range 10^{-4} to 2 X 10^{-5} m/s. Killey and Munch (1987) described the hydrogeology of this groundwater-contaminant plume. In 1994 the advancing front of the ^{90}Sr plume was believed to be about 7 m wide and was located within the deep portions of a 12 m-thick aquifer. Groundwater at this location moves at about 150 m per year, but because of geochemical interactions, the leading edge of the ^{90}Sr plume moved much slower and required over forty years to migrate from its initial source area to Duke Swamp, about 440 m down gradient.

DESIGN OF WALL AND CURTAIN

A simple trench-type PRB may have been possible at this site, but it would not have met three desired goals: 1) if the standard for treatment is pump and treat, then effluent must be monitored, 2) if the position or width of the plume changes over time, then control of the capture zone would be necessary, 3) if a reactive barrier is the first of its kind, then it should be closely monitored and documented, so that a determination of the effectiveness of the barrier material can be made before future application to other contaminant plumes. Most present-day PRBs do not include effluent monitoring, but performance monitoring is expected for radioactive contaminants. A trench-type PRB, without hydraulic controls, would also have required excavation to nearly the full 12-m depth of the aquifer, which would have been considerably more expensive.

AECL's Wall-and-Curtain design (Figure 1) consists of a sealed-joint, steel-sheet pile cut-off wall (Waterloo Barrier®) located on the down-gradient side of a granular zeolite "curtain" or barrier (Lee et al, 1998). The cut-off wall prevents groundwater backflow from the wetland and, during construction, functioned as a retainer to allow excavation. The cut-off wall was 30 m in length and extended 9.5 to 12 m into till or to contact the bedrock. Individual lengths of the interlocking steel-sheet piling were vibrated into the ground to refusal. Tee sections allowed sheet piling to be driven perpendicular to the wall to form the sides of the box that contained the granular curtain. The wall and sides were permanently grouted with a cement-based grout to create an impermeable barrier. The front of the box was created using the same sheet piling and temporarily grouted. This section of piling was withdrawn from the ground once the curtain was in place.

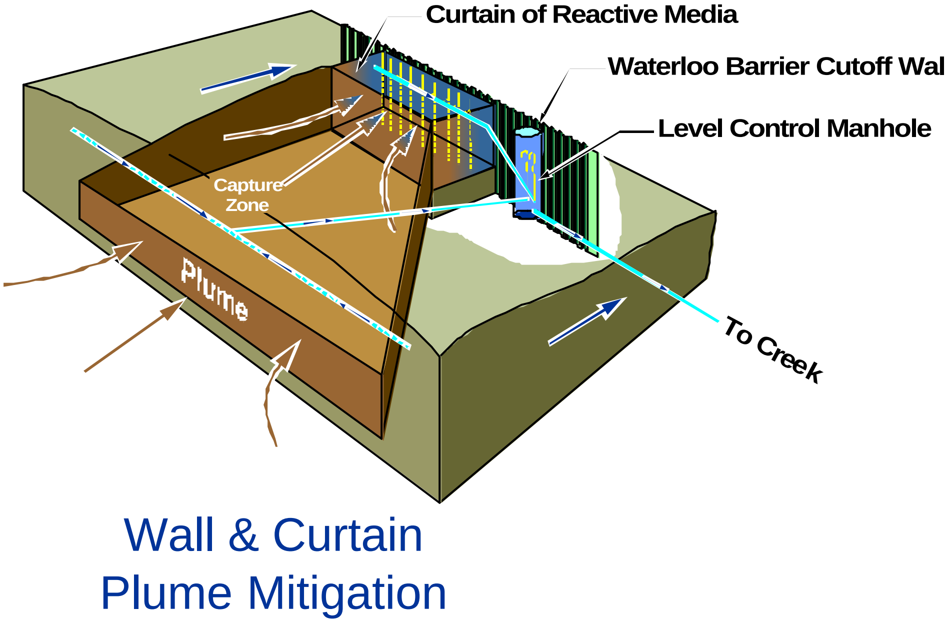

Figure 1: AECL's Wall-and-Curtain.

Six dewatering wells, two each on the up gradient and down gradient sides of the sheet piling box and one at either end were installed to a depth of 10 m. Pumping these wells lowered the water table to a depth approximately six meters below grade and allowed dry emplacement of the granular zeolite once the soil inside the sheet-piling box was excavated.

The reactive media used in the curtain at this site was 130-m^3 of 14 × 50 mesh granular clinoptilolite (zeolite). This granular zeolite has a high affinity for ^{90}Sr. The reactive material was situated in front of the cut-off wall to form the PRB and was 2.0 m in length, 11.0 m in width, and 6.0 m deep. The PRB extended from just below grade to about 6.1 m below the surface.

Two drainage systems allowed operators to exert hydraulic control over the groundwater flow system immediately up gradient and to perform effluent monitoring at a single discharge pipe.

1. A series of 10 vertical, continuously slotted, well screens was located in the curtain immediately in front of the wall. The wells were linked to a drain that terminated with a flexible outflow hose in a concrete manhole. This provided adjustment in outflow elevation, thereby allowing operational control of groundwater throughput and allowed for effluent monitoring at a single pipe. The concrete manhole drained to a low point in the local topography.

2. A second drainage pipe was located transverse to the path of groundwater and 60 m up gradient of the wall and curtain. This pipe also terminated in a flexible outflow pipe in the concrete manhole. The purpose of the horizontal up-gradient drain was to divert the shallow, contaminant-free groundwater around the treatment system, thereby extending the life of the ion-exchange material.

By adjusting the elevation of each of the two flexible outflow pipes to maintain groundwater levels within the PRB, operators set a flow rate and a capture zone that corresponded to a volume of moving groundwater that was slightly larger than the contaminant plume. The elevation of the PRB outflow pipe controlled the flow of contaminated groundwater into the zeolite. If adjusted properly, the inflowing groundwater also included an envelope of uncontaminated groundwater to the sides and beneath the contaminated groundwater. Adjustment of the groundwater capture zone in the lateral dimension has been confirmed with tracers. The elevation of the up-gradient drain outflow pipe allowed vertical capture to be controlled somewhat in that shallow, non-contaminated groundwater was diverted around the system, thus causing only the deeper part of the aquifer to move toward the WC. By following changes in the salinity profile of the aquifer (as the amount of diverted water was increased or decreased) the operators monitored the vertical dimension of the capture zone. The vertical profile of salinity, greater at depth, is predominantly due to road salting within the catchment.

Because measurement of flow out of the WC did not depend on estimates of permeability, water treatment volumes are uncertain by a factor of only 10%, and could be better if needed. This compares with uncertainties greater than 100% in most hydrogeological estimates of flow, owing to the uncertainties of permeability in natural deposits. Thus the flow and treatment character of the WC is considerably better than with a trench-type, traditional PRB where the effluent is usually not measurable directly because it exits from the down-gradient face of the permeable material, not through a pipe. Based on preliminary testing, using columns of zeolite in a test well in the contaminated portion of the aquifer, the 2-metre thick curtain of zeolite was predicted to retain ^{90}Sr for over 20 years.

PERFORMANCE

This facility treats 1.5×10^7 L per year (7.6 gpm) of ^{90}Sr contaminated ground water, while diverting 10^7 L per year of shallow, uncontaminated groundwater, which would otherwise enter the PRB. In the past two years, the WC has prevented the discharge of 1.7×10^9 Bq of ^{90}Sr into the adjacent wetland. Based on monthly sampling, gross beta measurements of effluent are less than 0.6 Bq/L as compared with influent groundwater with a ^{90}Sr activity of 85 Bq/L (Figure 2) (AECL, 2003). The drinking water standard is 5 Bq of ^{90}Sr per litre. The system has captured 99.5% of the ^{90}Sr that would otherwise have entered the wetland (AECL, 2003).

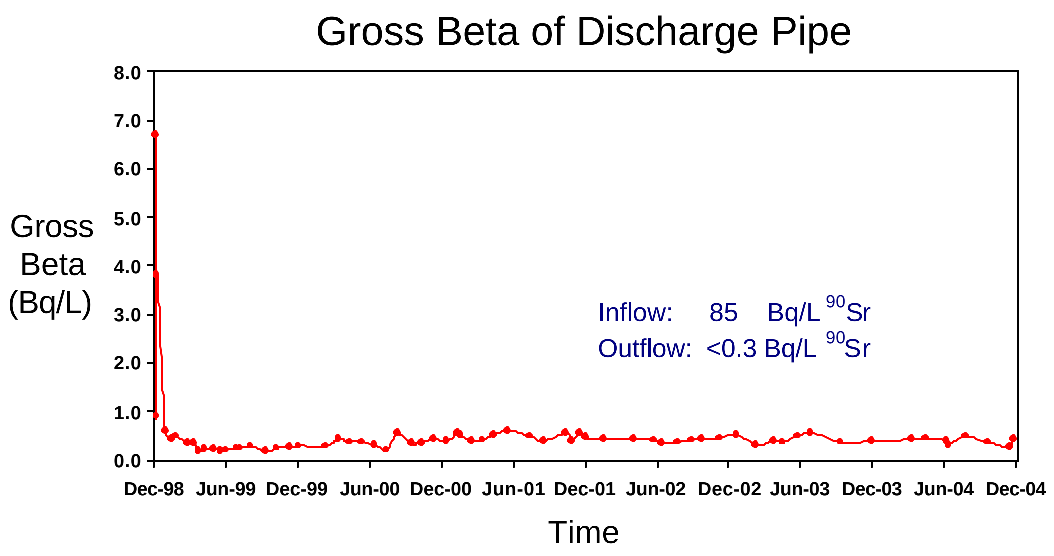

Figure 2: Ground-water outflow from the WC discharge pipe has remained low (AECL, 2003), well below the 5 Bq/L Canadian drinking water quality guideline for ^{90}Sr. One half of the measured gross beta is a good approximation of the ^{90}Sr concentration.

The total cost of the PRB was approximately $300,000. During the first 5 years of operation, this PRB saved $200,000 per year in operational costs as compared with a comparable "Pump and Treat" system, in which groundwater is pumped to surface where ^{90}Sr is concentrated, immobilized and disposed of. On that basis, the PRB paid for itself in less than two years.

Plan view dimensions of the capture zone were derived by simple flow-net analysis of water table maps prepared biannually. Water-table elevations were measured in wells at 5 m well spacings within 10 m of the WC, at 10 m well spacings within 20 m of the WC and 15 to 30 m well spacings within 40 and 100 m distances from the WC. Over 50 wells were used to produce a water table map within a 24,000-m^2 area.

POTENTIAL PROBLEMS

In the longer-term, there are issues with PRBs, such as maintenance and documentation of performance, common to all contaminated groundwater collection and treatment systems. There are also other issues related to site-specific groundwater chemistry and peculiarities of the installation itself. However, in general the performance of the WC during 5 years of continuous operation has been free of problems.

At this site, the contaminated groundwater is laden with ferrous iron in excess of 20 mg/L. If this water were to contact dissolved oxygen and precipitate iron oxy-hydroxide in the granular interstices of the reactive medium, permeability would decline and contaminated groundwater would by-pass the PRB and accumulate in the organic matter of the wetland. However, the mean and standard deviation for four hydraulic conductivity values measured in July 1999

(8.78×10^{-4} to 1.98×10^{-4} m/s) compared favourably with four values measured in September 2004 (8.20×10^{-4} to 7.46×10^{-5} m/s). From this we concluded that there has been no significant change in permeability so far.

Based on flow out of the small pond down gradient of the pilings, there appears to be leakage of basal waters in the aquifer, beneath the steel cut-off wall. The leakage, as much as 2.7 L/min (0.7gpm) could be due to an hydraulic gradient and the lack of a seal between the lower end of the steel sheet piling wall and the bedrock. The seriousness of this leakage is lessened because the basal groundwater is not contaminated and because the hydraulic head in the PRB is set few tens of centimetres below the local, natural level of the water table and thus induces upwelling into the reactive medium. If grouting were performed and were effective in stopping this leakage, performance monitoring over the long term would be simplified.

LESSONS LEARNED

Although performance monitoring has indicated excellent performance so far, the project team has learned a few lessons mostly related to the construction phase.

1. Allow a contingency budget for procedures that may be discovered, and effectively incorporated, during the construction phase.

Sand packing of selected dewatering wells with reactive zeolite proved to be prudent. This reduced the concentrations in the water released as a result of dewatering during construction. The method was easy to use and probably provided better sand pack properties than simple surging of the dewatering in wells. As effective as this step might proved to be, it was used at only 3 of the 6 dewatering wells, due to time limitations.

2. Components of the system must be as accessible as possible for inspection and cleanout.

Although the up-gradient drain performs the job of shunting shallow, uncontaminated ground water to the nearby wetland and reducing the ion load on the zeolite, the drain requires yearly cleaning at the base of the overflow tube where there is a constriction. The constriction provides a place for iron oxide to accumulate and, before discovered, had resulted in almost complete blockage of flow. Fortunately so far, 6 years into operations, this constriction is assessable and is being cleaned periodically.

CONCLUSIONS

AECL's Wall-and-Curtain has performed as intended both chemically and physically. It has prevented the movement of ^{90}Sr contaminated groundwater plume into a wetland.

The design has allowed: 1) direct measurement of flow and concentration of the water passing through the PRB, 2) hydraulic adjustment of the groundwater capture zone both vertically and horizontally and 3) detailed analysis of zeolite performance in terms of preferential flow rate measurements as well as detailed sampling as the contaminant front moved into, and through, the ion-exchange barrier.

This particular design requires almost no cost to operate.

On the basis of cost relative to an alternative "Pump and Treat" system, this PRB paid for itself in less than two years.

Plan view dimensions of the capture zone can be adequately determined using biannual water table maps generated from water table elevations measured in more than 50 wells within a 24,000 m^2 area adjacent to the Wall-and-Curtain.

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