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**ENVIRONMENTAL
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PROGRAM**

**Development of a Passive-Flow
Treatment System for
⁹⁰Sr-Contaminated Seep Water at
Waste Area Grouping 5 at Oak Ridge
National Laboratory**

P. A. Taylor

MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
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DEPARTMENT OF ENERGY

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⁹⁰Sr-Contaminated Seep Water at Waste Area Grouping 5 at
Oak Ridge National Laboratory

P. A. Taylor

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ABBREVIATIONS

CERCLA	Comprehensive Environmental Response and Liability Act
EE/CA	Engineering Evaluation and Cost Analysis
H-3	tritium
ORNL	Oak Ridge National Laboratory
PWTP	Process Waste Treatment Plant
RI/FS	Remedial Investigation and Feasibility Study
Sr-90	strontium-90
SWSA 5	Solid Waste Storage Area 5
TCLP	toxicity characteristic leaching procedure
WAG 5	Waste Area Grouping 5
WTTF	Wastewater Treatment Test Facility
ZDS	Zeolite Demonstration System

EXECUTIVE SUMMARY

Seep C is a free-flowing stream of groundwater (typical flow of 0.2 to 2 L/min) that emerges in a narrow valley below the old low-level waste disposal trenches in Solid Waste Storage Area 5 (SWSA 5), which is part of Waste Area Grouping 5 (WAG 5). The seep water contains high concentrations of Sr-90 (10,000 to 20,000 Bq/L) and contributes about 25% of all the Sr-90 leaving Oak Ridge National Laboratory. 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 seep water. The goal of the removal action is to have a system operational by November 1, 1994, that reduces the Sr-90 concentration in the water collected and treated by at least 90%.

In order to provide design and operating data for the full-scale system, a pilot-scale system, consisting of a 5-gal bucket with an inlet connection in the lid and a screened outlet on the bottom filled with 16 L of chabazite zeolite, was used to treat the seep water. The test was started on March 17, 1994, and concluded on June 15, 1994. The system treated 63,470 L (3967 bed volumes) of water and 22.7 mCi of Sr-90 from the seep water. The system removed over 99.5% of the Sr-90 from the first 43,000 L of water treated, after which the removal efficiency slowly decreased as the zeolite became loaded until it reached 84% for the final sample. The passive system performed at least as well as comparable pumped zeolite systems in terms of removal efficiency and zeolite utilization. The test was terminated just before the construction crew mobilized at Seep C to build the full-scale system.

1. INTRODUCTION

Seep C is a free-flowing stream of groundwater that emerges 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 enters Melton Branch at the bottom of the small valley. The seep water contains high concentrations of strontium-90 (Sr-90) and tritium (H-3). The Sr-90 concentration ranges from 10,000 to 20,000 Bq/L, with H-3 concentrations of about 100,000 Bq/L. The flow rate of the seep varies considerably, with measured flows ranging from 0 to 59 L/min, but more typical values are 0.2 to 2.0 L/min. Measurement of Sr-90 total flux from the Seep C area into Melton Branch suggests that there is a significant underground component of the flow (about 0.3 L/min) during dry weather. The seep contributes about 25% of all the Sr-90 leaving Oak Ridge National Laboratory (ORNL). Typical chemical data for the seep water is shown in Table 1.

**Table 1. Typical metals concentrations
in Seep C water**

Metal	Concentration (mg/L)
Ba	0.096
Ca	86.0
Fe	0.078
Mg	16.0
Mn	0.22
Mo	0.04
Na	8.2
Ni	0.010
Si	4.1
Sr	0.17
Zn	0.011

SWSA 5 was opened in 1958 and closed in 1973, during which about 86,000 m³ of waste was buried. 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 12 ft wide, about 15 ft deep, and 40 to 500 ft long. The trenches generally run parallel to the slope, so water can easily flow down through the trenches 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. WAG 5 is currently undergoing a Remedial Investigation and Feasibility Study (RI/FS) to determine final closure methods.

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 Analysis¹ (EE/CA)

considered a range of alternatives for treating the water at Seep C. 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 is to have operational by November 1, 1994, a system that reduces the Sr-90 concentration in the water treated by at least 90%. Lab-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 demonstration of the zeolite system was initiated following release of the EE/CA report (December 1993).

2. 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. 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 (see Figs. 1-3). 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, Wash.), an inorganic ion exchange resin. Compared to common organic ion exchange resins, zeolite is more selective for strontium over calcium and magnesium, which are present in high concentrations in the seep water. Zeolite also provides a stable waste form for disposal. Laboratory-scale tests on Seep C water had previously shown that chabazite zeolite was effective in removing the Sr-90 from the water. Since the same zeolite is also used at the PWTP, Waste Operations—which runs the PWTP and will operate the full-scale Seep C facility—is experienced in handling zeolite. 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). It was necessary to locate the treatment system in the floodplain of the creek in order to have enough hydraulic head to force water through the zeolite, so a chain basket was placed around the treatment bucket and attached to a tree to secure the system during high water. Water from Melton Branch covered the treatment system several times during the course of the experiment but did not cause any damage.

Several different configurations were tested before the successful system described above was developed. The original concept for the full-scale system was a trench filled with bags of zeolite, so bags of zeolite were 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 of 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 filled with 13.5 kg each of zeolite. Four of these bags were initially placed in the trough. After a few days, the surface of the bags was coated with iron hydroxide that 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 later lab-scale tests demonstrated that the geotextile fabric promoted iron hydroxide precipitation on the surface of the fabric. Various attempts were made to reduce the formation of iron hydroxide, including covering the top of the zeolite bag with plastic 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.

Fig. 1. Flow diagram of Seep C treatment system.

Fig. 2. Seep C area and treatment system.

Fig. 3. Interior of treatment system.

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. For most of these tests about 50% of the Sr-90 in the seep water was removed, but much higher removal efficiencies were needed.

To eliminate the geotextile fabric bags, the zeolite column (bucket) described above was tested. The inlet line of 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 up-flow 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 start up 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 s. An average of 30 to 60% of the Sr-90 was removed. The flow arrangement was then changed to the downflow system described above, which operated successfully for 90 days.

3. TEST RESULTS

The downflow column test was started on March 17, 1994, and was terminated on June 15, 1994, 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 were treated by the system. (Bed volume = water volume / zeolite volume, and is a convenient way to compare the performance of different-sized ion-exchange systems.) The flow rate through the system varied considerably (see Fig. 4), with an average flow rate of 0.83 L/min. The flow from the seep was generally higher than the flow capacity of the treatment system, so some water bypassed the zeolite by leaking out 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 built up 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 no significant channeling of water through the zeolite. The system was turned off several times during the course of the experiment to allow for creek sampling to measure Sr-90 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 Sr-90 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 (see Fig. 5). 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. 6), but after that, strontium was the only ion that was loading on the zeolite. As Fig. 6 also shows, the apparent total strontium removal was less than Sr-90 removal, which was caused by the slow dissolving of strontium that is naturally present in the zeolite.

Fig. 4. Flow rate variations for Seep C treatment system.

Fig. 5. Removal efficiency for Sr-90.

Fig. 6. Breakthrough of major cations in Seep C treatment system.

Figure 7 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 (WTF) contains two columns 7.6 cm ID by 91 cm high connected in series that are each filled with 2.5 L of chabazite zeolite. Process wastewater is pumped through the columns at 0.5 L/min. The typical composition of process wastewater is shown in Table 2. This water is similar to Seep C water, except for a lower Sr-90 concentration. The Zeolite Demonstration System (ZDS) is a larger pumped column (0.9 m ID by 2 m high) filled with 1.3 m³ of chabazite zeolite that treats process wastewater at a flow rate of 75 L/min. Unlike the Seep C treatment system, ZDS and WTF both operate at a constant flow rate, but the wastewater composition varies more than the Seep C water. Because of the lower Sr-90 concentration in the process wastewater, the removal efficiencies for ZDS and WTF are lower than at Seep C, even though the outlet Sr-90 concentrations are similar. Initial Sr-90 breakthrough occurred at about 3000 bed volumes for each of these systems.

A sample of spent zeolite from the Seep C treatment system was sent to CompuChem Environmental Corp. (Research Triangle Park, N.C.) for toxicity characteristic leaching procedure (TCLP) analysis to determine if the spent zeolite will need to be disposed of as a hazardous waste. The sample was leached using standard EPA extraction procedures, and the leachate was analyzed for volatiles, semivolatiles, pesticides, and metals. The results (Table 3) show that the spent zeolite will not be a hazardous waste, so it can be disposed of at ORNL as low-level waste.

Fig. 7. Comparison of Sr-90 removal efficiency of zeolite treatment systems.

Table 2. Typical composition of process wastewater

Component	Concentration (mg/L) ^a
Ba	0.043
Ca	75.0
Fe	2.2
Mg	12.0
Mn	0.16
Mo	<0.04
Na	64.0
Ni	0.024
Si	2.6
Sr	0.19
Zn	0.27
Sr-90	750 Bq/L
Cs-137	110 Bq/L

^aConcentrations of Sr-90 and Cs-137 in Bq/L.

Table 3. Results of TCLP analysis of loaded zeolite

Contaminant	Concentration (mg/L)	Regulatory limit (mg/L)
Metals		
Arsenic	0.003	5.0
Barium	2.5	100.0
Cadmium	0.005	1.0
Chromium	0.007	5.0
Lead	0.002	5.0
Mercury	0.0002	0.2
Selenium	0.003	1.0
Silver	0.006	5.0
Volatiles		
Vinyl chloride	<0.05	0.20
1,1-Dichloroethene	<0.025	0.70
2-Butanone	0.099	200.0
Chloroform	<0.025	6.0
Carbon tetrachloride	<0.025	0.5
Benzene	<0.025	0.5
1,2-Dichloroethane	<0.025	0.5
Trichloroethene	<0.025	0.5
Tetrachloroethene	<0.025	0.7
Chlorobenzene	<0.025	100.0
Semivolatiles		
Pyridine	<0.05	5.0
1,4-Dichlorobenzene	<0.05	7.5
2-Methylphenol	<0.05	200.0
3-Methylphenol	<0.05	200.0
4-Methylphenol	<0.05	200.0
Hexachloroethane	<0.05	3.0
Nitrobenzene	<0.05	2.0
Hexachlorobutadiene	<0.05	0.5

Table 3 (continued)

Contaminant	Concentration (mg/L)	Regulatory limit (mg/L)
2,4,6-Trichlorophenol	<0.05	2.0
2,4,5-Trichlorophenol	<0.05	400.0
2,4-Dinitrotoluene	<0.05	0.13
Hexachlorobenzene	<0.05	0.13
Pentachlorophenol	<0.25	100.0
Pesticides		
Heptachlor	<0.0002	0.008
Heptachlor epoxide	<0.0002	0.008
Toxaphene	<0.005	0.5
Gamma-BHC (Lindane)	<0.0002	0.4
Chlordane	<0.0006	0.03
Endrin	<0.0003	0.02
P,P'-Methoxychlor	<0.0015	10.0

4. SUMMARY AND CONCLUSIONS

In order to provide design and operating data for the full-scale system that will be constructed at Seep C, a pilot-scale system, consisting of a 5-gal bucket with an inlet connection in the lid and a screened outlet on the bottom filled with 16 L of chabazite zeolite, was used to treat the seep water. The test was started on March 17, 1994, and concluded on June 15, 1994, just before the construction crew mobilized at Seep C to build the full-scale system. The system treated 63,470 L (3967 bed volumes) of water, and removed 22.7 mCi of Sr-90 from the seep water. The system removed over 99.5% of the Sr-90 in the incoming water for the first 43,000 L of water treated, and then the removal efficiency slowly decreased, as the zeolite became loaded, until it reached 84% for the final sample. The passive system performed at least as well as comparable pumped zeolite systems in terms of removal efficiency and zeolite utilization. The results of a TCLP analysis of spent zeolite from the treatment system show that it will not be a hazardous waste and can be disposed of at ORNL as low-level waste.

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, December 17, 1993.

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