INTERCEPTION OF A GROUNDWATER PLUME CONTAINING STRONTIUM-90

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ABSTRACT

In December 1998, a vertical, permeable "barrier' of granular zeolite (clinoptilolite) was emplaced underground in the path of groundwater that was transporting strontium-90 (⁹⁰Sr) toward a wetland on the property of Atomic Energy of Canada Limited's Chalk River Laboratories (CRL). The primary components of the barrier were a 30-m long, steel-sheet piling cut-off wall and a 130 m³ volume of granular clinoptilolite. The granular clinoptilolite was to act as an ion exchange permeable reactive barrier (PRB) to intercept the ⁹⁰Sr groundwater plume. It was 2-m thick in the direction of groundwater flow, 11-m wide and 6-m deep. It was not emplaced at the depth of groundwater contamination. Instead, the clinoptilolite was "hung" like a "curtain" with its base at an elevation 3 to 4 m above the natural pathway of the groundwater. A locally reduced elevation head in the curtain was intended to cause the groundwater plume to angle upward and to flow through the curtain.

The concentration of 90 Sr in the influent groundwater has been about 400 Bq/L and the 12-year record of gross beta measurements has shown no sign of 90 Sr breakthrough. Attempts to measure 90 Sr radiochemically revealed less than 0.1 Bq/L 90 Sr in the effluent. Hydraulic controls on the incoming groundwater have performed as intended. As of this writing, the groundwater plume continued to be intercepted and the reactive barrier had been effective for long-term subsurface control and mitigation of groundwater contaminated with 90 Sr.

1. INTRODUCTION

A plume of strontium-90 (⁹⁰Sr)-contaminated groundwater was generated during the operation and closure of a 1950's vintage pilot plant on the property of Atomic Energy of Canada Limited, at the Chalk River Laboratories (CRL). The purpose of the plant was to decompose and reduce the volumes of ammonium nitrate solutions containing mixed fission products. The source of the contaminated groundwater was the release of solutions containing a variety of nuclides, including ⁹⁰Sr, into a pit lined with crushed limestone [1]. Groundwater transport and control of ⁹⁰Sr is of considerable interest because it is a significant product of nuclear fission, is somewhat mobile in groundwater systems and tends to accumulate in vegetation and animals as an analog to calcium.

Killey and Munch [2] characterized the hydrogeology of the area and documented the migration of the ⁹⁰Sr groundwater plume from its source toward the Duke Swamp wetland. The saturated thickness of the medium- to fine-grained sand aquifer ranged from 5 to 13 m. The hydraulic conductivity of the aquifer was in the range of 10^{-4} to 2×10^{-5} m/s [2]. In the early 1990's, it became apparent that the advancing front of the ⁹⁰Sr plume was approaching a point of discharge in the wetland [3]. The leading edge of the plume was at a depth of about 10 m and was about 7 m wide. Groundwater movement from the source to the wetland discharge zone, a distance of about 440 m, had a transit time of about 1.5 years, whereas the front of the ⁹⁰Sr plume required over forty years to travel this distance [3].

2. BACKGROUND

Two methods were considered for treatment of contaminated groundwater: 1) pump water from the ground and treat it at the surface using extraction wells in the contaminated aquifer, and 2) create a subsurface, reactive zone through which the groundwater would pass, where the contaminant is retained or attenuated by natural mechanisms. This method is commonly called a permeable reactive barrier (PRB). Natural processes of attenuation include ion exchange, precipitation and radioactive or microbial decay. PRBs can often be cost-effective in controlling groundwater plumes [4].

A trench-type PRB would have been possible to construct at this site, but it would not have met the goals we set for ourselves: 1) cost-effective, 2) ability to allow detailed long-term monitoring of the effluent, 3) ability to allow determination of the hydraulic conductivity and reactivity of the PRB, 4) control of the width of the capture zone so that if, over time, the plume became broader or changed direction, it could still be captured and, 5) no requirement to excavate contaminated soil so as to avoid the costs associated with handling and storage of contaminated soil. If the goals could be met, the design would allow monitoring of monthly performance and would provide a basis for application on other contaminant plumes.

We designed and implemented a system to meet the above goals and intercept the groundwater ⁹⁰Sr plume emanating from the former pilot plant [5]. The Wall and Curtain was installed in December of 1998 (Figure 1). It consisted of a cut-off <u>wall</u>, a <u>curtain</u> of permeable reactive media, an <u>upgradient horizontal drain</u>, and a <u>flow control</u> that may be used to adjust the groundwater capture zone. The actual facility was modified somewhat from that described in reference [6].

The cut-off wall is 30 m in length and extends 9.5 to 12 m into till or into contact with bedrock (Figure 1). The cut-off wall was intended to prevent the induced reduction in hydraulic head from causing groundwater flow into the reactive barrier from the wetland side. A locally reduced hydraulic head in well screens, at the back of the curtain of reactive media, was to propagate into the groundwater plume. The curtain of permeable reactive material is granular (14 x 50 mesh) clinoptilolite which was intended to retain ⁹⁰Sr by the process of ion exchange. A horizontal, upgradient drain was installed to divert the shallow, uncontaminated groundwater so it would not enter the barrier and carry competing ions into the ion exchange material. The range of adjustable elevation overflows in the Level Control Manhole (Figure 1) was to allow operators to minimize the dimensions and costs of the facility and to provide the ability to accommodate a wider groundwater capture zone, if needed, in the future [5, 6].

Prior to installation of the PRB, we performed a down-hole geochemical column experiment to test clinoptilolite performance *in situ*. This provided confidence that a volume of granular clinoptilolite, permeable than the aquifer, could retain ⁹⁰Sr. To our knowledge, this was the nuclear industry's first PRB. The design was more than a unique combination of three existing technologies (cut-off wall, horizontal drain and emplacement of reactive material to treat the groundwater underground), which allowed testing of groundwater plume capture. It was also a system that provided ready access to its effluent for flow and quality monitoring and sampling instrumentation to facilitate future evaluation of geochemical performance.

Before the design was implemented, we solicited world-class, external, critical review of the conceptual design; consulted with sealable joint sheet piling suppliers as to best practice; prepared proposals for managerial approval; presented a seminar on the design to technical staff at the Atomic Energy Control Board, our regulator; constructed a working sandbox model with hydraulic controls; supervised a numerical modeling contract to link the design to site-specific conditions; supervised an engineering contract to translate the design into construction drawings and specifications; buried existing aboveground instrumentation so construction could take place without damaging it; coordinated with CRL heavy equipment personnel to prepare the site; and presented site details and overall design concepts to an international conference [5].

During construction, we served as CRL technical representatives. We implemented design changes when faced with unforeseen situations and videotaped the construction sequence.

Following construction, groundwater throughput was adjusted to accommodate the groundwater plume. The Annual Safety Reports of AECL [7] include a brief summary of each year's results.

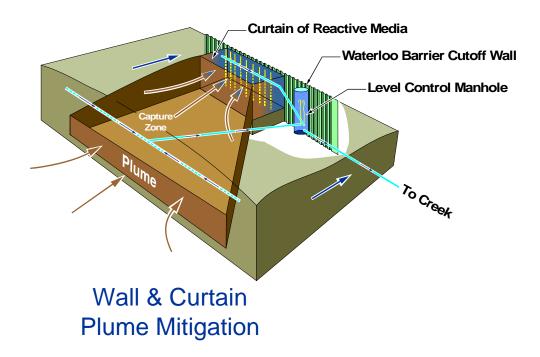


Figure 1. Schematic of AECL's Wall and Curtain permeable reactive barrier.

3. **RESULTS AND DISCUSSION**

The record of radioactivity, measured as gross beta, in the effluent/discharge from the Wall and Curtain reactive barrier indicates that it contains virtually no ⁹⁰Sr (Figure 2). When ⁹⁰Sr is a significant contaminant in CRL aquifers, half of the measured gross beta is often a good approximation of the ⁹⁰Sr concentration. However, in this case, where levels of gross beta are at environmental background levels of about 0.5 Bq/L, it is likely that the gross beta measurements represent only the naturally occurring radioactivity, such as ⁴⁰K and uranium- and thorium-series nuclides. Attempts to measure ⁹⁰Sr in the effluent with a method having a detection limit of 0.1 Bq/L found no detectable ⁹⁰Sr in the effluent of the Wall and Curtain. The method involved evaporating a 10 L sample, processing through four columns to separate and purify Sr-90 and quantifying by liquid scintillation counting. In fact, effluent concentrations have remained at or near 0.5 Bg/L (as gross beta) since shortly after start-up in 1999. The effluent remained at about 0.5 Bq/L, i.e. it was virtually free of ⁹⁰Sr. Based on the concentrations of ⁹⁰Sr in the influent groundwater and the measured rate of flow through the PRB, the apparent total capture of ⁹⁰Sr since the facility became operational in 1998 has been 5.03 E+10 Bq. Thus far, the facility has had a trapping efficiency indistinguishable from 100% because no breakthrough has been observed in monthly analysis of the effluent. The Wall and Curtain has retained ⁹⁰Sr that would otherwise have discharged to the wetland [7].

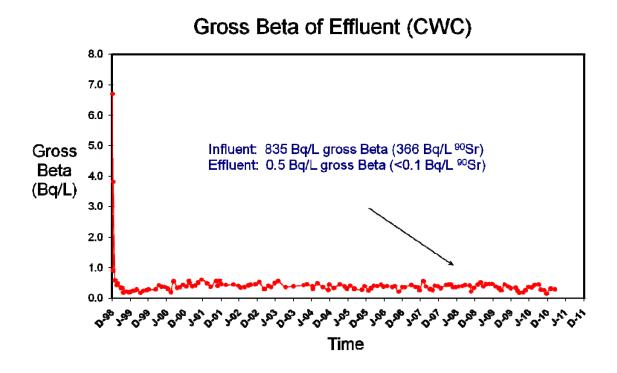


Figure 2. Gross beta concentrations in the Wall and Curtain effluent over the 12-year period from December 1998 to January 2011.

PRBs can have maintenance, permeability and proof-of-performance issues. There can also be issues related to site-specific groundwater chemistries and the peculiarities of PRB installations [4]. While no groundwater collection and treatment system is perfect, the monitoring of the effluent, a unique feature of the Wall and Curtain design, has aided in collecting the data needed to test for changes in permeability and performance as a barrier to contaminant migration. Records on the hydraulic gradient through the barrier and on the groundwater throughput and effluent quality have provided confidence in the performance of the Wall and Curtain PRB versus what would have had to be accepted had a trench or funnel-and-gate-PRB been constructed instead. Having experienced few issues or problems, we concluded that the Wall and Curtain has performed as intended.

4. CONCLUSIONS

The Wall and Curtain PRB had intercepted the Nitrate Plant Plume for 12 years as of January 2011. The use of commercially-available granular zeolite (clinoptilolite) proved suitable for long-term subsurface control and mitigation of groundwater contaminated with ⁹⁰Sr. The multi-year record showed no sign of ⁹⁰Sr breakthrough and the hydraulic controls have performed as intended. Had a trench or funnel-and-gate PRB been used instead, the facility would have been more expensive to build, difficult to monitor for performance and administratively messy and excessively costly regarding handling, storage and disposal of excavated/contaminated soil. A

trench or funnel-and-gate PRB would also have not allowed us to control the dimensions of the capture zone. The facility design has allowed on-going, long-term performance evaluations. Results have encouraged the use of clinoptilolite for control of other groundwater plumes at CRL and at West Valley, NY. In addition, the physical features of the Wall and Curtain could be used for evaluating the control of other groundwater contaminants.

5. ACKNOWLEDGEMENTS

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