

DEVELOPMENT AND OPERATION OF A PASSIVE-FLOW TREATMENT SYSTEM FOR ⁹⁰SR-CONTAMINATED GROUNDWATER

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ABSTRACT

Seep C was a free-flowing stream of groundwater that emerged in a narrow valley below the old low-level waste (LLW) disposal trenches in Solid Waste Storage Area 5 (SWSA 5) at Oak Ridge National Laboratory (ORNL). The flow rate of the seep water was strongly influenced by rainfall, and typically ranged from 0.5 to 8 L/min. The seep water entered Melton Branch, a small stream that joins White Oak Creek before exiting the ORNL boundary. The seep water contained high concentrations of ⁹⁰Sr (10,000 to 20,000 Bq/L) and, before the full-scale treatment system was installed, contributed about 25% of all the ⁹⁰Sr leaving ORNL. Seep C was identified as a primary source of off-site contaminant transport and was designated for an early removal action under the Comprehensive Environmental Response and Liability Act (CERCLA). A passive flow treatment system was chosen as the most cost-effective method for treating the water.

In order to provide design and operating data for the full-scale system, a pilot-scale treatment system consisting of a 5-gal bucket filled with 16 L of chabazite zeolite, was used to treat the seep water. The test was started on 3/17/94 and concluded on 6/15/94. The system treated 63,470 L (3967 bed volumes) of water and removed 22.7 mCi of ⁹⁰Sr from the seep water. The system removed more than 99.5% of the ⁹⁰Sr from the incoming water for the first 43,000 L of water treated. Then the removal efficiency slowly decreased, as the zeolite became loaded, until it reached 84% removal for the final sample. The passive system performed at least as well as comparable pumped zeolite systems in terms of ⁹⁰Sr removal efficiency and zeolite utilization. The test was terminated just before the construction crew mobilized at Seep C to build the full-scale system.

The full-scale system consists of a french drain to collect the water and eight 55-gal drums of zeolite (Norit Americas, Atlanta, GA), located in an underground vault, to provide treatment. The zeolite drums are arranged in a lead-lag configuration, with the four lead drums connected to a common header and the four lag drums supplied from a common header on the exit of the lead drums. There is an elevation drop of about 1.5 m from the french drain to the zeolite drums, which pushes the water successively through the zeolite, an air lock, and finally into Melton Branch.

The treatment system typically removes over 99.9% of the ⁹⁰Sr from the water passing through the drums of zeolite. The system has treated more than 3.3×10^6 liters of water since startup, and has removed over 1.6 Ci of ⁹⁰Sr. The iron hydroxide present in the seep water, which can plug the zeolite and restrict flow through the treatment drums, has been a continuing problem, and has resulted in additional costs of at least \$50,000. Nitrogen gas, which is being added to the french

drain at a flow rate of about 10 L/min, has reduced, but not eliminated, the iron hydroxide problem.

Pilot-scale filtration tests have been conducted at the site to determine whether it would be feasible to pump water from the french drain, through a pressure-filtration system, and then at low pressure through the existing zeolite drums. The tests showed that removal of the very small iron hydroxide particles required a 0.5 μm pore-size filter, which plugged very quickly. It does not appear feasible to use a filtration system on the seep water.

INTRODUCTION

Seep C was a free-flowing stream of groundwater that emerged in a narrow valley below the old disposal trenches in Solid Waste Storage Area 5 (SWSA 5), which is part of Waste Area Grouping 5 (WAG 5). The seep water entered Melton Branch, a small stream that joins White Oak Creek before exiting the ORNL boundary. The seep water contained high concentrations of strontium-90 (^{90}Sr) and tritium (^3H). The ^{90}Sr concentration ranged from 10,000 to 20,000 Bq/L, with ^3H concentrations of about 100,000 Bq/L. The flow rate of the seep varied considerably, with measured flows ranging from 0 to 59 L/min; however, more typical values were 0.5 to 8 L/min. Measurement of the total ^{90}Sr flux from the Seep C area into Melton Branch suggested that there was a significant underground component of the flow (about 0.3 L/min) during dry weather. The seep contributed about 25% of all the ^{90}Sr leaving Oak Ridge National Laboratory. Typical chemical data for the seep water are shown in Table I.

Table I. Typical Metals Concentrations in Seep C Water

Metal	Concentration (mg/L)	Metal	Concentration (mg/L)
Ba	0.096	Mo	0.04
Ca	86.0	Na	8.2
Fe	0.078	Ni	0.010
Mg	16.0	Sr	0.17
Mn	0.22	Zn	0.011

About 86,000 m^3 of waste was buried in SWSA 5 between the time it opened in 1958 and closed in 1973. The southern portion of SWSA 5, which is above Seep C, was used as a shallow-land burial ground for solid low-level waste. The trenches were 4 m wide, about 5 m deep, and 12 to 150 m long. The trenches generally ran parallel to the slope, so water can easily flow down through them to the bottom of the hill, which is just upgradient from Seep C. Water from one or

more of the trenches in SWSA 5 contributes the radionuclides found in the Seep C water. A Remedial Investigation and Feasibility Study (RI/FS) has been completed for WAG 5, and the Record of Decision (ROD), which will specify the final closure methods, is expected within the next few years.

Seep C was identified as a primary source of off-site contaminant transport, and was designated for an early removal action under the Comprehensive Environmental Response and Liability Act (CERCLA). An Engineering Evaluation and Cost Analysis (EE/CA) considered a range of alternatives for treating the water at Seep C (1). A passive treatment system located at the seep was identified as the least expensive option for treating the wastewater, but the innovative nature of the proposed treatment increased the level of uncertainty. A backup option identified was to truck the water to the Process Waste Treatment Plant (PWTP), a full-scale wastewater treatment facility for slightly contaminated water located on the west end of ORNL. The goal of the removal action was to have a system operational by 11/30/94 that reduced the ^{90}Sr concentration in the water treated by at least 90%. Laboratory-scale tests on a passive treatment system using zeolite, an inorganic ion-exchange media, and Seep C water had started earlier as part of another project. A pilot-scale treatment demonstration of the zeolite system was initiated following release of the EE/CA report (1).

PILOT-SCALE TEST SYSTEM DESCRIPTION

Following several unsuccessful system configurations (described below), a downflow column system was developed. A 5-gal plastic bucket (28 cm ID by 36 cm high) was modified to act as a column. An inlet hose was connected from a water collection trough to a bulkhead fitting placed in the top of the bucket, and a small splash plate was attached below the fitting to help distribute the incoming water (see Fig. 1). The trough provided hydraulic head to drive water through the treatment system and allowed most of the solids in the seep water to settle before the water entered the bucket. A perforated pipe, wrapped with 100-mesh screen, was connected through the side of the bucket (about 2 cm from the bottom), and another 100-mesh screen covered the bottom portion of the bucket just above the pipe. The bucket was filled to within 4 cm of the top with 16 L (26-cm bed height) of chabazite zeolite (TSM-310, Steelhead Specialty Minerals, Spokane, WA), an inorganic ion-exchange resin. Chabazite zeolite is a naturally occurring sodium aluminosilicate mineral that has both cation-exchange and molecular sieve properties (i.e. ion selectivity depends on both size and charge). Compared to common organic ion-exchange resins, zeolite is more selective for strontium than for calcium and magnesium, which are present in high concentrations in the seep water (2). Zeolite also provides a stable waste form for disposal. Laboratory-scale tests on Seep C water had previously shown that chabazite zeolite was effective for removing the ^{90}Sr from the water.

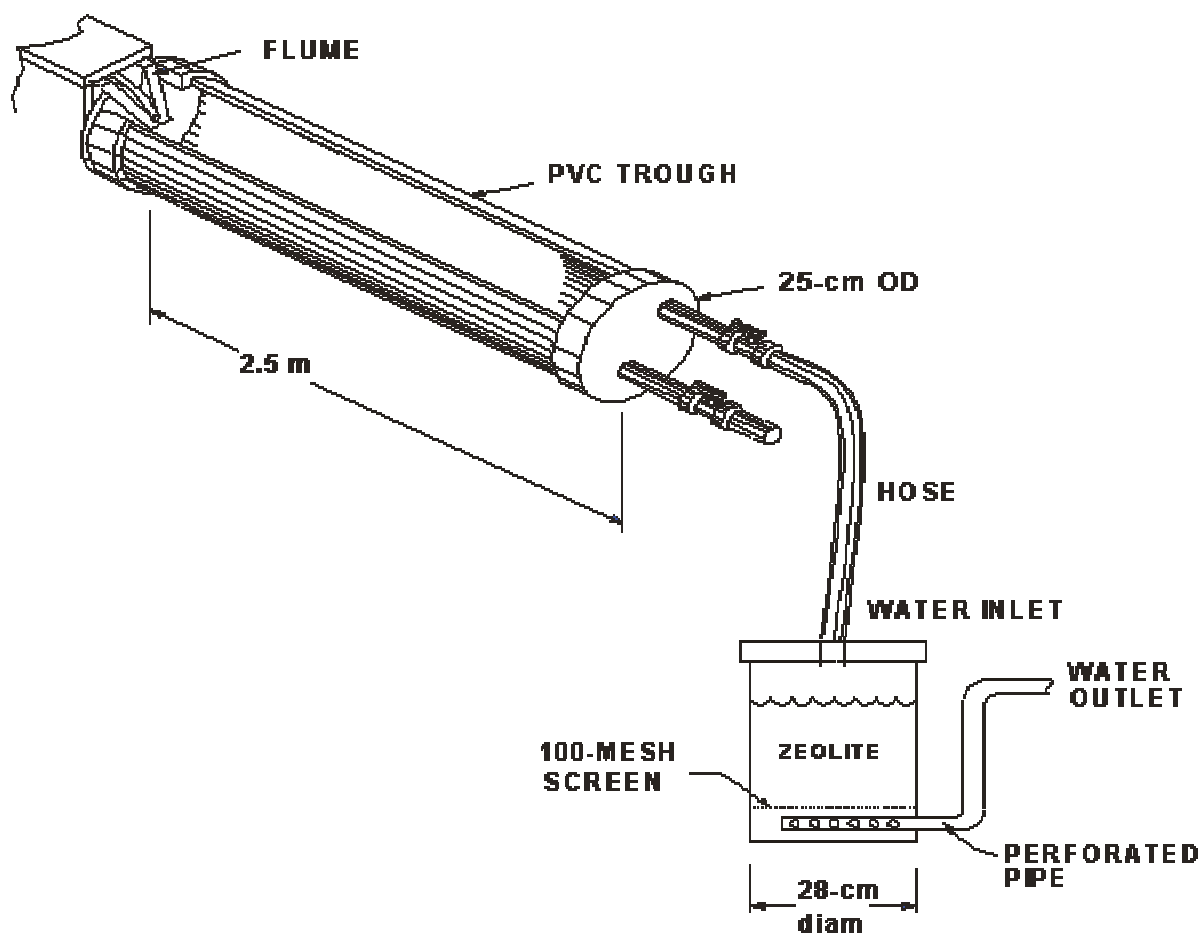


Figure 1. Diagram of the pilot-scale treatment system at Seep C.

Several different configurations were tested before the successful system described above was developed. Because the original concept for the full-scale system was a trench filled with bags of zeolite, this system was pilot-tested first. A trough (made from 10-in. PVC pipe that was cut open to make a trough) was installed at Seep C. The upper end of the trough was placed underneath the flume that had been previously installed to measure the flow rate of the seep water. Water from the seep flowed down through the trough and out either of two valves located in the end of the trough, one near the bottom and the other near the top. Several bags (10-in. OD and 24-in. long) were constructed from geotextile fabric, and each was filled with 13.5 kg of zeolite. Four of these bags were initially placed in the trough. After a few days, the surface of each bag was coated with iron hydroxide, which prevented water from flowing through the geotextile fabric. The iron concentration in the seep water was quite low (0.05 to 0.2 mg/L), but laboratory-scale tests demonstrated that the geotextile fabric promoted precipitation of iron hydroxide on the surface of the fabric. Various attempts were made to reduce the formation of iron hydroxide, including covering the top of the trough with a plastic sheet to reduce air exchange, adding argon gas underneath the plastic, and using a bag filter at the end of the flume

to remove any solids previously formed before the water contacted the zeolite bags. These methods reduced the rate at which the iron hydroxide formed but did not completely solve the problem. About 50% of the ^{90}Sr in the seep water was removed in most of these tests, but much higher removal efficiencies were needed.

In order to eliminate the geotextile fabric bags the zeolite column (bucket) described above was tested. The inlet line of the bucket was connected to the outlet valve of the trough by a rubber hose. In the first test, water entered the bottom of the bucket through a perforated pipe and flowed up through the zeolite and out a hole near the top of the bucket. It was felt that an upflow arrangement would help move any iron hydroxide or other solids out of the zeolite bed. The water repeatedly channeled up through a small part of the zeolite bed, resulting in minimal contact time between the water and the zeolite. Once the channeling started, that part of the zeolite would stay fluidized, providing a low-resistance path for the water, which would perpetuate the channeling. Repeated attempts to startup the column without channeling were unsuccessful. Dye tracer tests showed that the hydraulic residence time for the seep water in the zeolite bed was 10 to 15 seconds. ^{90}Sr removal averaged 30 to 60%. The flow arrangement was then changed to the downflow system described above, which operated successfully for 90 days.

RESULTS OF THE PILOT-SCALE TEST

The downflow column test was started on 3/17/94 and was terminated on 6/15/94, when the construction crew mobilized at the site to build the full-scale treatment system. A total of 63,470 L (3967 bed volumes) of water was treated by the system (bed volumes = water volume/zeolite volume, and is a convenient way to compare the performance of ion exchange systems of different sizes). The flow rate through the system varied considerably, averaging 0.83 L/min. The flow from the seep was generally higher than the flow capacity of the pilot-scale treatment system, so some water bypassed the zeolite by leaking out from under the bucket lid, which was not sealed to the bucket. The outlet pipe of the bucket was located 15 cm lower than the top of the water level in the trough; however, because of the water leakage, the hydraulic head for the system was somewhere between 15 and 4 cm (4 cm is the distance between the top of the bucket and the outlet pipe). Sediment from the seep water slowly accumulated on the top of the zeolite and decreased the flow. In order to maintain flow through the system, it was necessary to remove the sediment almost every day. The top portion of the zeolite bed was stirred up and then the sediment and water were poured off after the zeolite had settled. Several dye tests showed that there was not any significant channeling of water through the zeolite. The system was temporarily turned off several times during the course of the experiment to allow for creek sampling to measure ^{90}Sr flux into Melton Branch from the Seep C area. Starting and stopping the treatment system did not have any noticeable effect on the performance of the system.

The zeolite removed an average of 99.7% of the ^{90}Sr from the first 43,000 L (2688 bed volumes) of seep water treated. The treatment efficiency then decreased slowly, as the zeolite became loaded, until it reached 84.4% by the time the experiment was terminated. For the first 6400 L (400 bed volumes) of water treated, calcium in the seep water was adsorbing on the zeolite and

sodium was being liberated (see Fig. 2); however, after that time strontium was the only ion that was loading on the zeolite. Figure 2 also shows that the apparent total strontium removal was less than the ^{90}Sr removal, which was caused by leaching of the strontium that is naturally present in the zeolite.

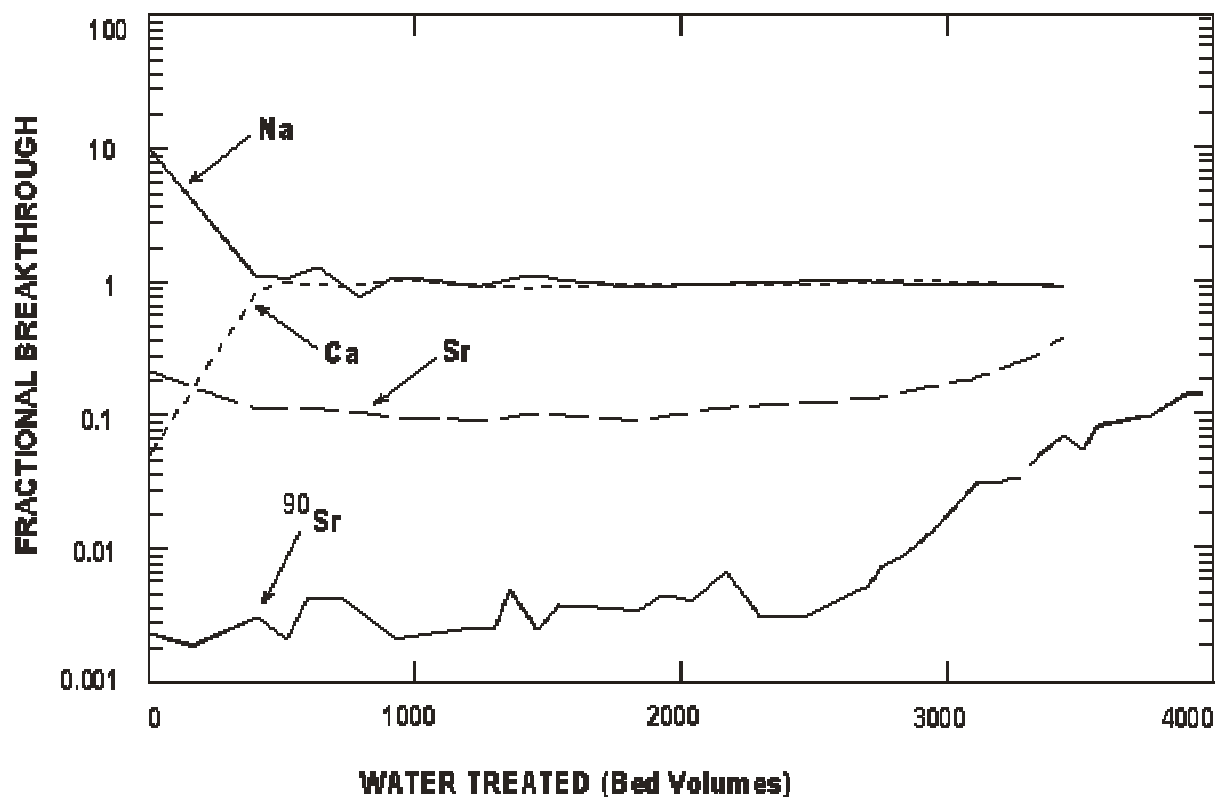


Figure 2. Fractional breakthrough (outlet concentration/inlet concentration) of selected cations for the pilot-scale treatment system at Seep C.

Figure 3 shows the performance of the Seep C system in comparison to two other zeolite systems that have been operated at ORNL. The Wastewater Treatment Test Facility (WTTTF) was a pilot-scale system that used two 7.6-cm ID by 91-cm high columns connected in series, each containing 2.5 L of chabazite zeolite. ORNL process wastewater (see Table II) was pumped through the columns at 0.5 L/min. This wastewater is similar to Seep C water, except that it has a lower ^{90}Sr concentration. The Zeolite Demonstration System (ZDS) was a larger column (0.9 m ID by 2 m tall) filled with 1.3 m³ of chabazite zeolite that was used to treat process wastewater at a flow rate of 75 L/min. Unlike the Seep C treatment system, both the ZDS and the WTTTF operated at constant flow rates, but the composition of the wastewater varied more than that of

the Seep C water. The ZDS was particularly affected by periods of high pH and/or high salt concentrations in the process wastewater, which severely impacted the performance of the zeolite. Because of the lower ^{90}Sr concentration in the process wastewater, the removal efficiencies for the ZDS and WTTF were lower than at Seep C, even though the outlet ^{90}Sr concentrations are similar. Initial ^{90}Sr breakthrough occurred at about 3000 bed volumes for each of these systems.

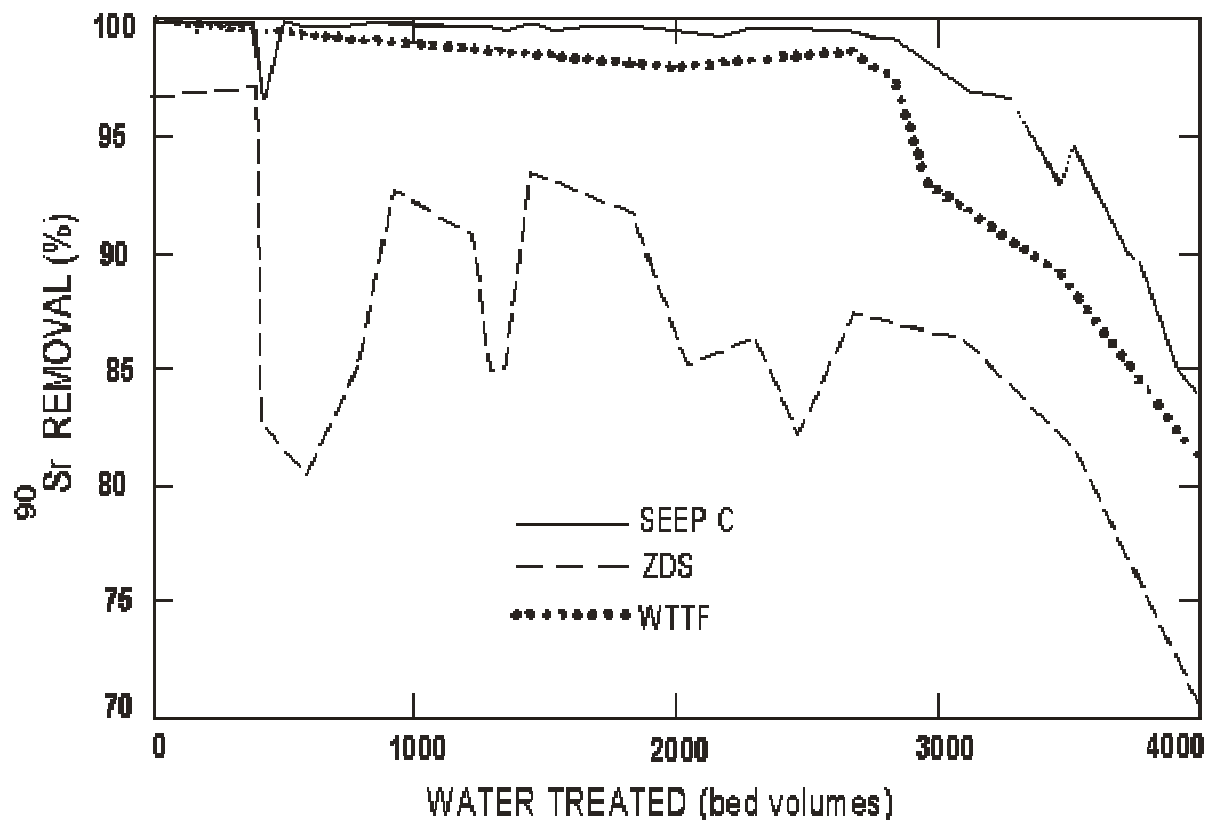


Figure 3. Comparison of the results for the Seep C pilot-scale treatment system with those from the Wastewater Treatment Test Facility (WTTF) and the Zeolite Demonstration System (ZDS).

Table II. Typical Composition of Process Wastewater

Component	Concentration (mg/L)	Component	Concentration (mg/L)
Ba	0.043	Na	64.0
Ca	75.0	Ni	0.024
Fe	2.2	Sr	0.19
Mg	12.0	Zn	0.27
Mn	0.16	⁹⁰ Sr	750 Bq/L
Mo	<0.04	¹³⁷ Cs	110 Bq/L

DESCRIPTION OF THE FULL-SCALE TEST SYSTEM

The full-scale treatment system was the first Environmental Management Project Integrating Contractor (EPIC) Project completed at ORNL. A team consisting of the operating contractor (Martin Marietta Energy Systems, Inc.), the architect-engineer (Ensearch), and the construction contractor (MK Ferguson) worked together to complete the project. DOE approved the project concept, the cost estimate, and the project scope on 3/15/94. Detailed design took from 3/15 to 5/20/94, which overlapped the pilot-scale demonstration previously described. The site plan and access road, the french drain, and the concrete vault that would hold the treatment system were designed first. The details of the treatment system were designed after the pilot-scale system had been operating for about a month. Procurement of long-lead-time items started on 4/26/94. The construction schedule began on 6/9/94 with initial mobilization, and the system was completed on 11/11/94. The total project cost was \$1.4M, with a design cost of \$401,000, procurement and construction costs of \$918,000, and project management and miscellaneous costs of \$95,000. The system started operation on 11/11/94; ahead of schedule and below the original cost estimate of \$1.5M.

The full-scale system consists of a french drain to collect the water and eight 55-gal drums of zeolite (Norit Americas, Atlanta, GA), located in an underground vault, to provide treatment. The zeolite drums are arranged in a lead-lag configuration, with the four lead drums connected to a common header and the four lag drums supplied from a common header on the exit of the lead drums. There is an elevation drop of about 1.5m from the french drain to the zeolite drums, which pushes the water successively through the zeolite, an air lock, and finally into Melton Branch. The flow rate of the effluent from the treatment system and the water level in the french drain are monitored continuously by instrumentation.

RESULTS OBTAINED WITH THE FULL-SCALE SYSTEM

The treatment system typically removes over 99.9% of the ^{90}Sr from the water passing through the drums of zeolite. The system has treated more than 3.3×10^6 L of water since startup, and the average flow rate has been 1.8 L/m. The zeolite has removed over 1.6 Ci of ^{90}Sr from the groundwater. The iron hydroxide in the seep water, which coats the top layer of the zeolite and restricts flow through the treatment drums, has been a continuing problem. When the flow is restricted, the water level in the french drain increases, possibly allowing water to bypass the treatment system. The water level in the french drain has averaged 75 cm. Figure 4 shows a graph of the treatment system flow rate and water level in the french drain since January 1996. Ferrous ions in the seep water can be oxidized by air to ferric ions, which precipitate to form iron hydroxide particles in the water. The system was designed to restrict the movement of air into the seep water, but apparently changes in atmospheric pressure can pump enough air into the french drain to cause some oxidation. The groundwater typically contained 0.1 to 0.2 mg/L Fe during the first year of operation of the full-scale treatment system, but occasional samples since 1996 have shown iron concentrations around 1 mg/L. The reason for this increase is not known.

The layer of iron hydroxide was scraped from the top of each of the first four zeolite drums several times during the first year of operation, which usually restored the flow through the drums. However, the operation was very labor intensive and involved a risk of radiological contamination from handling the radioactive iron hydroxide slurry in the very limited space of the underground vault. During the past three years, the drums of zeolite have been replaced five times, after they became clogged. The old drums of zeolite are currently stored onsite, awaiting final disposal as LLW. The cost of a change-out, including manpower, new zeolite and expected disposal costs is about \$10,000. If the clogging had not occurred, it should have been possible to treat all of the 3.3×10^6 L of water treated to date with the original eight drums of zeolite, so the iron hydroxide plugging has resulted in additional costs of about \$50,000. The new drums typically become clogged again fairly rapidly, so the flowrate through the treatment system is generally quite low. Nitrogen gas, which is being added to the french drain at a flowrate of about 10 L/min, has reduced, but has not eliminated, the iron hydroxide problem.

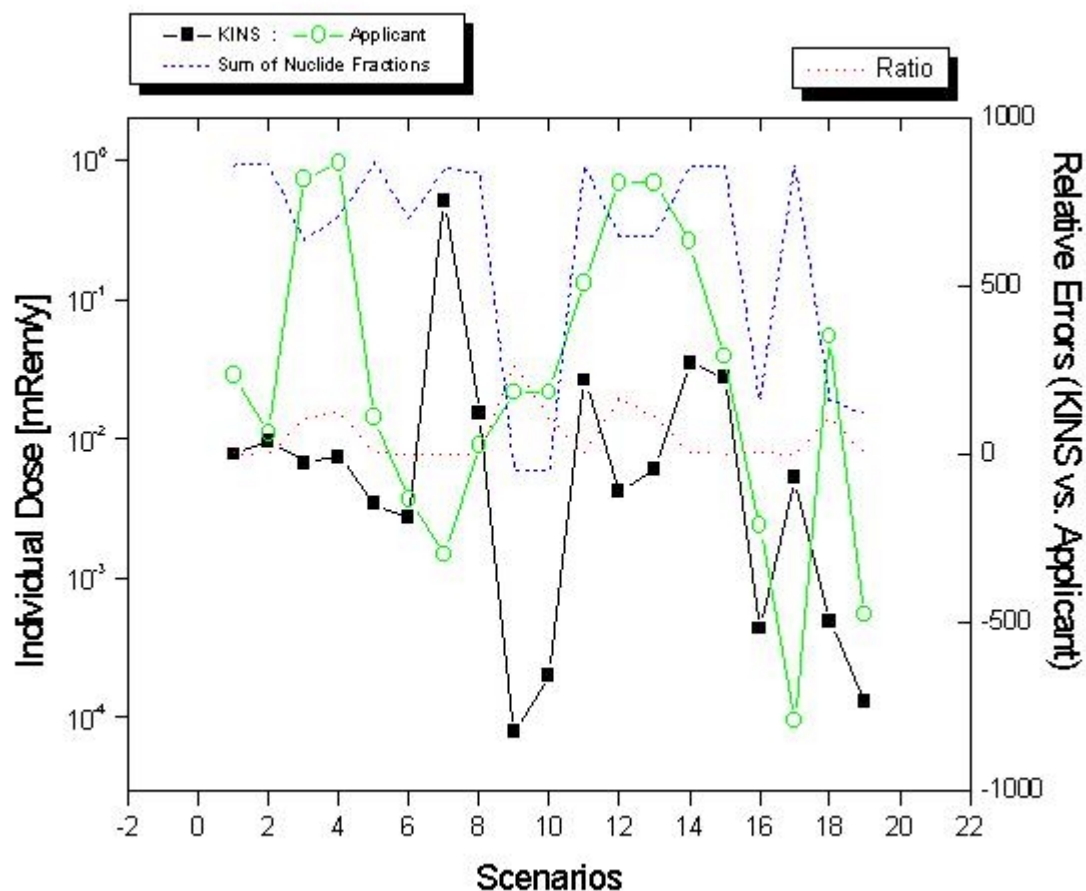


Figure 4. French drain water level and effluent flow rate for the full-scale treatment system at Seep C.

Pilot-scale filtration tests have been conducted at the site to determine whether it would be feasible to pump water from the french drain, through a pressure-filtration system, and then at low pressure through the existing zeolite drums. The tests showed that removal of the very small iron hydroxide particles required a 0.5- μm pore-size filter, which plugged very quickly. It does not appear to be feasible to use a filtration system on the seep water.

CONCLUSIONS

A pilot-scale treatment system at Seep C provided design and operating data for the full-scale system that was constructed. The system treated 63,470 L (3967 bed volumes) of water and removed 22.7 mCi of ^{90}Sr from the seep water. The passive-flow system performed at least as well as comparable pumped zeolite systems in terms of removal efficiency and zeolite utilization. Design and construction of the full-scale treatment system was completed in eight

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months, and the system started operation in November 1994. The system has treated over 3.3×10^6 L of water since startup, and typically removes over 99.9% of the ^{90}Sr from the influent. Iron hydroxide in the seep water, which coats the top layer of the zeolite and restricts flow through the treatment drums, has been a continuing problem.

REFERENCES

1. "Engineering Evaluation/Cost Analysis for the Seep Removal Action at Waste Area Grouping 5 at the Oak Ridge National Laboratory", DOE/OR/02-1217&D1, prepared by Radian Corporation for U.S. Department of Energy, Oak Ridge, Tennessee (1993).
2. S. M. Robinson, "Evaluation of Alternative Flow Sheets for Upgrade of the Process Waste Treatment Plant", ORNL/TM-10576, Oak Ridge National Laboratory (1991).