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Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1996

Annual Report

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Prepared for
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ABSTRACT

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program, funded by the U.S. Nuclear Regulatory Commission, is (a) studying the degradation effects in organic ion-exchange resins caused by radiation, (b) examining the adequacy of test procedures recommended in the Branch Technical Position on Waste Form to meet the requirements of 10 CFR 61 using solidified ion-exchange resins, (c) obtaining performance information on solidified ion-exchange resins in a disposal environment, and (d) determining the condition of liners used to dispose the ion-exchange resins.

During the field testing experiments, both portland type I–II cement and Dow vinyl ester-styrene waste form samples were tested in lysimeter arrays located at Argonne National Laboratory-East (ANL-E) in Illinois and at Oak Ridge National Laboratory (ORNL). The study was designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over an extended period. Those experiments have been shut down and are to be exhumed. This report discusses the plans for removal, sampling, and analysis of waste form and soil cores from the lysimeters. Results of partition coefficient determinations are presented, as well as application of a source term computer code using those coefficients to predict the lysimeter results. A study of radionuclide-containing colloids associated with the leachate waters removed from these lysimeters is described. An update of upward migration of radionuclides in the sand-filled lysimeter at ORNL is included.

Job Code A6876—Field Lysimeter Investigations:
Low-Level Waste Data Base Development Program

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EXECUTIVE SUMMARY

Research is being conducted at the Idaho National Engineering and Environmental Laboratory on materials from four liners containing organic and inorganic ion-exchange resin under three tasks of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program.

For resin solidification, portland type I–II cement and vinyl ester-styrene (VES) waste form specimens incorporating ion-exchange resin waste were periodically subjected to the tests specified in the "Technical Position on Waste Form" issued by the U.S. Nuclear Regulatory Commission. Waste form sample performance data were obtained as a result of the work, and the results were compared to data obtained from similar waste form specimens tested earlier in the program. That information was reported in a NUREG/CR report this fiscal year.

Field testing consists of examining the effect of disposal environments on solidified resin wastes. The purpose of this task, using lysimeter arrays at Oak Ridge National Laboratory (ORNL) in Tennessee and Argonne National Laboratory-East (ANL-E) in Illinois, is to expose samples of solidified ion-exchange resin to the actual physical, chemical, and microbiological conditions of a disposal environment. The study, which ran for 10 years before shutdown occurred this fiscal year, was designed so that continuous data on chemical species and nuclide release and movement, as well as environmental conditions, were obtained. Each month, data were retrieved from the data acquisition system. At least quarterly, water was drawn from the porous cup soil-water samplers and from the lysimeter leachate collection compartment. Those water samples were analyzed for chemical species and beta- and gamma-producing nuclides.

Since shutdown, planning has been under way to remove the waste form samples and extract soil samples from the lysimeter soil columns. Those tasks will be accomplished by soil coring. Waste forms will be characterized and degradation determined. Soil samples will be analyzed for radionuclide movement and the presence of microbes.

Lysimeter soils and sand partition coefficients (K_ds) were determined by laboratory analysis. Those analyses and the results are presented.

Leachates from both ORNL and ANL-E lysimeters were examined for radionuclide-containing colloids. That study is described and the results given. This was an extension of previous work done on this program.

Final results of the study of upward migration of radionuclides in the sand-filled lysimeter at ORNL are discussed.

A source term computer code is used to model the release of radionuclides from the lysimeter waste form samples using the newly developed K_ds. Also, comparisons of code prediction to actual lysimeter data have been made.

ACRONYMS AND ABBREVIATIONS

ANL-E	Argonne National Laboratory-East
ASTM	American Society for Testing and Materials
CFR	Code of Federal Regulations
DAS	data acquisition system
DUST	Disposal Unit Source Term
EDX	energy dispersive x-ray
INEEL	Idaho National Engineering and Environmental Laboratory
NAA	neutron activation analysis
NRC	U.S. Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
SEM	scanning electron microscopy
VES	vinyl ester-styrene
XRF	x-ray fluorescence analysis

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INTRODUCTION

The March 28, 1979 accident at Three Mile Island Unit 2 released approximately 560,000 gal of contaminated water to the auxiliary and fuel handling buildings. The water was decontaminated using a demineralization system called EPICOR-II developed by Epicor, Inc.[a] The contaminated water was cycled through three stages of organic and inorganic ion-exchange media. The first stage of the system was designated the prefilter, and the second and third stages were called demineralizers. After the filtration process, the ion-exchange media in 50 of the prefilters contained radionuclides in concentrations greater than the limits for low-level wastes. These prefilters were transported to the Idaho National Engineering and Environmental Laboratory (INEEL) for interim storage before final disposal. A special overpack (high-integrity) container was developed during that storage period to dispose of the prefilters at a commercial disposal facility in the State of Washington. As part of the EPICOR and Waste Research and Disposition Program funded by the U.S. Department of Energy, 46 prefilters were disposed. Four prefilters used in U.S. Nuclear Regulatory Commission (NRC) studies were stored in temporary storage casks outside the Hot Shop of Test Area North Building 607 at the INEEL. Those four prefilters were disposed during this reporting year at the Radioactive Waste Management Complex on the INEEL Site.

Under the EPICOR and Waste Research and Disposition Program, continuing research has been conducted by the INEEL on materials from those EPICOR-II prefilters.[1,2] That work is now funded and directed by the NRC as part of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program. Studies were conducted on organic ion-exchange resins from selected prefilters. The resins were examined to measure degradation, tests are being performed to characterize solidified ion-exchange media, and experiments are being conducted to field-test solidified wastes using lysimeters.

The results of resin degradation from studies of the first and second sampling, as described in References 3 and 4, were compared with those of the third sampling described in Reference 5. The degradation studies determined the acceptability of EPICOR-II prefilters for disposal in high-integrity containers at the commercial disposal site at Hanford, Washington by identifying (a) degradation effects on the ion-exchange resins caused by contained radiation and (b) the possible release of contained radionuclides from ion-exchange resins. Those studies are complete and are not reported here.

Another aspect of this program was investigated—the solidification of EPICOR-II wastes from prefilters PF-7 and PF-24 using portland type I–II cement and vinyl ester-styrene (VES), a proprietary solidification agent developed and supplied by the Dow Chemical Company.

The formulations used for the immobilization of EPICOR-II wastes were developed to produce waste form samples meeting the regulatory requirements of 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste." The NRC, in its "Technical Position on Waste Form, Rev. 1,"[6] provides guidance to waste generators on waste form sample test methods and acceptable results for compliance with the waste

a. References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof.

form sample requirements of 10 CFR 61. In this study, EPICOR-II waste form samples were subjected to the recommended tests of Reference 6 (reported in References 7 and 8) and, on an annual basis, were subjected to the specified compression test to ensure compliance with stability requirements. The data as reported in Reference 9 indicated that the cement waste form sample strength had stabilized after increasing with age for 8 years. In the case of VES, the waste form sample strength had begun to decrease.

Solidified waste form samples containing EPICOR-II ion-exchange resin waste were field-tested using lysimeters. The objectives of the field testing task were to (a) examine the performance of the waste form samples in typical low-level waste disposal environments, (b) compare field results with short-term laboratory leach studies, (c) compare field results with U.S. Department of Energy Special Waste Program field test results, (d) develop a low-level radioactive waste field leach-rate data base for use in performance assessment source term calculations, and (e) use the Disposal Unit Source Term (DUST) computer code to compare predicted cumulative radionuclide release to actual field data.

The intent of the testing was to expose waste form samples to the physical, chemical, and microbiological environment of typical disposal sites in the eastern United States (References 1 and 2). The lysimeters were expected to monitor the release of nuclides from buried waste form samples and provide data that accurately determine movement as a function of time and environmental conditions. Emphasis was placed on investigating the requirements of 10 CFR 61. The study was designed so that continuous data on nuclide release and movement, as well as environmental conditions, were obtained over the test period. The experiment was shut down at the beginning of fiscal year 1996.

This report contains a description of the waste form and soil core sampling plans to be implemented on these shut-down lysimeter experiments. Soil coring will be employed to remove the waste forms from the lysimeters as well as to obtain soil cores. The waste forms will be characterized to determine the extent of degradation after 10 years in a disposal environment. Soil samples will be analyzed to obtain a map of radionuclide movement through the soil column in each lysimeter.

Methods used and results of laboratory determinations of partition coefficients associated with the soils and inert sands at each experimental site are presented.

Colloid studies were carried out on leachates removed from the lysimeter at each site that was experiencing the highest concentration of radionuclide release to the leachate collector. The examinations are described and the results included in this report.

Final results of the study of upward migration of radionuclides in the sand-filled lysimeter at ORNL are presented.

This report contains a comparison of results of application of a source term code used to compute release of radionuclides from the lysimeter waste forms to actual releases into leachate collectors observed over the 10 years of operation. The calculations apply the partition coefficients determined in the laboratory.

RESIN SOLIDIFICATION

As part of the resin solidification project, EPICOR-II waste form samples solidified with portland type I–II cement and VES were periodically subjected to compression testing per ASTM C39[10] over a 12-year time span. The samples were tested dry using sulfur leveling caps poured per ASTM C617.[11] Normally, one sample of each type of ion-exchange resin waste form (all organic and organic resins with zeolite) in each solidification agent (cement and VES) are tested. On August 9, 1995, eight 12-year-old waste form samples were compression-tested using a Tinuis Olsen 60,000-lb testing machine. The samples had mass and contact radiation dose readings similar to those tested in 1984.[7,8] The test results were presented in Reference 9, which was published in this fiscal year.

Both cement waste form types exhibited an increase in strength with age and self-irradiation dose over the 12 years of this study. After a buildup over the first 8 years, that increase seemed to have stabilized at about 5,500 psi for both cement types. In the interval from 7 through 11 years, the cemented organic/inorganic resin waste form sample data were scattered very high and low. However, the cemented all-organic resin waste samples exhibited more consistent results, with one low point at 11 years. Comparing these data to the externally irradiated compressive strength data obtained in 1985 indicates that the strength of the cemented wastes will slowly decrease with further age to about 3,000 psi after 300 years as predicted by end-of-life testing. This value is well above the 500-psi minimum recommended by the Technical Position, Rev. 1.

Both VES waste form types also exhibited an increase in strength with age and self-irradiation dose through 8 years. The maximum strength of the VES samples was between 4,500 and 5,000 psi. At 11 years, strength had significantly decreased with the organic/inorganic resin waste form sample (20%). At 12 years, both types of waste form samples suffered a further strength decrease (16% for all-organic and 11% for organic/inorganic resin wastes). Comparing these data to those of the externally irradiated end-of-life test compressive strength data obtained in 1985 confirms that the VES sample strength will continue to decrease with age to about 2,000 psi. This value is also well above the 60-psi minimum required for VES by the Technical Position, Rev. 1.

The total beta/gamma exposure dose experienced by the 12-year-old waste form samples was between 0.6E+6 and 3.1E+6 rad. That is two orders of magnitude less than the 300-year end-of-life doses received by the externally irradiated samples of earlier INEEL tests, and these aged samples had not reached the low strength exhibited by those earlier externally irradiated samples.

FIELD TESTING

Materials and Methods

Experiment Description

Solidified waste forms containing EPICOR-II ion-exchange resin waste were field-tested using lysimeters. Lysimeter sites were established at Oak Ridge National Laboratory (ORNL) and Argonne National Laboratory-East (ANL-E). Instrumentation within each of the five lysimeters at each site includes porous cup soil-water samplers and soil moisture/temperature probes. The probes were connected to an onsite data acquisition system (DAS), which also collected data from a field meteorological station located at each site. A detailed description of the lysimeters and their installation is presented in Reference 12, while data from the 10 years of operation are contained in earlier reports.[13-22] Those experiments were shut down and are awaiting waste form removal and soil sampling.

Description of Waste Forms

Waste forms used in the field test are composed of solidified EPICOR-II prefilter resin wastes. Two waste types were used in the solidification project. One is a mixture of synthetic organic ion-exchange resins from prefilter PF-7 (phenolic cation, strong acid cation, and strong base anion resins), and the other is a mixture of synthetic organic ion-exchange resins from prefilter PF-24 (strong acid cation and strong base anion resins) with an inorganic zeolite. PF-7 waste contains 5% Sr-90, while PF-24 waste contains about 1% Sr-90. Of the other radionuclides in those wastes, Cs-137 and Cs-134 are the major constituents, with traces of Co-60 and Sb-125 included.

Portland type I-II cement and VES were used to solidify both types of resin wastes. Individual waste form samples were manufactured by allowing a mixture of solidification agent and resin waste to solidify in polyethylene molds that were 4.8 cm in diameter by 10.2 cm high. Enough of the mixture was added to each vial to produce samples with an average diameter of 4.8 cm and a height of 7.6 cm (137.5 cm^3). Each lysimeter contains seven of these 4.8 × 7.6-cm waste form samples stacked end-to-end to form a 1-L waste volume. Table 1 shows the types of samples placed in the lysimeters. A complete description of waste form manufacture is given in Reference 7. Bench testing of similar waste forms, per the requirement of the Technical Position, Rev. 1, is described in Reference 8.

Description of Lysimeters

The lysimeters were designed as self-contained units that can be easily disposed at the termination of the field test experiment. A total of 10 lysimeters were used, with five placed at each field site. Each lysimeter is a right-circular cylinder (0.91 m in diameter by 3.12 m in height), constructed of 12-gauge, 316 L stainless steel (Figure 1). Internally, the lysimeter is divided into two sections, the upper volume being 1,532 L and the lower volume being 396 L. A 3.8-cm, Schedule 40 stainless steel tube provides access to the lower compartment. The upper compartment of each lysimeter contains the soil column with waste forms, three temperature/moisture probes, and five soil moisture cups. The cups are numbered 1 through 5 as noted in Figure 1. The lower compartment serves as a leachate collector, which is emptied and sampled through the 3.8-cm tube.

Table 1. Lysimeter waste form composition.

Lysimeter Number	Fill Material	Waste Form Description
1	Soil	Cement with PF-7 resin waste
2	Soil	Cement with PF-24 resin waste
3	Soil	VES with PF-7 resin waste
4	Soil	VES with PF-24 resin waste
5 ANL-E	Silica oxide	Cement with PF-7 resin waste
5 ORNL	Silica oxide	Cement with PF-24 resin waste

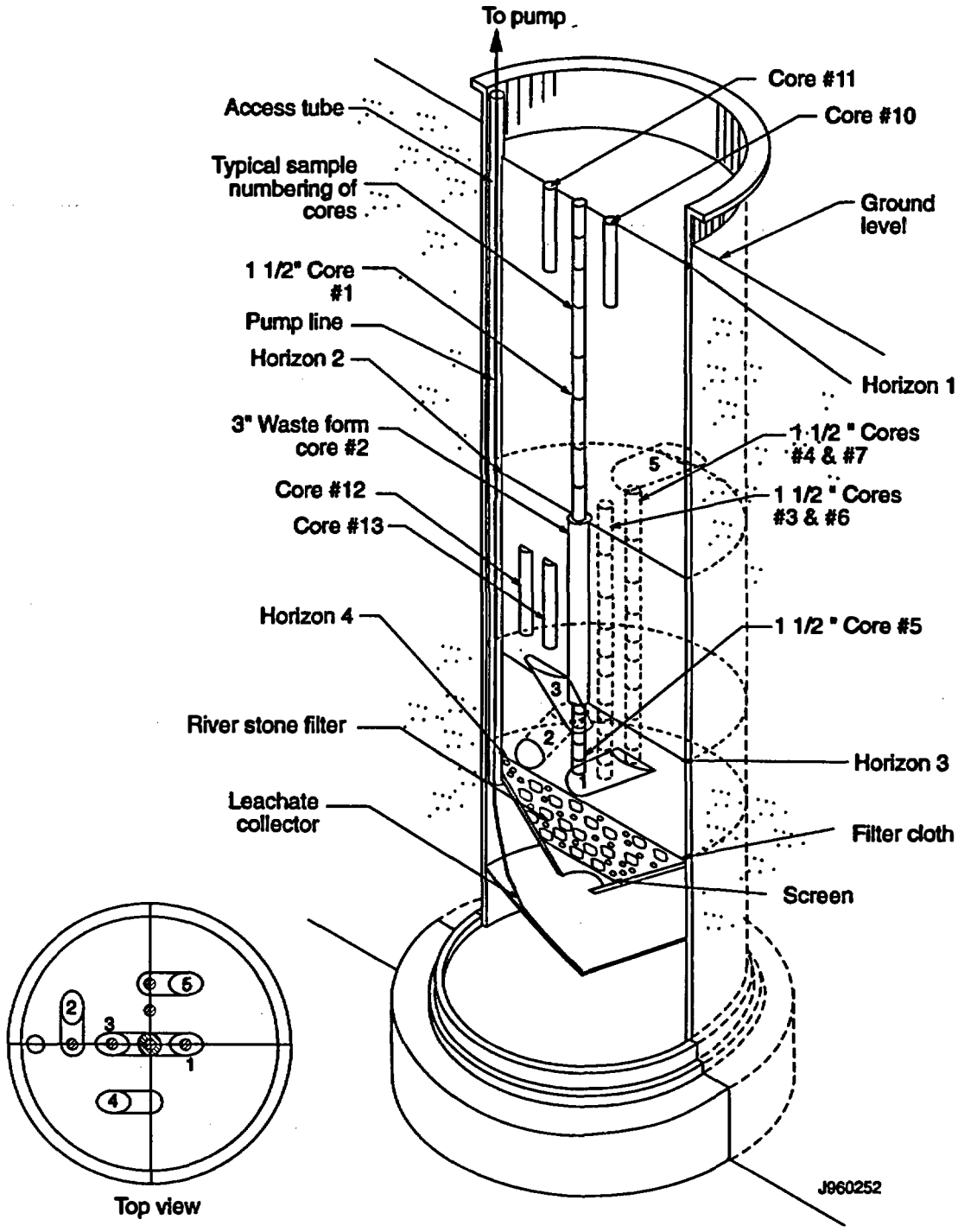


Figure 1. Isometric drawing showing the lysimeter experiment, cores, and samples.

Four lysimeters at each field site (numbered 1 through 4) are filled with soil; the remaining one (number 5) is a control lysimeter filled with an inert silica sand (Reference 12). Two different soils were used. One was representative of Midwestern soils; the other was intended to approximate soil found at Barnwell, South Carolina. ANL-E used local indigenous soil that fits NRC criteria for Midwestern soil. It is a Morley silt loam with the surface layer removed. The resulting subsurface soil is a clay loam. Soil at ORNL was not found to be a suitable substitute for Barnwell soil; therefore, acceptable soil (a clay sand[12]) was transported to ORNL from the Savannah River Plant adjacent to the Barnwell facility in South Carolina.

Soil temperature and moisture sensors are physically located within a common housing or probe. These probes are located at three elevations (149, 77.9, and 28.8 cm from the bottom of the soil column) within each lysimeter. The function of these probes was to provide data on whether or not the buried waste forms experienced freezing temperatures and if the surrounding soil was moist. Because all of the soil lysimeters at each site were exposed to the same environment, the placement of probes provided a planned redundancy in data collection.

Data Retrieval and Analysis

Electrical impulses from the environmental instruments were collected by, processed in, and stored by the DAS for periodic retrieval. The DAS processed input into recognizable data using programmable steps. Output from the soil moisture probes, for example, was processed by a polynomial equation that was derived from laboratory calibration of the probes (Reference 12).

Data output from the DAS was stored on a cassette tape and was then retrieved and translated to an IBM PC-compatible disk file. These files were printed either as graphs or in an alphanumeric format.

Water from each lysimeter was drawn from porous cup soil-water samplers and lysimeter leachate collection compartments at least quarterly. These water samples were analyzed routinely for gamma-producing nuclides and, as required, for the beta-producing nuclide Sr-90.

Both experiments were shut down at the beginning of this fiscal year. Covers were fabricated and bolted down to the top of the vessels after each leachate collector had been drained of leachate. This precluded the entry of any precipitation and provided an opportunity for the soil columns to drain. Also, at that time, the DASs were disconnected and moved to an indoor storage facility.

Continuing Studies

Lysimeter Waste Form and Soil Core Sampling

The primary objective of the waste form and soil sampling will be to obtain the entire waste form from each of the five lysimeters at each site in a cylindrical core, which could be transported to the INEEL for detailed examination. Secondary objectives will be to extract soil cores, soil microbial samples, selected moisture cups, filter fabric samples, and filter support stone samples. Seven soil cores will be taken per lysimeter, one for the waste form and six for soils. Four soil grab samples, one filter cloth cut sample, and one filter support rock grab sample per lysimeter are planned. Moisture cup numbers 1 and 3 from each lysimeter also are to be collected.

A diagram of the sample locations and sizes is shown in Figure 1, and Table 2 lists the cores and samples. The waste form cores (number 2) will be 7.5 cm in diameter and 58.5 cm long and extend from horizon 2 to 3. That length contains all seven waste form samples. The soil cores and microbial soil samples will be 3.3 cm in diameter and various lengths. All will be taken with coring tools made up of 25-cm-long segments. Cores number 1 will be taken with a 75-cm top segment, then a 25-cm-long bottom segment. All other cores are to be taken at full depth. The microbial soil samples will also be taken with the 3.3-cm-diameter tool using one segment. All cores will be contained in plastic, cylindrical core tool liners that will be closed with plastic caps on the ends.

Radial and vertical position of the coring tools is to be controlled during coring operations by use of special guide plates and bushings. The coring tools and tips were specifically designed for this

Table 2. Lysimeter waste form, soil, and other samples.

Sample Designation	Number of Samples/ Makeup	Sample Length (cm)	Planned Analysis
Core Number 1	8	12.5	Radionuclide
Core Number 2	1	54.5	Radionuclide
	1	4.0	Radionuclide
Core Number 3	4	12.5	Radionuclide
	1	7.5	Radionuclide
Core Number 4	4	12.5	Archival
	1	7.5	Archival
Core Number 5	5	6.3	Radionuclide
Core Number 6	6	6.3	Radionuclide
	1	7.5	Radionuclide
Core Number 7	3	12.5	Archival
	1	7.5	Archival
Sample Number 8	Filter cloth	NA	Radionuclide
Sample Number 9	Rock	NA	Radionuclide
Core Number 10	Soil	25	Archival
Core Number 11	Soil	25	Microbial
Core Number 12	Soil	25	Microbial
Core Number 13	Soil	25	Microbial
Sample Number 14	Soil	NA	Microbial
Sample Number 15	Waste form scraping	NA	Microbial
Sample Number 16	Moisture cup 3	NA	Radionuclide
Sample Number 17	Moisture cup 1	NA	Archival

task by Art's Manufacturing and Supply of American Falls, Idaho. The tool driver was a Bosch rotary hammer. The INEEL designed the special core tool guide system.

Radiochemical characterization, including full-length gamma scanning of each seven-sample waste form, will provide information on the remaining waste form radionuclide inventory. Each set of seven waste form samples will have one or two samples radiochemically analyzed. That random analysis combined with full-length gamma scan will be used to determine the inventory of each waste form. Waste form physical condition will be determined by visual examination, weighing of all samples, and compressive testing of three samples from each waste form. Soil cores will be radiochemically analyzed for nuclide content, as will the filter cloth, filter support stone, and moisture cups. These data will then be used to determine radionuclide material balance within each lysimeter soil column, radionuclide pathways through the soil columns, and radionuclide holdup factors of the various components of the lysimeter systems. Other soil samples will be examined for microbial activity, which may then be related to waste form physical condition indirectly or more directly by microbial examination of waste form surface scraping samples.

Partition Coefficients of Geomaterials Used in Lysimeters

Introduction. The ratio, at steady state, of the concentration of a contaminant on the solid to its concentration in the leachate water defines the partition coefficient, K_d. In this case, the K_d value is the radioactivity associated with the solid phase (as cpm/g) divided by the activity in the liquid (as cpm/mL). It is a critical parameter that describes the degree to which a sediment will retain or immobilize a contaminant. The K_d can be determined from a single experiment, or the data for a set of experiments at different tracer concentrations can be plotted to give an isotherm. An isotherm is a plot of the contaminant concentration sorbed on the solid versus its concentration remaining in the liquid at steady state. The advantage to this approach is the acquisition of information on the effect that tracer concentration has on K_d.

Materials. Partition coefficients, through the use of isotherms, were developed for the various geomaterials used in the lysimeters. The materials investigated were:

1. Soil from the ORNL lysimeters (designated ORNL soil)
2. Soil from the ANL-E lysimeters (designated ANL-E soil)
3. Sand from the ORNL control lysimeter (designated ORNL sand)
4. Sand from the ANL-E control lysimeter (designated ANL-E sand)
5. Silica flour used as packing around moisture cups.

Both soil samples, as received, contained a large percentage of coarse but friable aggregate. This was gently disaggregated by very light action with a mortar and pestle. The material was then passed through a 0.84-mm (#20) sieve. Only a small percentage was retained on this sieve: 9.52% and 18.35% for ANL-E and ORNL soils, respectively. The material that passed through the sieve was used for the sorption experiments. Rainwater, collected at each site, was used as the contact solution. To this rainwater, tracers containing cesium and strontium were added.

Experiments were conducted with three different sets of tracer solutions. One solution contained elemental cesium at five different concentrations. The other two solutions contained Cs-137 and Sr-85, both containing elemental carriers, at different concentrations. This was required by the great difference in sorption capacity between the soils and the sand and silica samples. Each experiment consisted of tracer solutions containing Cs-137 and Sr-85 in rainwater from the same site from which the soil or sand were obtained.

Methods. Kinetics experiments were performed for the two soil samples to determine an appropriate amount of time for the sorption isotherm experiments to run. For each sample, two grams of soil and 20 mL of tracer solution were weighed into a polyethylene centrifuge tube. These were placed on a mixing table and sampled six times over a period of 12 days. The samples were analyzed on an intrinsic Ge gamma counting system. Count rates were determined (corrected for mass) and compared to count rates of reference samples of the tracer solution.

Isotherm experiments were conducted using a set of four concentrations (five for the elemental cesium experiment) of the stock tracers. Two grams of solid (soil, sand, or silica flour) were weighed into plastic bottles, and 20 mL of tracer were added. These were placed on a shaker table for 12 days and sampled six times over that period. The quantity sorbed was determined by the difference from a set of reference tracer solutions. A similar set of experiments using elemental CsCl solution was also conducted. In this case, the solution was analyzed for cesium by atomic absorption spectrometry.

Sorption Kinetics Results. Sorption of cesium and strontium on the two soils is initially rapid with most uptake occurring in the first 1.5 hours. After that time, a small percentage (about 4%) of strontium desorbed, probably in response

to changing pH and other chemistry in solution. The cesium continued to be sorbed slowly over the 12-day experiment. Given the time constraints of this project, 12 days was taken as the optimum time for the isotherm experiments.

Elemental Cesium Results. Isotherms for the elemental cesium experiments are non-linear, implying that sorption takes place on a wide array of sites having different sorption energies. This is expected for natural materials containing many minerals. As a result, as more cesium is available, the fraction sorbed becomes less and less. The concentrations in these experiments are very high, relative to radioactive contamination. This was necessary to observe cesium in solution. Both isotherms can be fitted with a Freundlich isotherm equation of the form

$$S = aC^b \qquad (1)$$

where

S = concentration on the solid

C = concentration in the liquid at steady state

a and b = fitting factors where b is constrained between 0 and 1.

The ORNL soil isotherm for elemental cesium was fit with a = 4.43 and b = 0.68. The first three points on the plot (Figure 2) are linear and pass close to the origin, possibly showing that the isotherm is close to the Henry's Law regime in which the isotherm is linear at low sorption fractions. If this is the case, the slope equals the K_d and is 37 mL/g at low loadings. The ANL-E soil isotherm was also non-linear and could be fit with a Freundlich equation. In this case, a = 6.19 and b = 0.46. This soil has a much greater sorption capacity than the ORNL soil. To use this equation to determine K_d in the lysimeters, the concentration of cesium in the pore fluid (i.e., porous cup and leachate collector samples) must be known. For the ANL-E soil, estimating from the slope of the two lowest points and the origin, we obtained an initial K_d of 180 mL/g for cesium. The isotherms for elemental cesium were constrained by detection limits for cesium; consequently, an additional set of isotherms were developed for Cs-137 (plus carrier) and Sr-85.

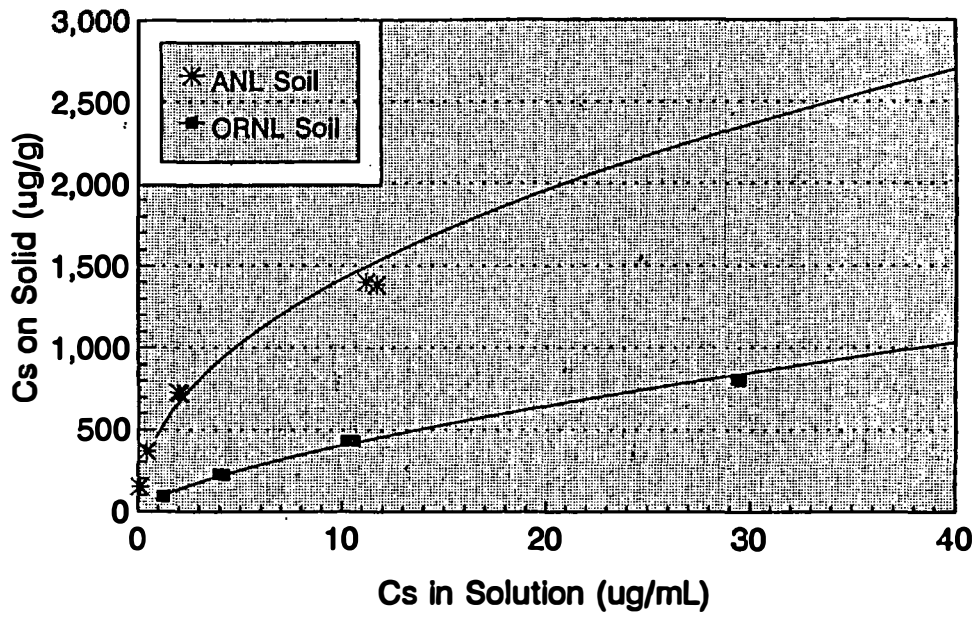
Radioactive Tracer Results. Results for all of the sorption experiments involving Cs-137 and Sr-85 are shown in Table 3. Cesium sorbs much more readily than does strontium, and both elements have discernable K_d values for all materials including the sand and silica flour. The two soils, especially the ANL-E soil, have high capacities for cesium. Both the Cs-137 and Sr-85 tracers used in these experiments contained carriers (the cesium carrier was quite concentrated in order to provide sufficient cesium in solution, after sorption, to allow analysis). Consequently, we have calculated results in terms of μg of cesium per gram of solution or per gram of solid. When isotherms are non-linear, the total concentration of that element in solution (at steady state with the solid phase) must be known, including the concentration of the stable element that is naturally present. This can be applied to the lysimeter study by converting the activity per mL to microgram quantities through the specific activity of the various radionuclides of cesium and strontium in the waste. In addition, the concentration of stable cesium and strontium in the water that passed through the lysimeter should be known. In this way, the position along the isotherm can be determined, from which K_d is calculated.

All of the isotherms for Cs-137 can be described by the power regression equation, which is equivalent to a Freundlich isotherm. Results are shown in Figures 2 through 4. The isotherm is non-linear and is described by two terms, one being an exponent, as shown in Equation (1). This being the case, partitioning is related to the concentration of cesium in solution after equilibration with the solid. Only the results for the silica flour can be reasonably fit to a linear function (Figure 4). With the Freundlich isotherm (i.e., power regression), the K_d is a function of the total concentration of that element in solution. Therefore, it is necessary to know not only the concentration of radioactive strontium and cesium in solution but also the elemental

Table 3. Partition coefficients (mL/g) and Freundlich parameters.

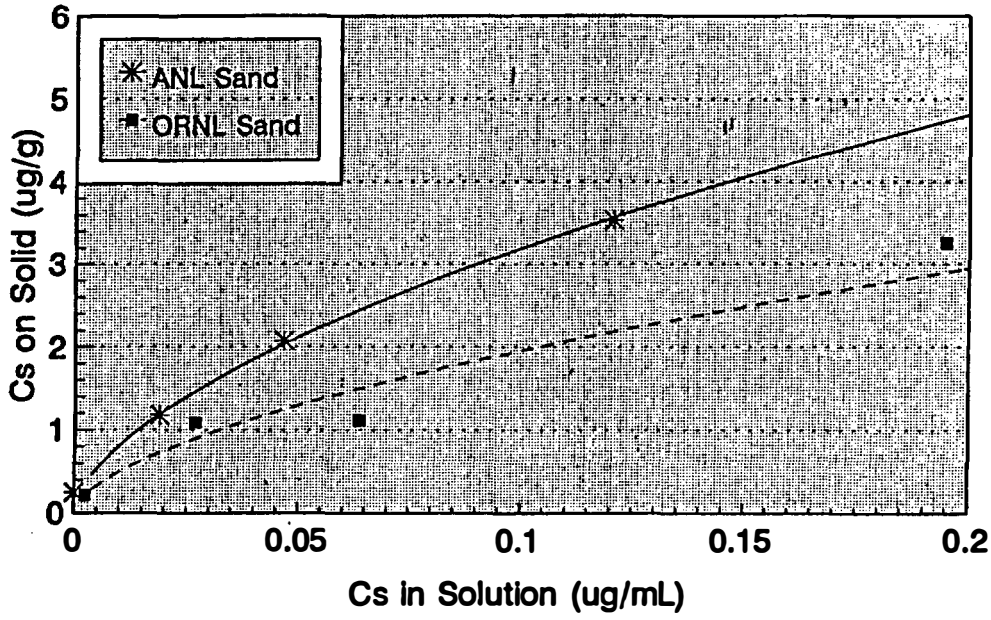
Material	Cesium Form[a]	Freundlich Parameters for Cesium	Strontium Form	Freundlich Parameters for Strontium
ANL-E Soil	Freundlich Linear	a = 6.19, b = 0.46 K_d = 390	Linear	K_d = 81
ORNL Soil	Freundlich Linear	a = 4.43, b = 0.68 K_d = 37	Freundlich Linear	a = 2.44, b = 0.67 K_d = 20 - 40
ANL-E Sand	Freundlich Linear	a = 2.53, b = 0.59 K_d =55	Linear	K_d = 6.4
ORNL Sand	Freundlich Linear	a = 2.03, b = 0.59 K_d = 40	Linear	K_d = 6.3
ANL-E Silica	Freundlich Linear	a = 2.79, b = 0.74 K_d = 42	Linear	K_d = 2.2
ORNL Silica	Freundlich Linear	a = 2.86, b = 0.76 K_d = 49	Linear	K_d = 2.1

a. Linear estimate represents the K_d at low concentrations in the groundwater.



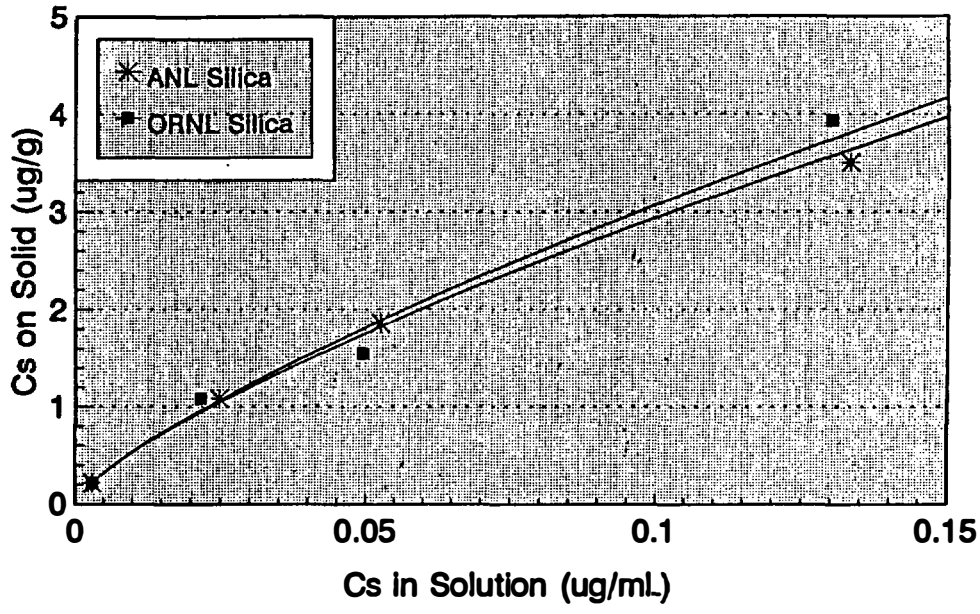
ANL Soil: Power Regression, a = 6.19 b = 0.463
ORNL Soil: Power Regression, a = 4.43 b = 0.681

Figure 2. Cesium-137 (plus carrier) isotherms for soils from the ANL-E and ORNL lysimeters.



ANL Sand: Power Regression, a = 2.53 b = 0.593
ORNL Sand: Power Regression, a = 2.033 b = 0.593

Figure 3. Cesium-137 isotherms for sand from the ANL-E and ORNL lysimeters.



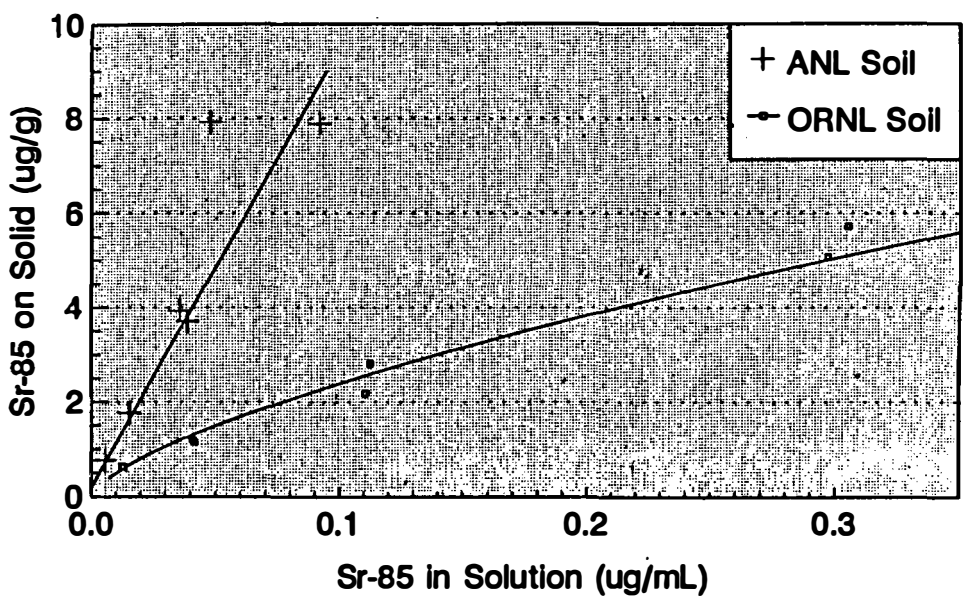
ANL Silica: Power Regression, a =2.79 b = 0.74
ORNL Silica: Power Regression, a = 2.86 b = 0.76

Figure 4. Cesium-137 isotherms for silica flour from the ANL-E and ORNL lysimeters.

strontium and cesium. Using the lowest five points from each curve, we have recalculated K_d based on a linear regression (this is reasonable at low cesium concentrations). From these data, the K_d for cesium on the ORNL soil is 37 mL/g, and for the ANL-E sample, the K_d is 390 mL/g. These values are also given in Table 3. At concentrations higher than about 500 µg/g on the solid, the non-linear function should be used.

Strontium is sorbed less than cesium in all cases. Results are shown in Figures 5 through 7, with results calculated in terms of µg of strontium. Sand and silica samples have linear isotherms, with no difference between the ANL-E and ORNL geomaterial samples (the difference in the experiments is the water that was used). When calculating the K_d of strontium for the ORNL lysimeter soil, it is necessary to account for the non-radioactive, natural strontium present in the soil, as well as the Sr-90 from the waste.

We determined the concentrations of stable strontium and cesium in deionized water that had been left in contact for 4 days with soil from the ANL-E and ORNL lysimeters. Four grams of dry soil were mixed with 40 grams of deionized water. The strontium content of water exposed to the ANL-E and ORNL soils was 0.014 µg/mL and 0.009 µg/mL, respectively, as analyzed by inductively coupled plasma, atomic emission spectroscopy. No stable cesium was detected in these samples at a detection limit of 0.2 µg/mL by atomic adsorption spectroscopy. The implication of this analysis and the assumption that the leachate from the waste is very low in elemental concentrations of cesium and strontium (even though the radioactivity may be high) is that the K_d values for the lysimeters are on the low end of the isotherms. This being the case, it is probably justifiable to use simple extrapolations of linear regressions to approximate K_d in modeling transport in the lysimeters.



ANL Soil: Linear Regression, a =0.758 b = 81.4
ORNL Soil: Power Regression, a = 2.44 b = 0.67

Figure 5. Strontium-85 isotherms for soil from the ANL-E and ORNL lysimeters.

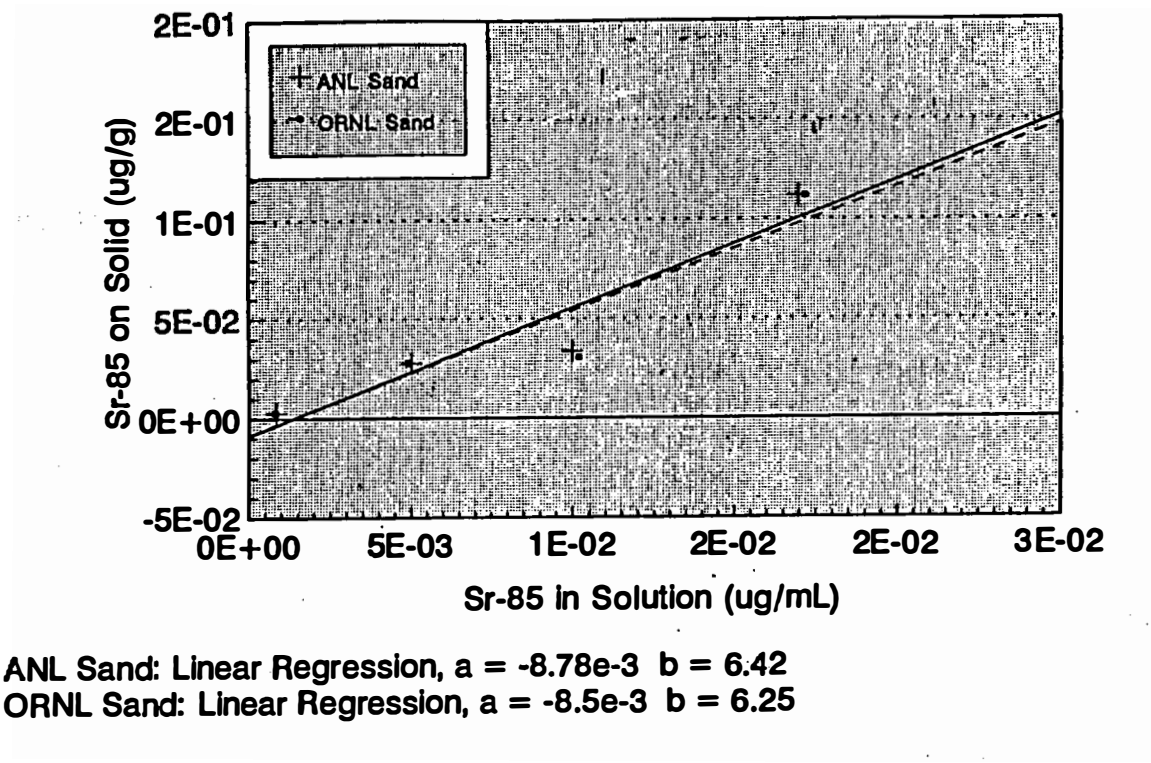


Figure 6. Strontium-85 isotherms for sand from the ANL-E and ORNL lysimeters.

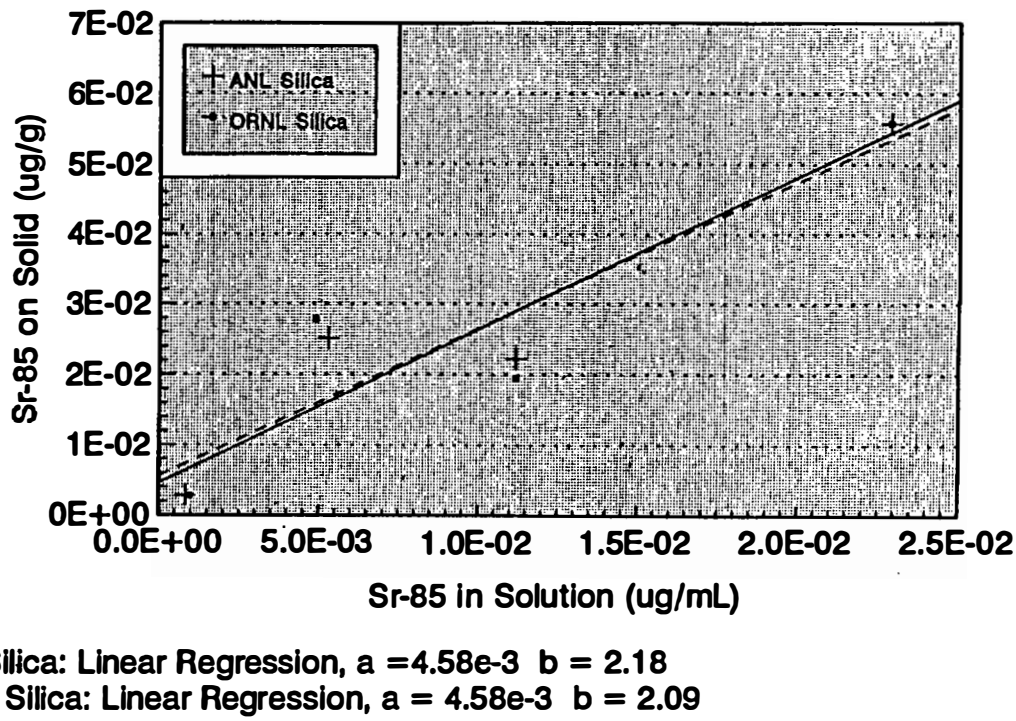


Figure 7. Strontium-85 isotherms for silica flour from the ANL-E and ORNL lysimeters.

Colloid Study

Introduction. During a preliminary investigation of leachate from the NRC Field Lysimeter Low-Level Waste Data Base Development Program,[23] qualitative evidence indicated the presence of beta-emitting radiocolloids. The present work was intended to first verify the initial results by similar experimentation and then identify the radionuclides associated with the colloids and their chemical composition. Based on additional information about particle morphology and particle composition, the source of the particles was to be determined. There were three hypotheses made prior to this work: (1) particles observed during preliminary work were generated as a result of degradation of the waste forms present within the lysimeters, (2) particles observed during preliminary work are chemical precipitates that occur on the filters used for fractionation, and (3) the primary radionuclide associated with the observed particles is Sr-90. These hypotheses were evaluated by investigating two sets of leachate from different soil-filled lysimeters. One contained a cement waste form and the second contained a VES waste form. A large volume of each leachate, approximately 2.0 L, was passed through each filter in a set of six filters ranging in size from 0.01 to 0.6 μm. Detailed radioanalyses including gamma spectroscopy, liquid scintillation spectrometry, and gross particle proportional counting were performed on the filters and the filtrate that passed through each filter. Energy dispersive x-ray analysis (EDX), neutron activation analysis (NAA), x-ray fluorescence analysis (XRF), protron induced x-ray emission analysis, gravimetric analysis, and scanning electron microscopy (SEM) were also performed on filters. Filtration data verified preliminary results, which demonstrated a relationship between the quantity and size of captured particles and the quantity of radioactive material on filters.

Additionally, increases in filtrate activity were observed as filter pore sizes decreased. The combination of gamma spectroscopy, liquid scintillation spectrometry, and proportional counting data supports a conclusion that the primary radionuclide associated with colloids observed in lysimeter leachate is Sr-90. About 2% of the Sr-90 in the lysimeter leachate studied appears to be associated with particulate material. At least four different types of colloids, ranging from about 0.02 μm to 20 μm in size, can be observed in leachate samples. Based on morphology and elemental analysis, one type appears to be a chemical precipitate of $Ca(OH)_2$. This type was in abundance in the leachate from the lysimeter containing a cement-solidified waste form. Another type is an organic with a spherical appearance. After being irradiated with a high-intensity electron beam for the purpose of EDX, these were observed to deform into the shape of a torus. This type was found only in the leachate from the lysimeter with a solidified VES waste form. The third and fourth types of colloids were common to leachate from both waste forms. The third type has an angular morphology and elemental composition consistent with weathered rock. The fourth type has a rounded morphology and appears similar to fine sand grains.

The chemical speciation of leachate demonstrates variability associated with the location of sample collection within the lysimeter and over time. Table 4 provides typical values of speciation for each lysimeter. The samples in which the Table 4 values were measured were obtained in April 1994 and were reported in Reference 21.

Methods and Materials. Commonly, water samples are acidified to prevent radionuclide deposition onto the walls of storage and shipping containers. This may destroy or deteriorate colloidal particles, which initially exist in alkaline solutions. The lysimeter leachate samples used in this investigation were not acidified.

Table 4. Chemical speciation of typical leachate samples.

Leachate Chemical Speciation Information		Cations				
Sample	Solidification Agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)
ANL-E 3-1	VES	119	4.8	15	0.9	45
ANL-E 3-3	VES	86	6.1	14	0.3	47
ANL-E 3-5	VES	93	2.6	15	0.2	45
ORNL 1-1	Cement	37	3.0	18	<2	1.7
ORNL 1-3	Cement	33	2.2	23	2.1	1.4

Leachate Chemical Speciation Information		Anions			
Sample	Solidification Agent	Cl (mg/L)	Nitrate (mg/L)	Phosphate (mg/L)	Sulfate (mg/L)
ANL-E 3-1	VES	1.6	1.1	<0.5	21
ANL-E 3-3	VES	6.6	<0.1	<0.5	22
ANL-E 3-5	VES	1.5	0.4	<0.5	24
ORNL 1-1	Cement	2.9	58	<0.5	18
ORNL 1-3	Cement	1.3	4.7	<0.5	14

One of the underlying goals of this investigation was to determine the radioactivity distribution between solid suspended phases and liquid phases within samples of lysimeter leachate. Solid phases were removed from suspension, and colloidal particle isolation was achieved using vacuum filtration techniques. Two-liter aliquots of leachate were passed through 47-mm-diameter polycarbonate track-etch membrane filters. The maximum pressure differential measured inside the vacuum flask during filtration was 9.3E+4 Pa; however, most samples passed through the pores of the membrane filter at much lower vacuum levels. Six polycarbonate filters, with pore diameters of 0.6 μm, 0.4 μm, 0.2 μm, 0.1 μm, 0.05 μm, and 0.01 μm, were used in the analysis of each sample. Approximately 2 L of fresh leachate was passed through each filter size. This fractionation technique has been employed previously with some success.[23] To evaluate the distribution of radioactivity between the solid and liquid phases, an array of different radioanalyses was performed. This included gamma spectroscopy, liquid scintillation counting/spectroscopy, and proportional counting. The following is a description of the samples obtained and how analyses were performed on them.

One-liter Marinelli beakers of leachate from both ORNL lysimeter 1 and ANL-E lysimeter 3 were acidified and then analyzed by high-resolution gamma spectroscopy using HP(Ge) detectors. Ten-milliliter aliquots of raw leachate and filtrate from each filtration were analyzed using liquid scintillation spectroscopy. Two-hundred-and-fifty-milliliter aliquots of filtrate were evaporated and analyzed for gross alpha and gross beta activity. After gravimetric analysis, each polycarbonate filter was analyzed using gamma spectroscopy and using proportional counting to obtain gross alpha and gross beta activity. After these analyses, each polycarbonate filter was sectioned into fifths. One-half of each filter was analyzed using liquid scintillation spectroscopy. The internal standard method was used to determine the counting efficiency during each liquid scintillation analysis. One-quarter of each filter was prepared for NAA. One-eighth of each filter was prepared for SEM analysis. SEM analysis included sputter coating the samples with a layer of Au-W. One-eighth of each sample was prepared for gross XRF analysis. To evaluate the occurrence of radioactive material deposition within the leachate storage/shipping containers, a small volume (~1.0 L) of acidified (HNO_3) water (pH ~2.0) was added to each nearly empty container. The contents were vigorously shaken to ensure contact with the container walls. This fluid was analyzed by gamma spectroscopy. Table 5 summarizes the samples obtained and the sample analyses performed.

Results and Discussion. Table 6 provides the results of proportional counter and liquid scintillation counter analyses. Table 6 data demonstrate a correlation between filter pore size and filter radioactivity per unit volume. Generally, as pore size decreases, filter radioactivity increases. One observes slight differences among the data sets when comparing liquid scintillation counting data and proportional counter data. This is expected for several reasons. First, the liquid scintillation data are based on efficiency values derived from Sr-90/Y-90 standards. The gross beta proportional counter values are based on an assumed Cs-137 efficiency. This difference could easily account for such discrepancies. Secondly, only half a filter was analyzed during liquid scintillation analysis, whereas the whole filter was analyzed during proportional counter analysis. Differences between these two analysis techniques could reflect slight inhomogeneity of activity distribution over the sample surface.

Table 6 data also demonstrate increasing activity of ORNL filtrate as pore size is increased. The data illustrate that the presence of particles in the ORNL leachate is associated with some fraction of the total activity found in the leachate. However, they do not show the same clear relationship between ANL-E filtrate radioactivity and pore size.

No gross alpha emissions were detected in evaporated 0.250-L samples of raw leachate. Consequently, no further alpha analyses were performed. Neither Co-60 nor Cs-137 energy lines were observed during gamma spectroscopic analyses of raw leachate, filters, and filtrate.

Table 5. Sample and analyses array.

		Analyses Types									
Sample Type	Filter Pore Size	Gamma Spectroscopy	Gross Alpha	Gross Beta	Gravimetric	LSC	SEM	EDX	XRF	PIXE	NAA
Leachate ANL-E 3		X	X	X		X					
Leachate ORNL 1		X	X	X		X					
Filters ANL-E 3	0.01 µm	X		X	X	X	X	X	X	X	X
	0.05 µm	X		X	X	X	X	X	X	X	X
	0.10 µm	X		X	X	X	X	X	X	X	X
	0.20 µm	X		X	X	X	X	X	X	X	X
	0.40 µm	X		X	X	X	X	X	X	X	X
	0.60 µm	X		X	X	X	X	X	X	X	X
Filters ORNL 1	0.01 µm	X		X	X	X	X	X	X	X	X
	0.05 µm	X		X	X	X	X	X	X	X	X
	0.10 µm	X		X	X	X	X	X	X	X	X
	0.20 µm	X		X	X	X	X	X	X	X	X
	0.40 µm	X		X	X	X	X	X	X	X	X
	0.60 µm	X		X	X	X	X	X	X	X	X
Filtrate ANL-E 3	0.01 µm	X				X					
	0.05 µm	X				X					
	0.10 µm	X				X					
	0.20 µm	X				X					
	0.40 µm	X				X					
	0.60 µm	X				X					
Filtrate ORNL 1	0.01 µm	X				X					
	0.05 µm	X				X					
	0.10 µm	X				X					
	0.20 µm	X				X					
	0.40 µm	X				X					
	0.60 µm	X				X					

Table 5. (continued).

Sample Type	Filter Pore Size	Analyses Types									
		Gamma Spectroscopy	Gross Alpha	Gross Beta	Gravimetric	LSC	SEM	EDX	XRF	PIXE	NAA
Acidified Container Remains											
ORNL 1		X									
ANL-E 3		X									

Gamma spectroscopy = high resolution spectroscopy performed with an HP(Ge) detector.

Gross alpha and gross beta = proportional counting with analyses for gross alpha and gross beta activity.

LSC = liquid scintillation counting/spectroscopy for the analysis of beta activity.

SEM = scanning electron microscopy.

EDX = energy dispersive x-ray analysis.

XRF = to x-ray fluorescence analysis.

PIXE = proton-induced x-ray emission analysis.

NAA = neutron activation analysis.

Table 6. Results of proportional counter and liquid scintillation counter data.

Filter Measurements

Filter Pore Size (µm)	ORNL Filters LSC (Bq/L)	Uncertainty (Bq/L)	Gross Beta (Bq/L)	Uncertainty (Bq/L)
0.01	2.97	0.27	3.49	0.22
0.05	0.17	0.02	0.39	0.03
0.10	0.05	0.00	0.03	0.01
0.20	0.08	0.00	0.36	0.03
0.40	0.05	0.00	0.06	0.01
0.60	0.05	0.00	0.12	0.02

Filter Pore Size (µm)	ANL-E Filters LSC (Bq/L)	Uncertainty (Bq/L)	Gross Beta (Bq/L)	Uncertainty (Bq/L)
0.01	0.27	0.02	0.21	0.02
0.05	0.11	0.00	0.06	0.01
0.10	0.07	0.00	0.06	0.01
0.20	0.08	0.00	0.09	0.01
0.40	0.08	0.00	0.06	0.01
0.60	0.06	0.00	0.06	0.01

Filtrate Measurements

Filter Pore Size (µm)	ORNL Filtrate LSC (Bq/L)	Uncertainty (Bq/L)	Gross Beta (Bq/L)	Uncertainty (Bq/L)
0.01	148.38	1.17	133.48	0.44
0.05	191.95	1.26	167.78	0.49
0.10	196.74	1.26	173.01	0.50
0.20	197.07	1.27	162.90	0.49
0.40	193.81	1.27	172.56	0.50
0.60	192.89	1.27	179.71	0.51

Filter Pore Size (µm)	ANL-E Filtrate LSC (Bq/L)	Uncertainty (Bq/L)	Gross Beta (Bq/L)	Uncertainty (Bq/L)
0.01	12.98	0.82	10.83	0.13
0.05	12.3	0.82	11.50	0.13
0.10	15.72	0.83	9.56	0.12
0.20	11.62	0.82	11.49	0.13
0.40	12.96	0.82	10.61	0.13
0.60	10.06	0.80	10.43	0.13

Gross beta = proportional counting with analyses for gross alpha and gross beta activity.
LSC = liquid scintillation counting/spectroscopy for the analysis of beta activity.

These data support preliminary results suggesting a colloidal transport mechanism. It appears that the ORNL leachate contains a tightly bound radiocolloid with a diameter slightly smaller than 0.2 μm. This observation arises from a review of Table 6 filter data, which show a decrease in activity up to a filter pore diameter of 0.2 μm. Table 6 filtrate data show an increase in activity up to a 0.2-μm diameter pore size.

The shapes of the summed ORNL filter liquid scintillation spectrums and the summed ANL-E filter liquid scintillation spectrums were compared to the shape of the summed spectrums from a sample spiked with Sr-90/Y-90. Although the spiked sample had slightly different quenching characteristics than the filter samples, there was an apparent relationship between the shapes of these spectrums. In particular, a broad spectrum consistent with the radionuclides Sr-90 and Y-90 was observed. There was an absence of Cs-137 and Co-60 in the leachate during gamma spectroscopy. No gross alpha activity was observed in either leachate sample. These leachate samples historically contain higher concentrations of Sr-90 than other radionuclides. These observations support the hypothesis that the primary radionuclide in the leachate samples, which is associated with colloidal particles, is Sr-90.

NAA and XRF results were of limited use. The difficultly with these analyses was that they suffered from sample mass that was too small. Trace quantities of Co-60 and Mn-56 arising from their elemental precursors, stable Co-59 and Mn-55, were observed on neutron-activated filter samples. The relationship between these results and the overall transport of radioactive material in the porous lysimeter matrix is not clear. Gross XRF analysis was likewise limited, showing only elemental manganese.

SEM and EDX analyses demonstrated that at least four different types of colloids ranging from about 0.02 to 20 μm in diameter were present. Figure 8 is an example of the morphology of one predominant type of particle found in filters used with ORNL leachate. These are hexahedron crystalline particles ranging in size from about 0.2 to 10.0 μm. Using EDX, the elements detected in these particles were calcium and oxygen.

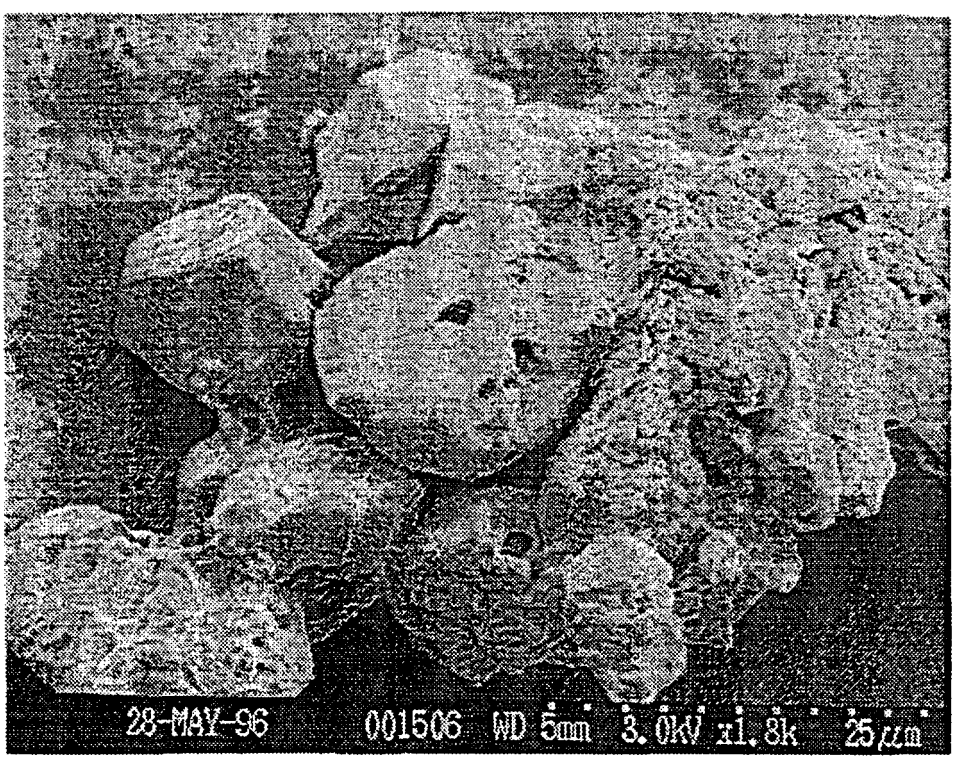


Figure 8. ORNL leachate particles: an example of particles composed primarily of calcium and oxygen.

Figures 9 and 10 show particles common to leachate from both lysimeter systems investigated. The particles in both figures contain trace amounts of phosphorous, calcium, silicon, oxygen, and manganese as detected using EDX. Figure 9 shows angular features but does not appear to be crystalline. This type of particle was observed to be from around 5 to 20 μm in diameter. Figure 10 demonstrates the last common type of particle observed. This type of particle has a smooth or rounded surface. It ranges in size from 0.05 to about 10.0 μm.

This investigation supports previous findings of a radiocolloid transport mechanism through the lysimeter soils of the NRC Field Lysimeter Low-Level Waste Data Base Development Program. These findings are consistent with concerns regarding this transport mechanism as developed or discussed within several contemporary works on this subject.[24–27]

The hypothesis that Sr-90 is the primary radionuclide associated with collodial particles found in the EPICOR leachate examined is well supported. No gamma or alpha radiation emissions could be detected on filters or filtrate. Filter sample beta spectra were consistent with Sr-90/Y-90 source spectra. About 2% of the Sr-90 activity in the leachate seems to be associated with colloidal material.

There is morphological and stoichiometric evidence to indicate that one type of particle frequently observed in the ORNL lysimeter, which contains a solidified cement waste form, is composed of $Ca(OH)_2$. It is speculated that this may arise from the cement-solidified waste form since there is an excess of $Ca(OH)_2$ in cement matrices.[28] It seems plausible that $Ca(OH)_2$ is the colloidal particle most closely bound with strontium due to the similarity between calcium and strontium. However, this investigation cannot empirically substantiate this theory. There is a unique type of spherical organic particle associated with the leachate from the ANL-E lysimeter, which contains a VES-solidified waste form. Based on particle behavior while being examined by EDX, it is likely to be a polymer. It is possible that these particles may be a degradation product of the VES waste form. But as in the case of $Ca(OH)_2$ particles, there is no definitive evidence to verify these speculations. These items might be further investigated by examination of the waste form when it is removed from the lysimeter. The hypothesis that particles observed are a result of waste form degradation has some speculative support based on these investigations and cannot be rejected. However, there is no clear support of this hypothesis.

The third hypothesis, stating that the colloids previously identified are chemical precipitates occurring on the filter, can be rejected based on morphology. Neither the angular nor smooth, rounded-shape particles have a morphology consistent with precipitation onto the filter. The spherical organic particles and $Ca(OH)_2$ particles, because of their shapes, were clearly formed while in suspension. There is some precipitation of solid materials onto the filters used for fractionation purposes. Based on gravimetric analysis showing no or only weak correlation between particle deposition mass and filter activity, normalized by volume filtered, this appears to be incidental and of little significance.

Upward Migration of Radionuclides at ORNL

During previous samplings, the presence of both Cs-137 and Sr-90 were discovered at the surface of lysimeter ORNL-5, which is the sand-filled control. Radionuclide activity was first detected during a routine gamma survey of the lysimeter's surface in 1991. At that time, more activity was found near the center than at the edges. Surface samples were obtained from the center of the lysimeter at depths from 0 to 2.5 cm and from 2.5 to 5 cm for analysis of cesium and Sr-90. Analysis detected Cs-137, Cs-134, and Sr-90. These data showed that more nuclides were at the surface, suggesting some type of an active deposition mechanism. There remained a question, however, concerning the source of the nuclides. In August of 1992, samples were again taken from the lysimeter and analyzed for Cs-137 and Cs-134. The results were similar to the previous sampling.

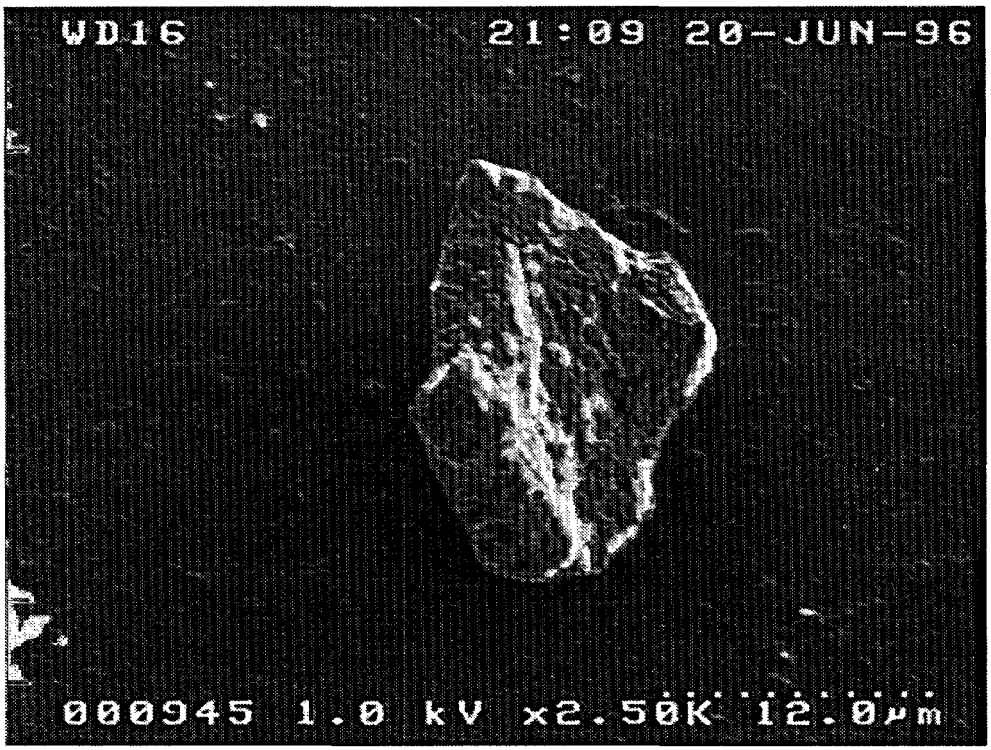


Figure 9. Angular particle common to both ANL-E and ORNL leachate samples.

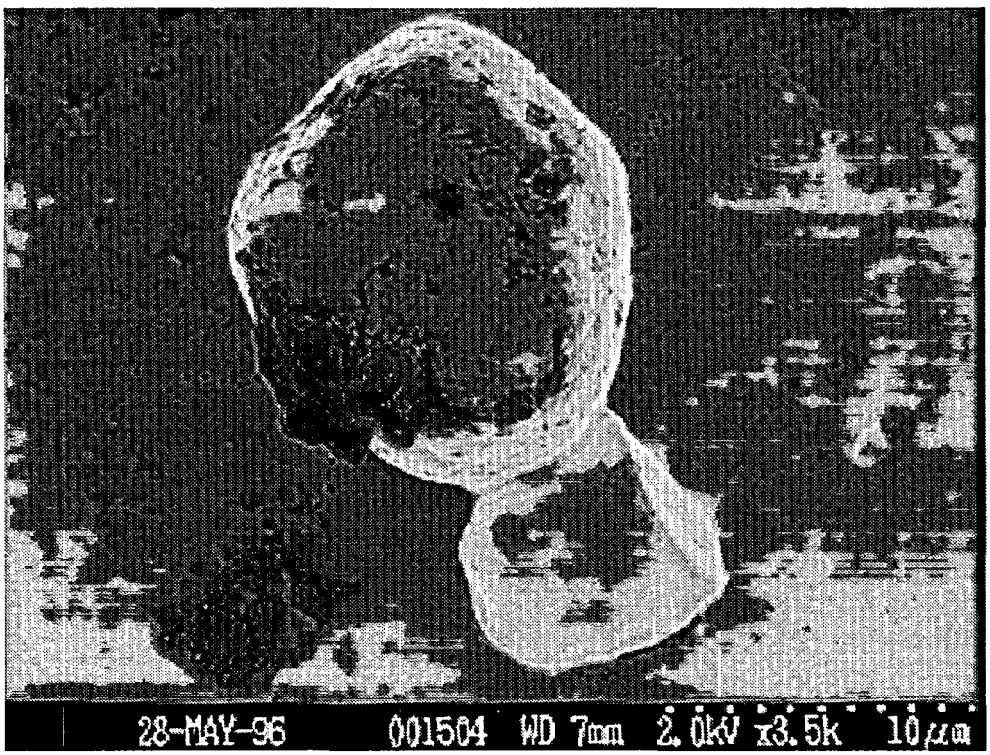


Figure 10. Smooth particle common to both ANL-E and ORNL leachate samples.

On January 31, 1994, two cores of sand 80 cm long were collected from lysimeter number 5 to be analyzed for cesium and strontium. One core was collected from the side of the lysimeter 13 cm from the wall, and the other was collected from the center of the lysimeter directly above the buried waste form (located approximately 100 cm below the sand surface). These sand cores were sectioned into 5-cm segments. Radiocesium and strontium activity were measured for each segment.

The analyses show that Cs-137 and Cs-134 were present throughout the length of the center core (Table 7). Cesium-137 was also found in all segments of the side core, but Cs-134 was only found in the upper half of that core. Cesium-134 is an activation product that is formed in the core of nuclear reactors and does not occur naturally; therefore, the ratio of Cs-137 to Cs-134 in the sand segments can be used to determine if there was an outside source of cesium added to the lysimeter. By decay-correcting the original ratio of the waste form to the date of sand collection (and assuming that both radioisotopes behave chemically identical), the ratio should be equal to 399. The data presented in Table 7 and Figure 11 indicate that the ratio in all segments of the center core fall close to this value, except for segment 2 (66.5 to 61.5 cm in depth). This confirms that the cesium seen throughout the length of the sand core is a result of upward vertical transport from the waste form sample.

During the sectioning of the core, it was noticed that there was a fine plant root present throughout the depth of the core. The root material was extracted from each segment and counted. The results are presented in Table 7 and Figure 11. Cesium-137 activity is associated with the roots, and peaks in the root data occur at the same depths as do peaks in the sand activity. There are three peaks seen in cesium content (Figure 11): one at 24 cm, a large peak at 39 cm, and a smaller peak at 64 cm. These peaks may be indicative of some sort of periodic movement of the cesium, but completion of the soil core studies is necessary before this can be verified. It can be seen that there are higher concentrations of Cs-137 associated with the roots than with the sand. Sand from the deepest three segments was analyzed three separate times. The first time was the whole segment, and the other two times were subsamples of the sand. Segment 2 (Table 7) has a fairly wide range of activities between the whole segment and the two subsamples, suggesting that the activity in the sand is not evenly distributed. This could be a result of the root being involved in the transport process.

Strontium-90 analysis results show that there is significant strontium throughout the entire depth of the core (Table 7 and Figure 11). Peaks occur in the distribution at the same depths as for cesium in both the sand and roots. This suggests that the same mechanism may be involved for transporting strontium upward as for cesium. Strontium and cesium behave very differently chemically, but if the process of migration is more physical than chemical, then the ratio of Cs-137 to Sr-90 should be similar at all depths. Table 7 includes a tabulation of this ratio versus depth. It can be seen that the ratios are similar for most of the segments, indicating that the upward transport is possibly related to a physical phenomenon such as evaporation enhanced by transpiration through the root. The fact that the sand has a very low cation-exchange capacity is probably the reason that the physical aspect of migration is so evident.

A comparison of the Cs-137 concentrations in the center and side cores (Figure 12) shows that Cs-137 concentrations are much lower at the edge of the lysimeter as might be expected. Those areas are dependant on dispersion as well as evaporation to move the nuclides to them. The obvious peaks of concentration in the center core are less apparent and appear to be delayed about 5 cm.

The radionuclide data confirm that the cesium and strontium present in the sand core were transported by an undetermined mechanism upward from the buried waste form. The partition coefficients measured for cesium and strontium in the clean sand indicate that strontium should be nearly six times more mobile than cesium. However, the strontium and cesium profiles in the sand core are quite similar (Figure 11), with subsurface activity peaks occurring at the same depths. The

Table 7. Cesium (Cs) and strontium (Sr) analyses for sand core segments from the center (Core C) and side (Core S) and root fragments from the center of ORNL lysimeter 5 collected on January 31, 1994.

Segment Number		Depth (cm)	Sand										Plant Root	
			Segment Dry Weight (g)		Cs-137 (pCi/g)		Cs-134 (pCi/g)		Sr-90 (pCi/g)	Ratio[b] Cs-137/Cs-134		Ratio[e] Cs-137/Sr-90	Sample Weight (g)	Cs-137 (pCi/g)
Core C	Core S		Core C	Core S	Core C	Core S	Core C	Core S	Core C	Core C	Core S	Core C		
—[a]	1	76.5-71.5	—	126.11	—	0.26	—	ND[c]	—	—	d		—	—
1	2	71.5-66.5	100.68	139.80	598.1	0.20	1.5	ND	1.5	399		450	0.0134	18,900
1	—	71.5-66.5	17.08	—	704.5	—	1.6	—	—	448			—	—
1	—	71.5-66.5	17.16	—	660.8	—	1.6	—	—	425			—	—
2	3	66.5-61.5	118.92	120.32	1,303.4	0.22	1.8	ND	3.5	724		650	0.0172	20,660
2	—	66.5-61.5	17.48	—	2,241	—	2.3	—	—	969			—	—
2	—	66.5-61.5	20.37	—	1,550	—	1.8	—	—	882			—	—
3	4	61.5-56.5	121.53	131.40	356.7	0.19	0.7	ND	2.0	484		200	0.0301	20,480
3	—	61.5-56.5	19	—	400.7	—	0.8	—	—	498			—	—
3	—	61.5-56.5	14.85	—	376	—	0.7	—	—	517			—	—
4	5	56.5-51.5	115.25	109.76	490.2	0.24	1.1	ND	2.1	447		300	0.0234	22,540
5	6	51.5-46.5	117.07	115.29	403.3	0.17	0.8	ND	2.7	492		200	0.0216	27,520
6	7	46.5-41.5	125.28	141.24	1,594	0.19	3.2	ND	7.6	491		300	0.0224	27,360
7	8	41.5-36.5	129.06	113.21	37,283.1	0.40	30.4	ND	14.1	466		1,000	0.0220	81,970
8	9	36.5-31.5	121.14	124.99	551.2	1.14	1.4	ND	1.5	404		400	0.0302	13,620
9	10	31.5-26.5	116.32	117.30	866.6	38.9	2.3	0.08	3.5	376	467	300	0.0196	10,150
10	11	26.5-21.5	122.86	135.38	5,484.2	6.1	10.9	ND	7.6	475		750	0.0463	21,580
11	12	21.5-16.5	117.94	108.01	2,032.4	2.6	4.4	ND	16.0	458		200	0.0256	5,990
12	13	16.5-11.5	125.78	104.74	1,513	3.5	3.6	ND	0.5	423		3,400	0.1049	3,850
13	14	11.5-6.5	94.99	117.22	711.7	9.0	1.8	0.02	0.2	390	557	4,400	0.0615	5,940
14	15	6.5-0	150.30	142.25	715.2	53.6	1.6	0.12	0.6	451	462	1,200	0.3105	8,570

a. No measurement was taken at this location.
b. Theoretical ratio of Cs-137/Cs-134 = 399.
c. ND = none detected.
d. Blank indicates not enough information available to calculate ratio.
e. Theoretical ratio of Cs-137/Sr-90 = 440.

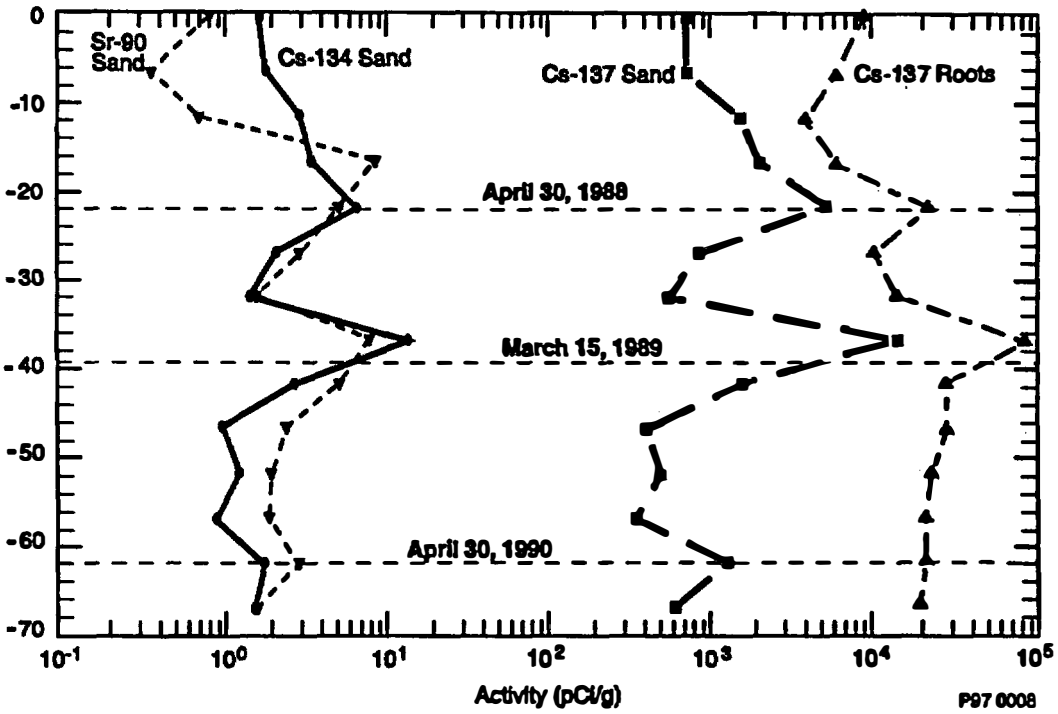


Figure 11. Cesium-137 and Sr-90 associated with plant roots and sand from the center core taken from ORNL lysimeter 5.

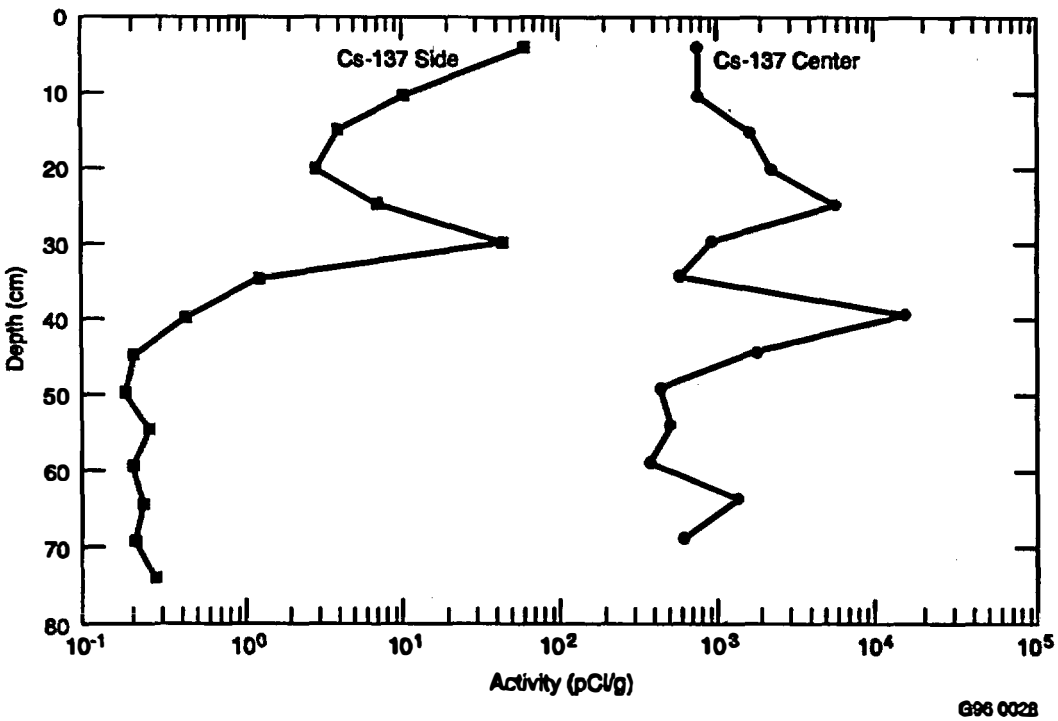


Figure 12. Comparison of Cs-137 analysis from center versus side sand cores from ORNL lysimeter 5.

chemical behavior of cesium is such that it would tend to be sorbed much like potassium to clays, organic matter, or other similar materials,[29,30,31] yet initially, the sand had a very low cation-exchange capacity (0.06 cmol/kg), indicating that there was little, if any, clay present in the sand to sorb the cesium.

The discrete peaks in radionuclide activity in the sand and associated with the roots suggest that there were periods of increased releases of the radionuclides from the waste forms or episodic upward transport, or both. During the 8.5 years of operation of this experiment prior to coring, there had been four times when there was sufficient rainfall between leachate collections where leachate built up in the collector and sand column to a level at or above the waste form. The dates that the lysimeters were drained for the flooding events are 30 April 1988, 15 March 1989, 30 April 1990, and 12 April 1991. The inundation of the waste forms may have allowed more radionuclides to become available for transport than under normal conditions. This becomes evident by examining the cumulative activity curves for both Sr-90 and Cs-137 in the leachate collected from the reservoirs shown in Figures 13 and 14. For the time period shown, nearly six times as much Sr-90 was collected in the reservoirs as was Cs-137. The cumulative curve for Sr-90 is smoother and more regular than for Cs-137. The times for flooding are shown on the figure, and it can be readily seen that there were increased releases of both Sr-90 and Cs-137 to the leachate following these events, with the increase in cesium much more noticeable, confirming that more cesium was made available to transport due to the inundation. In fact, for a flooding event that occurred after the core was collected (24 April 1994), the cumulative release of Cs-137 to the reservoir increased by more than 14-fold over the next two sampling periods.[32]

The total amount of Cs-134, Cs-137, and Sr-90 present in the center core was determined and compared to the initial inventory. It was found that for all three radionuclides, the fractional release present in the core was 3E-6. The measured effective diffusion coefficients for the radionuclides out of the waste forms suggests that there should have been a greater fractional release

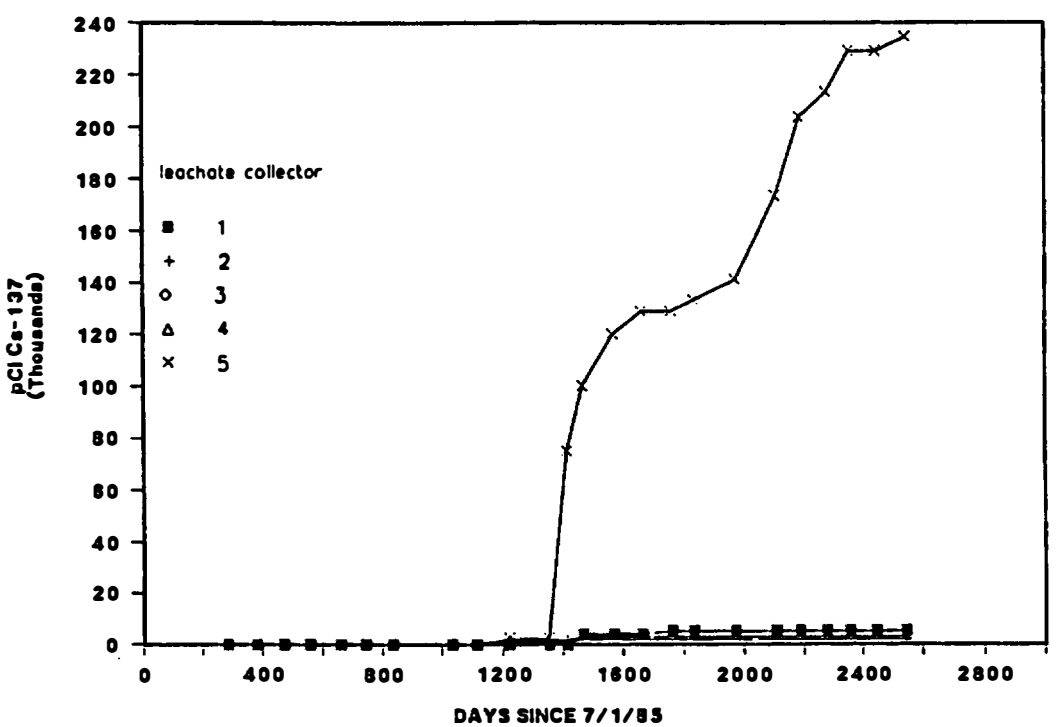


Figure 13. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

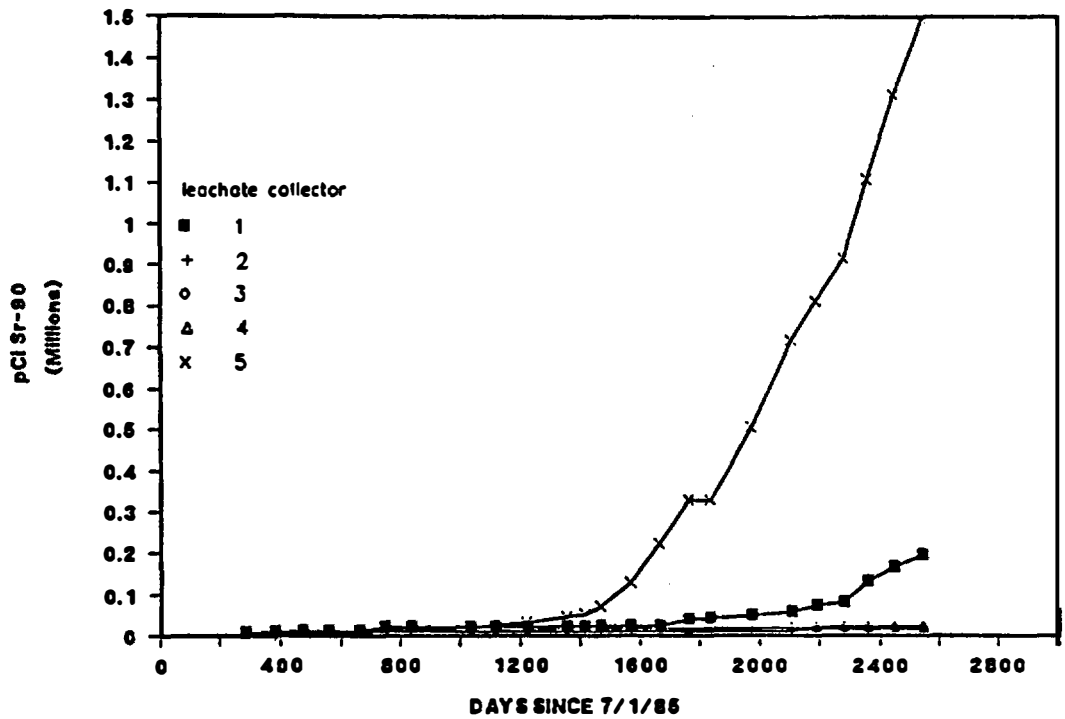


Figure 14. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

of strontium than cesium, which was not the case. The constant fractional release further supports the theory that the flooding events were instrumental in making additional radionuclides available for upward transport.

The increased cesium concentration associated with the root as compared to the sand segments and the concentration peaks occurring in both at the same depths strongly suggests a positive relationship between the transport of the radionuclides and the presence of the root. The discoloration around the root indicates that there was a modification of the soil water chemistry in the vicinity of the root, with the possible addition of organic matter to which the radionuclides could sorb. It is likely that the root would take up cesium in a manner similar to the uptake of nutrients by roots because of its similar behavior as potassium. This is also true for strontium, which is taken up by plants similarly to calcium.[33]

The upward transport of cesium and strontium is a phenomenon not commonly observed. The similar nature of the distribution of radionuclides associated with the root and within the sand core and the relatively small radius of increased radionuclide activity strongly suggests a corresponding relationship between the root and upward migration. It appears most likely that the transport of the radionuclides is within the evapotranspiration stream of the unidentified plant. Whether the migration was through the root or just outside cannot be determined from the data collected.

The upward transport was discovered because the sand that was used in the lysimeter had little or no charged surface areas for the strontium or cesium to sorb to. In most natural soils, clays and organic matter would adsorb the radionuclides before they could migrate significant distances, especially in the relatively short time observed here. In addition, the persistence of the high concentration of radionuclides in the vicinity of the root with time is quite remarkable given that nearly 140 cm of precipitation (over 900 L) on average flushes through the lysimeter every year.

The upward transport does have important implications in regard to subsurface disposal of wastes contaminated with cesium and strontium. Conventional wisdom suggests that cesium and

strontium will be relatively immobile because they are easily sorbed to soil particles, and any movement would be downward in areas where rainfall infiltration significantly exceeds evaporation. However, upward migration in low-level radioactive waste disposal facilities, under specific conditions, could lead to direct exposures and offsite transport by erosion.

Use of Lysimeter Data for Performance Assessment Radionuclide Release Calculations

This study[b] is a continuation of previous work,[34] which examined the release of cesium and strontium from these lysimeters over 8 years. In the previous study, the influence of key transport parameters (dispersion coefficient and distribution coefficient), waste form release rates, and the choice of the boundary condition on estimated release were investigated. A major finding of the previous study was the need to measure the equilibrium soil partition coefficient, K_d. This was accomplished and the data are used for the basis of the new analysis of the data.

Model. Measurable releases were collected from moisture cup 3, which is located approximately 23 cm from the bottom of the waste forms, and the lysimeter leachate collector system, located 51 cm below the bottom of the waste forms. The radionuclides found in the leachate collector to date are Cs-137, Sr-90, Cs-134, and Sb-125.

After review of the data, we chose to model the release of Cs-137 and Sr-90 from portland type I-II cement located in lysimeter 5 at ORNL and ANL-E because releases from the lysimeters filled with soil were so low that meaningful comparisons between the computer code and the experimental data could not be performed. After 10 years, releases from the control lysimeters for cesium were less than 4.3E-4%, while releases of strontium were less than 0.2% of their respective initial inventories. Fractional releases from soil-filled lysimeters are at least an order of magnitude less than from the sand-filled lysimeters.

The DUST computer code was selected as the tool for predicting releases to the lysimeter leachate collector. The DUST code models waste form release and contaminant transport in a one-dimensional representation of a porous media. Transport is modeled by a finite difference method or by a mixing cell cascade approach. The mixing cell model is limited in that it does not take diffusional waste form release into consideration nor does it model dispersion during transport. Therefore, for our simulation, we selected the finite difference model. More detailed descriptions of the models in the code are given in References 35 and 36.

Geometry and Boundary Conditions—The base geometry for our model placed 121 nodes with equal spacing of 1 cm. The seven waste forms were broken into 53 separate forms of 1 cm in height. However, diffusion-controlled release from the waste form was modeled using the total dimensions of the combined waste forms, that is, a cylinder of 2.4 cm in radius and a height of 53 cm. Concentration and flux traces were obtained to match moisture cup 3 and the lysimeter leachate collector. The concentration and flux traces were taken at the points 23 and 51 cm below the waste forms. In this report, we use the total mass released 51 cm beneath the waste form as our performance measure. Outputs were stored every quarter year, and the actual data were interpolated so the model and actual times coincided with one another. The soil diffusion coefficient had a minor role on the release measured at the leachate collector, and it was set to 1E-7 cm^2/s at both ORNL and ANL-E. Zero concentration was specified at the top boundary for the ANL-E and ORNL trials. Zero dispersive flux was specified at the bottom boundary in both cases. This boundary condition simulates release through drainage.

Model Parameters—At ANL-E, lysimeter 5 contained PF-7 resin waste solidified in portland type I-II cement, while lysimeter 5 at ORNL contained PF-24 resin waste also solidified in portland type I-II cement. Experimental

b. T. M. Sullivan, "Predictive Modeling of Contaminant Release from the Nuclear Regulatory Commission Low-Level Waste Lysimeters," letter report, July 12, 1996.

measurements of release from these waste forms indicate that diffusion is the rate-controlling mechanism.[8] Measured diffusion coefficient values for the resins under study solidified in portland type I-II cement are 9.6E-10 cm^2/s for Sr-90 and 6.0E-10 cm^2/s for Cs-137.[c] These values were used as the base case values. The total flow through the lysimeter collection system in 10 years was 8,864 L at ORNL and 6,081 L at ANL-E. The 10-year average volumetric flow per unit area (i.e., darcy velocity) was 93.5 cm/yr (2.96E-6 cm/s) at ANL-E and 136.2 cm/yr (4.3E-6 cm/s) at ORNL.[22] In the simulation, yearly averages consistent with the data were used when calculating flow and transport. The soil bulk density values were 1.55 g/cm^3 at ANL-E and 1.60 g/cm^3 at ORNL.[12] Soil moisture content values are found in Reference 20. In lysimeter 5 at both sites, the average moisture content was calculated as 21%. The distribution coefficients were measured for Sr-90 or Cs-137 by Brookhaven National Laboratory and are reproduced in Table 8.[d] While the sorption isotherms were not always linear, for the low concentrations of the contaminants, the distribution coefficients are treated as linear. The dispersion coefficients have not been measured. As such, they were estimated by fitting the model predictions to the data. Generally speaking, the dispersion coefficient is 0.01 to 0.1 of the transport length. In this experiment, the transport length ranges from 0.5 (bottom of the waste form) to 1.1 m. Thus, expected dispersion coefficients should range from 0.5 to 11 cm. The released mass collected from the lysimeters was used to make comparisons to the DUST code predictions.

Table 8. Linear partition (distribution) coefficient (K_d) values for cesium and strontium in lysimeter materials.

Material	Cesium K_d (cm^3/g)	Strontium K_d (cm^3/g)
ANL-E soil	390	81
ORNL soil	37	20 - 40
ANL-E sand	55	6.4
ORNL sand	40	6.3

c. J. W. McConnell, Jr., Information presented at the NRC Workshop on Performance Assessment, Washington D.C., June 2-5, 1992.

d. M. Fuhrmann, "Partition Coefficients of Geomaterials used in the Nuclear Regulatory Commission Low-Level Waste Lysimeters," letter report, January 1996.

Initial amounts of Cs-137 and Sr-90 varied at ORNL and ANL-E due to the fact that they were solidified with two different resin types. In ORNL lysimeter 5, the PF-24 resin waste form had a total initial inventory of 3.29E-3 Ci of Sr-90 and 1.432 Ci of Cs-137. The PF-7 resin waste form at ANL-E had a total initial inventory of 1.84E-2 Ci of Sr-90.[12] Cesium-137 was not modeled at ANL-E for lack of sufficient releases.

Previous Results: Modeling 8 Years of Data—Three parameters are known to strongly influence release through the lysimeter soil column. They are K_d (the distribution coefficient) and the dispersivity coefficient, which together control transport from the waste form through the soil column, and the waste form diffusion coefficient, which controls waste form release rates. In a previous study,[34] several cases were modeled in which either K_d (the dispersivity coefficient) or the waste form diffusion coefficient was varied to best match the actual release data from the lysimeters.

Release to the lysimeter leachate collector is a function of waste form release. In the modeling studies, increasing the waste form release rate by a factor of two doubles the lysimeter release rate. Based on measured waste form diffusion coefficients, approximately 50% of the Sr-90 inventory would be released within 10 years. At the leachate collector, less than 0.2% of the inventory of strontium has been released after 10 years. This indicates that transport through the lysimeter sand is the controlling process, or that the laboratory-measured leach rates do not apply in the field.

Parametric studies on the influence of the value for the dispersivity and the distribution coefficient indicated that predicted release at the

leachate collector was extremely sensitive to both of these parameters. Increasing the distribution coefficient lowers solution concentrations and increases the time required to travel from the waste form to the leachate collector. Increasing the dispersivity value increases the amount released at early times (in this experiment, less than 20 years). Over the range of expected dispersivity values, the predicted release at the leachate collector varied by several orders of magnitude over the first 7 years.

Modeling of Lysimeter Release.

Measured Data—The criterion by which the model results are deemed adequate is based on the measured cumulative release and concentration in the leachate. Strontium-90 release from lysimeter 5 at ORNL and ANL-E is the base case for these modeling efforts. Other radionuclides and lysimeters will be discussed later in this report.

Lysimeter leachate concentrations were calculated from the actual data using the amount of mass collected per volume of leachate sampled over a given time period. The volume of leachate and released amounts are presented in References 18 through 22.

Figure 15 presents the cumulative release of Sr-90 from lysimeter 5 (the sand-filled lysimeters) at ORNL and ANL-E. In both cases, the releases are small for the first 5 years and show a trend of increasing release rates between 5 and 10 years. After 10 years, 6E+6 pCi and 4.2E+6 pCi of Sr-90 have been collected in the leachate collectors at ORNL and ANL-E, respectively.

Base Case—Using the measured waste form diffusion coefficients and K_d values in Table 8 for sand-filled lysimeters, the DUST code was used to predict releases from the lysimeters. A dispersion value of 0.6 cm was selected for the base case. This is towards the low end of expected values and was selected because it was assumed that the sand forms a relatively homogeneous pore structure for transport; therefore, dispersion should be minimized. However, the lysimeters also have instrumentation (moisture cups) and the waste forms that might act to increase dispersion. For the base case, sensitivity analysis on the

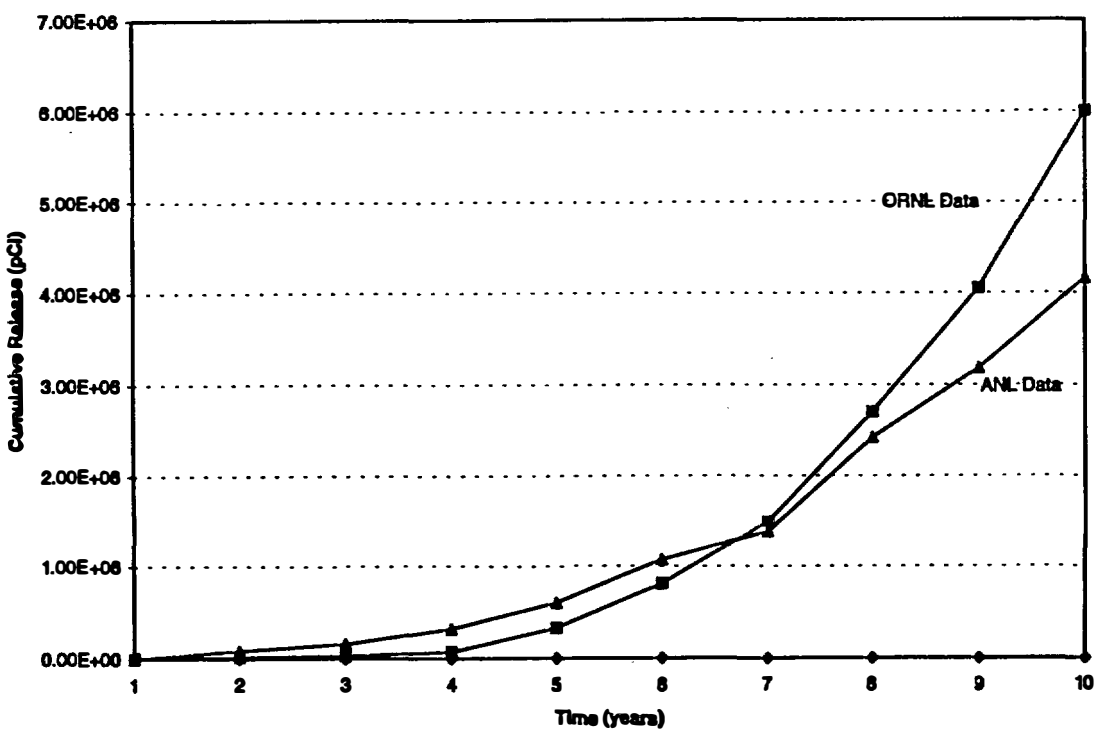


Figure 15. Measured cumulative release of Sr-90 from lysimeter 5 at ORNL and ANL-E.

effect of dispersivity was conducted, and the results are relatively insensitive to the dispersion value.

Figure 16 presents the predicted Sr-90 release from the sand-filled lysimeters at ORNL and ANL-E. The predictions clearly do not match the data. Modeled releases over the 10-year period approach 1E+9 pCi; this is almost three orders of magnitude greater than the measured data. Although not shown, the predicted concentrations in the leachate also exceed the measured amounts by orders of magnitude.

Analyzing the model results provides the cause for this discrepancy. Using the laboratory-measured waste form diffusion coefficient to predict release from the waste form over 10 years indicated that approximately one-half of the inventory will be released. Second, with the measured K_d values, the contaminant pore velocity is about 10 cm/yr. Therefore, the transport time from the bottom of the waste form to the leachate collector is on the order of 5 years. The transport time from the top of the waste form is about 10 years. Thus, most of the mass released over the 10 years would reach the leachate collector, provided the K_d parameter is correct. The DUST model predictions indicate that 10 to 20% of the mass released over the 10-year period should reach the leachate collector within the first 10 years of operation.

This leads to the conclusion that either the laboratory-measured waste form diffusion coefficients are not appropriate for leaching under field conditions, the laboratory-measured K_d values are not appropriate for field conditions, or both are inappropriate. The K_d values could be inappropriate due to differences in moisture content in the field (unsaturated) and in the laboratory (saturated). Several researchers have performed studies which show that the degree of absorption is a function of soil moisture. There is the possibility that different chemical species are forming in the field and have different sorption characteristics. In an attempt to identify possible scenarios that could cause this behavior, several numerical

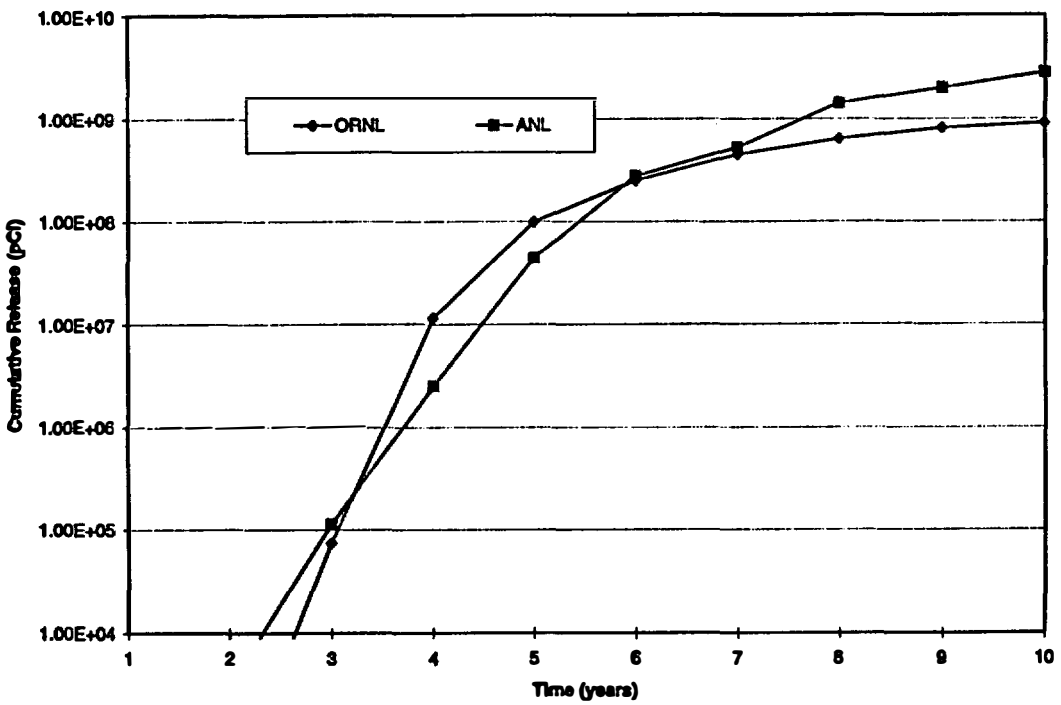


Figure 16. Predicted Sr-90 cumulative release from lysimeter 5 using measured K_d and waste form diffusion coefficient values.

investigations were performed. The investigations examined decreasing the waste form release rates to better fit the measured data while holding all other parameters constant, or increasing the K_d and dispersivity values while using the measured waste form diffusion coefficients.

Modeling to Match the Data for Sr-90 Release from ORNL and ANL-E Lysimeter 5—In this section, the cumulative release and lysimeter concentrations are examined for a number of different waste form release and transport scenarios. Three waste form release scenarios were modeled: diffusion-controlled, uniform, and solubility-controlled release. In these scenarios, all parameters were maintained at the base case value except the waste form release rate. Release rate parameters were chosen by trial and error to match the cumulative release after 10 years. In the diffusion-controlled model, the diffusion coefficient was reduced from the base case until a match with the release data was found. In the uniform release model, the model simulates a uniform fractional release each year corrected for radioactive decay. In the solubility-limited release model, release occurs up to the solubility limit. The solubility limit chosen will be less than actual solubility. The release depends on the volume of water flowing past the waste form. This solubility limit is an effective solubility for Sr-90 in the system and not a true solubility. The actual solubility would depend upon non-radioactive strontium as well. Further, since a one-dimensional model is used, the solubility used in the model is an average representation of the entire volume of the lysimeter. This is not necessarily the solubility at the waste form interface, which may be different due to changes in chemistry.

One alternative transport scenario was modeled. In this case, the waste form release rates were the same as found in the base case, and the transport parameters (K_d and dispersivity) were changed to obtain a fit to the data.

The "best" fit parameters for each model are displayed in Table 9. The three waste form release models all limit the total 10-year release from the waste form to about 1E+7 pCi, which is two orders of magnitude less than the release predicted in the base case (D_{wf} = 9.6E-10 cm^2/s). Attempts to optimize the fit to the data using statistical techniques (e.g., least squares) were not tried due to the speculative nature of the assumptions on release and transport parameters. The intent of these calculations was to show the types of behavior that would be required to match the data rather than to obtain the absolute "best" fit.

ORNL Lysimeter 5 Results—The "best" fit parameters for ORNL lysimeter 5 for each of the alternate models are found in Table 9. The "best" fit diffusion coefficient, 5E-14 cm^2/s, was four orders of magnitude below the laboratory-measured value. This low release rate is needed to match the data due to the relatively fast transport time that occurs when using the base case K_d values in Table 8. The "best" fit transport parameter, K_d=36, is much larger than the "best" fit value found previously based on 8 years of data (K_d = 24). After collecting the additional 2 years of data, it became clear that the previous "best" fit was too low because the

Table 9. "Best" fit parameters for cumulative release of Sr-90 from lysimeter 5 at ORNL and ANL-E.

Model	Parameter	ORNL	ANL-E
Diffusion-controlled release	Waste form diffusion coefficient (cm^2/s)	D_{wf} = 5E-14	D_{wf} = 1E-15
Uniform release	Uniform fractional release rate (yr^{-1})	4E-4	1.3E-4
Solubility-limited release	Solubility limit (pCi/L)	1,260	1,680
Transport-limited release	K_d (cm^3/g), dispersion coefficient (cm)	K_d = 36 Disp = 7.5	K_d = 36 Disp = 8

previous model predictions showed a sharp increase in release over the ninth and tenth year. The data did not support this.

Figure 17 displays the predicted cumulative release of the four different scenarios and compares them to the measured data. All predict the cumulative release over 10 years to within 20% of the measured value. This is the result of the fitting procedure used to select the appropriate release and transport parameters.

The match at earlier times varies substantially based on the conceptual model. The high K_d model shows an extremely poor fit to the data at early times, under-predicting the measured release. If this model was the appropriate model, the next few years should have shown a marked increase in release rate. Although the measured cumulative release to the lysimeter increased by approximately 50% over the tenth year, this is still a smaller increase than predicted by the model. This behavior is similar to the "best" fit prediction of $K_d = 24\ cm^3/g$ based on 8 years of data. It appears likely that if this process was repeated after 2 more years of data collection, the "best" fit K_d would need to be increased again. It should be noted that for $K_d = 36\ cm^3/g$, the approximate pore velocity of the contaminant is 2.3 cm/yr. Thus, the release from the bottom of the waste form would not be expected to reach the leachate collector in over 20 years (50-cm travel distance) due to advection. Thus, dispersion is the major factor in transporting mass to the leachate collector under this scenario. Further, in this scenario, the waste form has released approximately 50% of its inventory. Since less than 0.2% has reached the leachate collector, most of the mass remains in the region between the collector and waste form. This indicates that relatively high concentrations of Sr-90 will be found when performing soil sampling beneath the waste form.

The three models with restricted waste form release matched the data better at all times (see Figure 17). The solubility-limited release and diffusion-controlled release models over-predict actual release for the first 9 years and slightly under-predict release after 10 years. The uniform release model closely tracked the data for the first 9 years and under-predicted the data in the tenth year. In all cases, the release starts to become

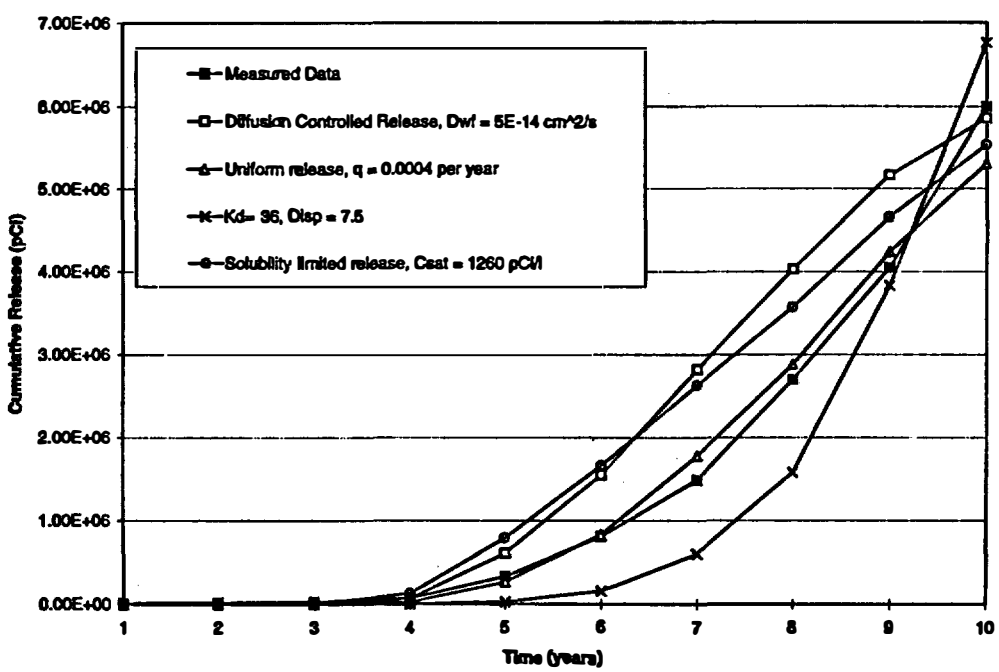


Figure 17. Comparison of ORNL Sr-90 cumulative release data with alternative conceptual models of release and transport.

substantial (>1E+5 pCi) after 4 or 5 years. At ORNL, the transport velocity using the measured K_d value is approximately 13 cm/yr. Thus, it would take about 4 years for waste released from the bottom of the waste form (50 cm above the leachate collector) to reach the leachate collector. In contrast to the high K_d model, these models restrict the amount of mass released from the waste form. Thus, they would predict much lower concentrations in the soil than the high K_d model.

Figure 18 presents the predicted cumulative release for the four alternative models over 20 years. The high K_d model predicts much greater release than the waste-form-restricted release models. This reflects the much higher predicted release from the waste form when using the base case waste form release parameters and altering the transport parameters to match the data. The three waste-form-limited release models showed similar release behavior over the 10- to 20-year period. After 20 years, the uniform release model predicted the greatest lysimeter release of the three.

ANL-E Lysimeter 5 Results—The "best" fit parameters for ANL-E lysimeter 5 for each of the alternative models are found in Table 9. The "best" fit parameters differ from those found at ORNL lysimeter 5. The waste form diffusion coefficient and fractional release rate are lower than at ORNL. The cause for this is the smaller measured cumulative fractional lysimeter release at ANL-E, which in turn leads to lower estimated waste form release rates. The ANL-E waste form "best" fit diffusion coefficient, 1E-15 cm^2/s, is a factor of 50 lower than the "best" fit value found for the ORNL data and almost six orders of magnitude lower than the laboratory-measured value. The solubility limit is slightly higher than found at ORNL due to the lower water flow rates found at ANL-E. (To obtain the same mass release with a lower flow rate, the concentration must be higher.) The reductions in release rate coefficients led to between 1 and 2E+7 pCi being released from the waste form over the 10-year period.

Figure 19 presents a comparison of the predicted and measured cumulative fractional release for all of the scenarios considered. At times less than 5 years, the measured data greatly exceed all of the predicted values. Substantial amounts of Sr-90 have reached the leachate collector in 2 years. Based on the measured K_d values and the average flow rate at ANL-E, the pore velocity of the contaminant should be 9.2 cm/yr. Therefore, is should take slightly more than 5 years for the contaminant to move the 50 cm from the bottom of the waste form to the lysimeter leachate collector. The arrival at times earlier than 5 years indicates either that preferential flow may be occurring and thereby leading to faster transport, or the distribution coefficient for the Sr-90 that is arriving early is not as high as the measured value of 6.3.

In Figure 19, the cumulative release based on the solubility-limited release model tracks the measured value the best. The uniform and diffusion-controlled waste form release models also follow the trends in the measured data reasonably well. The high K_d model shows a much different shaped curve than the data or any of the other models.

Figure 20 presents the predicted cumulative release over a 20-year period for the four conceptual models of the system. Similar to the results found at ORNL, the high K_d model predicts much greater release than the waste-form-restricted release models. The three waste-form-limited release models showed similar release behavior over the 10- to 20-year period. After 20 years, the uniform release model predicted the greatest lysimeter release of the three.

Analysis of Release of Cs-137 from ORNL Lysimeter 5 and Sr-90 Release from ORNL Lysimeter 1—Substantial release of cesium or strontium occurred in the lysimeter leachate collectors in only two cases other than the sand-filled units. Strontium-90 was found in ORNL lysimeter 1 at values of the same order of magnitude as the sand-filled lysimeter, and Cs-137 was found in ORNL lysimeter 5 at quantities greater than 1E+6 pCi.

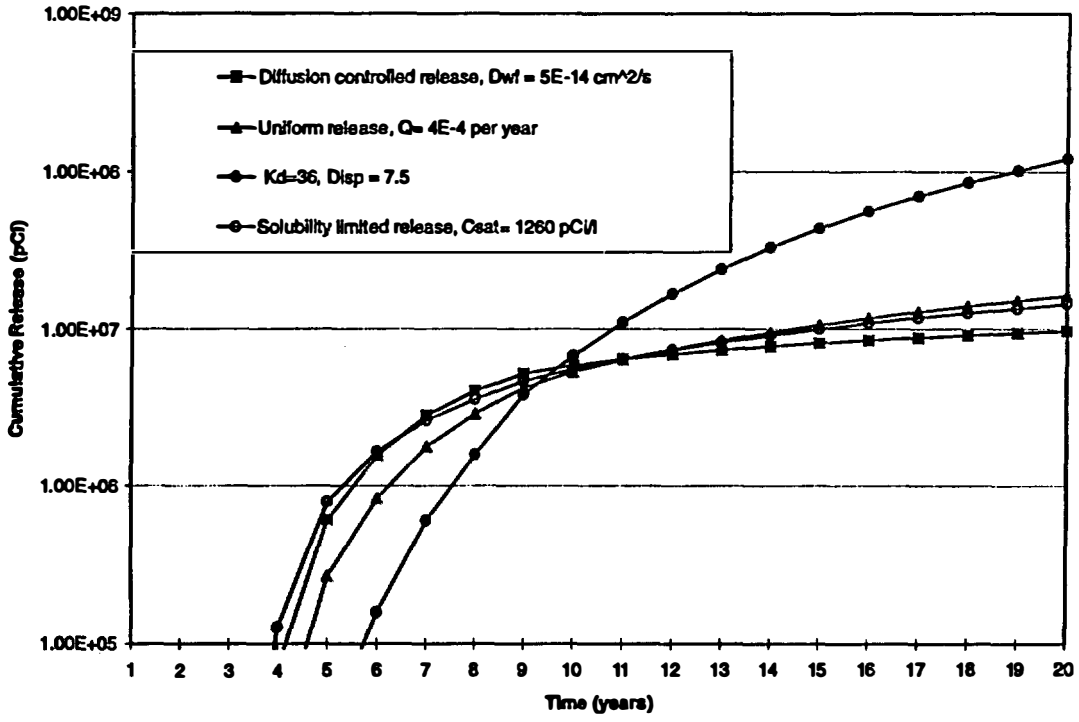


Figure 18. Comparison of predicted cumulative Sr-90 release over 20 years from ORNL lysimeter 5 based on alternative release and transport models.

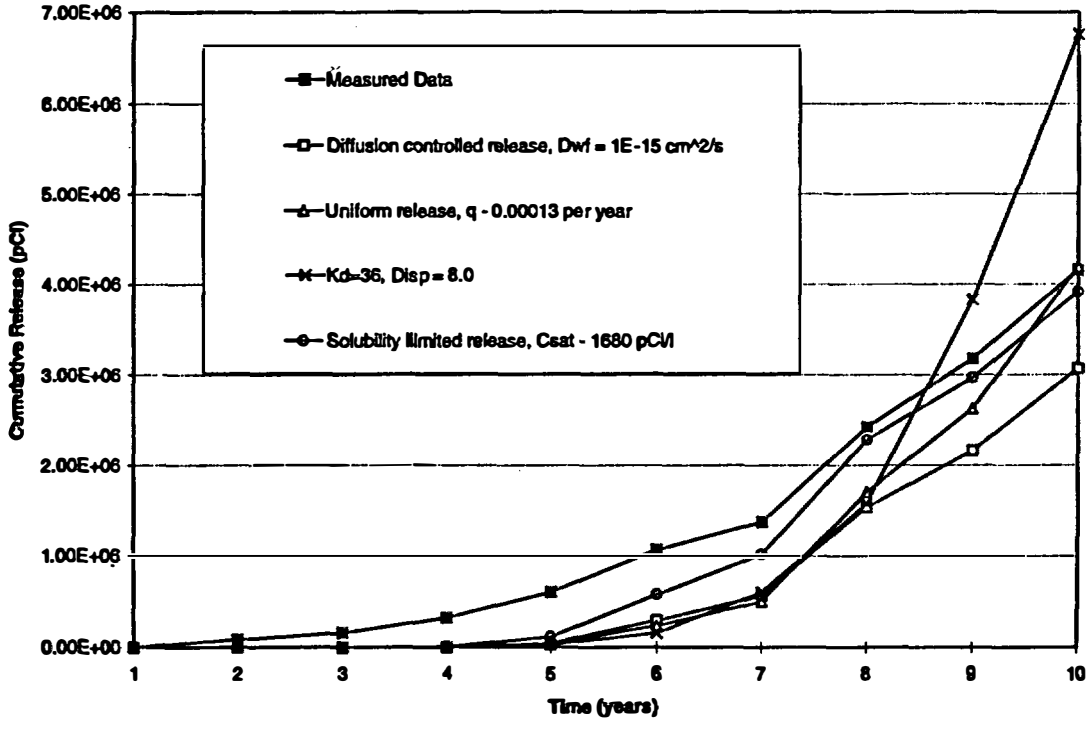


Figure 19. Comparison of measured and predicted cumulative Sr-90 release from ANL-E lysimeter 5.

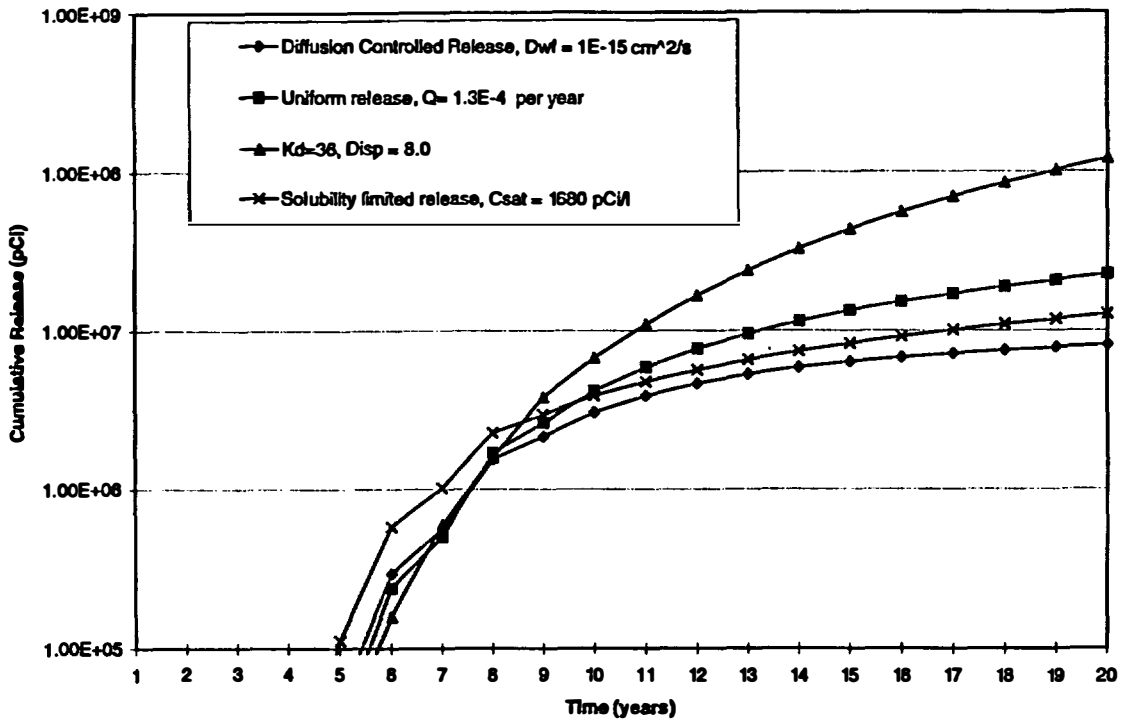


Figure 20. Comparison of predicted cumulative release of Sr-90 from ANL-E lysimeter 5 based on alternative release and transport scenarios.

At ORNL lysimeter 1, approximately 2.6E+6 pCi of Sr-90 was collected. The waste form is cement with PF-7 resin waste, similar to ANL-E lysimeter 5. This is slightly less than 2E-4 of the initial inventory. Thus, the cumulative release is an order of magnitude less than the sand-filled lysimeter at ORNL. The Sr-90 release from lysimeter 1 is consistent with a K_d value of between 20 and 40 cm^3/g. For this lysimeter and a K_d value of 20 cm^3/g, the average pore velocity of the contaminant would be less than 4 cm/yr. Thus, the transport of contaminant due to advection to the leachate collector would take approximately 14 years. Thus at this rate, if the distribution coefficient is 20 cm^3/g, the measured release would be due to dispersion phenomena. Since this value is not known, it is not possible to determine at this time if the release from the lysimeter is following the expected patterns (i.e., waste form release similar to laboratory-measured release rates and transport governed by laboratory-measured K_d values).

At ORNL lysimeter 5, Cs-137 releases showed a dramatic increase in year 9. In one sampling period, the release was 4.5E+6 pCi. In the previous 9 years, only 3.5E+5 pCi had been released in total. In the sampling period immediately following, another 8.9E+5 pCi was released. In the following two sampling periods, 1.5E+5 and 0.99E+5 pCi were released. The data clearly indicate a pulse of cesium passing through the system. This indicates some non-uniform process (i.e., preferential flow path, pulse release from the waste form, flooding, etc.) that cannot be modeled accurately using a one-dimensional model based on more uniform release processes. The cumulative fractional release in the one pulse was 3E-6. This small amount of release cannot be adequately modeled based on average properties of the system. It would be interesting to follow the time evolution of cesium release for longer periods of time.

Radionuclide Travel Time in Lysimeters. The data indicate minimal release for cesium from lysimeters 1 through 4 at ORNL. This is consistent with a measured K_d value of 37 cm^3/g. At this K_d value, the cesium would have a pore velocity of around 2 cm per year. Thus, it would take 25 years before the cesium would move 50 cm from the bottom of the waste form to the leachate collector.

Similar remarks apply to lysimeters 1 through 4 at ANL-E for both cesium and strontium. The measured K_d value for cesium is 390 cm^3/g and for strontium is 81 cm^3/g. Both of these values indicate that substantial time would be required to move the contaminants to the lysimeter leachate collectors.

As soil core samples are obtained beneath the waste form, it will be interesting to determine if the concentration profiles are consistent with the measured K_d values. For the relatively high measured K_d values, one would expect most of the activity to reside within 20 cm of where it is released (for K_d = 40 cm^3/g, movement of approximately 2 cm/yr for 10 years). Table 10 presents approximate transport distance over the 10-year period at ORNL and ANL-E based on average flow rates and moisture contents found in the sand lysimeters. A sand moisture content of 0.21 was assumed for the calculations. Transport would be different in the soil lysimeters due to the different flow rates and moisture contents. The distance presented in Table 10 is in the direction of flow and does not account for dispersion effects. It is meant as an approximate guideline to estimate transport distances. Distances traveled perpendicular to the flow direction would be much smaller.

Table 10. Radionuclide travel time as a function of retardation coefficient.

Site/Medium	Radionuclide	Darcy Flow Rate (cm/yr)	Bulk Density (g/cm^3)	K_d (cm^3/g)	Retardation Coefficient (dimensionless)	10-Year Travel Distance (cm)
ORNL/Soil	Cs-137	136.2	1.74[a]	37	307	21.1
ORNL/Sand	Cs-137	136.2	1.6	40	306	21.2
ORNL/Soil	Sr-90	136.2	1.74[a]	20-40	167 - 332	38.8 - 19.4
ORNL/Sand	Sr-90	136.2	1.6	6.3	49	132
ANL-E/Soil	Cs-137	93.5	1.74[a]	390	3,232	1.4
ANL-E/Sand	Cs-137	93.5	1.55	55	407	10.9
ANL-E/Soil	Sr-90	93.5	1.74[a]	81	672	6.6
ANL-E/Sand	Sr-90	93.5	1.55	6.4	48	92.3

a. The value for the density of ORNL soil was not available, so the value for the ANL-E soil was used (see Reference 37).

CONCLUSIONS

Resin Solidification

Both cement waste form types have exhibited an increase in strength with age and self irradiation dose. After a buildup over the first 8 years, that increase seems to have stabilized at about 5,500 psi for both types. Comparing these data to the irradiated compressive strength data obtained during INEEL testing in 1985 indicates that the strength of the cemented wastes will slowly decrease with age to about 3,000 psi in the future.

Both VES waste form types also exhibited an increase in strength with age and self-irradiation dose through 8 years. The maximum strength of the VES samples was between 4,500 and 5,000 psi. However, at 11 years, strength had dramatically decreased with the organic/inorganic resin waste form sample. At 12 years, both types of waste form samples suffered a further strength decrease. Comparing these data to those of the irradiation test compressive strength data of 1985 confirms that the VES sample strength will continue to decrease with age to about 2,000 psi in the next few years.

Field Testing

The lysimeter experiments that were operated successfully for over 10 years were shut down at the beginning of fiscal year 1996. A waste form and soil core sampling plan has been developed to more closely examine radionuclide movement in the lysimeter soil columns.

Studies were conducted to measure the partition coefficients of the lysimeter soils, inert sand, and moisture cup silica flour for strontium and cesium using rainwater collected at each site as the contact solution. The resulting coefficients are similar to those previously found in the literature.

The recent colloid investigation supports previous findings of a radiocolloid transport mechanism through the lysimeter soils. These findings are consistent with concerns regarding this transport mechanism as developed or discussed within several contemporary works on this subject. It was determined that Sr-90 is the primary radionuclide associated with collodial particles found in both ANL-E and ORNL leachates. That finding is well supported. No gamma or alpha radiation emissions could be detected on filters or filtrate. About 2% of the Sr-90 activity in the leachate seems to be associated with colloidal material. It is probable that particles observed in these investigations are a result of waste form degradation. The thought that the colloids previously identified are chemical precipitates occurring on the filter, can be rejected based on morphology.

The radionuclides Cs-137, Cs-134, and Sr-90 were detected in the upper region of the sand-filled lysimeter, 100 cm above the buried waste forms. It was conclusively shown that the radionuclides were released by the EPICOR-II waste forms and were not from outside the experiment. Physical examination discovered a root from an unidentified plant extending downward the length of the center core. Concentrations of Cs-137 peaked in both sand and root at the same depth levels. Also, concentrations of Cs-134, Cs-137, and Sr-90 within the sand core peaked at those same levels.

The intimate association of the radionuclide distribution with the root strongly suggests that the upward transport was within the evapotranspiration stream of the plant. Because the upward migration was unexpected, information that could shed more light on the exact mechanism of transport was not collected or preserved. However, the observation does have implications for locations where radionuclides may be disposed in the shallow subsurface. The presence of plants may enhance the upward movement of radionuclides to the surface where direct exposures or offsite migration are possible.

Attempts were unsuccessful to model the release of Sr-90 from sand-filled lysimeters at ANL-E and ORNL based on laboratory values of waste form release properties and soil distribution coefficients using DUST. Model predictions exceeded measured results by two orders of

magnitude. Attempts to match the experimental data with model predictions were made by either limiting the waste form release rate or the transport rate in the soil. Calculations limiting the waste form release rate were performed for three different conceptual models: diffusion-controlled release, uniform release, and solubility-limited release. In each case, the amount of mass released was approximately two orders of magnitude less than expected based on laboratory data. Best estimates for these parameters were made, and the fits to the data were reasonably good. Potential reasons for the release rates not matching the experimental data could be goechemical controls on release (e.g., formation of strontium carbonate or other strontium species that are immobile) or the inapplicability of saturated leach tests for the unsaturated field conditions. Calculations limiting the transport rate through the sand were done by increasing the partition coefficient by a factor of approximately six over its measured value to a value of 36. Through appropriate choice of the dispersion coefficient, a relatively good fit to the data was obtained. A potential reason why the measured K_d value does not apply could be the use of saturated systems when measuring K_d; while in the field, the systems are unsaturated. The degree of saturation for the sand-filled lysimeters is approximately 50%.

REFERENCES

1. R. C. Schmitt and H. W. Reno, *Program Plan of the EPICOR and Waste Research and Disposition Program of the Technical Support Branch*, EGG-TMI-6521, revised December 1983.

2. J. W. McConnell, Jr., *EPICOR-II Resin/Liner Research Plan*, EGG-TMI-6198, March 1983.

3. J. W. McConnell, Jr. and R. D. Sanders, Sr., *EPICOR-II Resin Degradation Results from First Samples of PF-8 and PF-20*, NUREG/CR-4150, EGG-2176, Idaho National Engineering Laboratory, EG&G Idaho, Inc., May 1985.

4. R. D. Sanders, Sr. and J. W. McConnell, Jr., *EPICOR-II Resin Degradation Results from Second Samples of PF-8 and PF-20*, NUREG/CR-4608, EGG-2452, Idaho National Engineering Laboratory, EG&G Idaho, Inc., October 1986.

5. J. W. McConnell, Jr., D. A. Johnson, and R. D. Sanders, Sr., *Radiation Degradation in EPICOR-II Ion Exchange Resins, Final Report*, NUREG/CR-5594, EGG-2603, Idaho National Engineering Laboratory, EG&G Idaho, Inc., September 1990.

6. U.S. Nuclear Regulatory Commission, "Technical Position on Waste Form," Revision 1, Low-Level Waste Management Branch, Washington, D.C., January 1991.

7. R. M. Neilson, Jr. and J. W. McConnell, Jr., *Solidification of EPICOR-II Resin Waste Forms*, GEND-INF-055, August 1984.

8. R. M. Neilson, Jr. and J. W. McConnell, Jr., *EPICOR-II Resin Waste Form Testing*, NUREG/CR-4637, EGG-2457, Idaho National Engineering Laboratory, EG&G Idaho, Inc., October 1986.

9. J. W. McConnell, Jr. and R. M. Neilson, Jr., *The Effects of Aging on Compressive Strength of Low-Level Radioactive Waste Form Samples*, NUREG/CR-6392, INEL-95/0506, Idaho National Engineering Laboratory, Lockheed Idaho Technologies Company, June 1996.

10. American Society for Testing and Materials, ASTM C39, "Standard Test Method for Compressive Strength of Cylindrical Concrete Specimens," 1972.

11. American Society for Testing and Materials, ASTM C617, "Standard Method of Capping Cylindrical Concrete Specimens," 1976.

12. R. D. Rogers, J. W. McConnell, Jr., E. C. Davis, and M. W. Findlay, *Field Testing of Waste Forms Containing EPICOR-II Ion Exchange Resins Using Lysimeters*, NUREG/CR-4498, EGG-2438, Idaho National Engineering Laboratory, EG&G Idaho, Inc., June 1986.

13. R. D. Rogers, J. W. McConnell, Jr., M. W. Findlay, and E. C. Davis, *Lysimeter Data from EPICOR-II Waste Forms—Fiscal Year 1986*, EGG-TMI-7417, October 1986.

14. R. D. Rogers, J. W. McConnell, Jr., M. W. Findlay, and E. C. Davis, *Lysimeter Data from EPICOR-II Waste Forms—Fiscal Year 1987*, EGG-TMI-8004, March 1988.

15. R. D. Rogers, J. W. McConnell, Jr., E. C. Davis, and J. D. Jastrow, *Annual Report of the TMI-2 EPICOR-II Resin/Liner Investigation: Low-Level Waste Data Base Development Program for Fiscal Year 1988*, NUREG/CR-5229, EGG-2553, Vol. 1, Idaho National Engineering Laboratory, EG&G Idaho, Inc., December 1988.

16. J. W. McConnell, Jr., R. D. Rogers, E. C. Davis, and J. D. Jastrow, *Annual Report of the TMI-2 EPICOR-II Resin/Liner Investigation: Low-Level Waste Data Base Development Program for Fiscal Year 1989*, NUREG/CR-5229, EGG-2577, Vol. 2, Idaho National Engineering Laboratory, EG&G Idaho, Inc., February 1990.

17. J. W. McConnell, Jr., R. D. Rogers, D. A. Johnson, J. D. Jastrow, and D. S. Wickliff, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1990*, NUREG/CR-5229, EGG-2577, Vol. 3, Idaho National Engineering Laboratory, EG&G Idaho, Inc., December 1990.

18. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, D. S. Wickliff, and R. R. Brey, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1991*, NUREG/CR-5229, EGG-2577, Vol. 4, Idaho National Engineering Laboratory, EG&G Idaho, Inc., January 1992.

19. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, and D. S. Wickliff, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1992*, NUREG/CR-5229, EGG-2577, Vol. 5, Idaho National Engineering Laboratory, EG&G Idaho, Inc., February 1993.

20. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, and T. M. Sullivan, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1993*, NUREG/CR-5229, EGG-2577, Vol. 6, Idaho National Engineering Laboratory, EG&G Idaho, Inc., May 1994.

21. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, and T. M. Sullivan, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1994*, NUREG/CR-5229, INEL-94/0278, Vol. 7, Idaho National Engineering Laboratory, Lockheed Idaho Technologies Company, May 1995.

22. J. W. McConnell Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, I. L. Larsen, and T. M. Sullivan, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1995*, NUREG/CR-5229, INEL-94/0278, Vol. 8, Idaho National Engineering Laboratory, Lockheed Idaho Technologies Company, June 1996.

23. R. R. Brey, J. W. McConnell, Jr., T. M. Sullivan, and J. D. Jastrow, "A Preliminary Investigation of the Existence of Radiocolloids in Leachate from the NRC Field Lysimeter Investigations," *Waste Management*, Vol. 14, No. 7, 1994, pp. 581–588.

24. I. M. Litator, G. M. Barth, and E. M. Zika, "Fate and Transport of Plutonium-239 + 240 and Americium-241 in the Soil of Rocky Flats, Colorado," *Journal of Environmental Quality*, Vol. 25, No. 4, 1996, pp. 1086–1094.

25. R. R. Brey, *A Study of the Transport Characteristics and Mechanisms of Submicron Particles in Unsaturated Porous Media*, Ph.D. Dissertation: Purdue University, West Lafayette, Indiana, 1994.

26. R. W. Puls and R. M. Powell, "Transport of Inorganic Colloids Through Natural Aquifer Material: Implications for Contaminant Transport," *Environmental Science and Technology*, Vol. 26, No. 3, 1992, pp. 614–621.

27. J. F. McCarthy and J. M. Zachara, "Subsurface Transport of Contaminants," *Environmental Science and Technology*, Vol. 23, No. 5, 1989, pp. 496–502.

28. S. Mindess and J. Young, *Concrete*, Francis Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1981.

29. B. L. Sawhney, "Kinetics of Cesium Sorption by Clay Minerals," *Soil Sci. Soc. Am. Proc.*, Vol. 30, 1966, pp. 565–569.

30. D. D. Eberl, "Alkali Cation Selectivity and Fixation by Clay Minerals," *Clays and Clay Minerals*, Vol. 28, 1980, pp. 161–172.

31. R. N. J. Comans, M. Haller, and P. DePreter, "Sorption of Cesium on Illite: Nonequilibrium Behavior and Reversibility," *Geochimica et Cosmochimica Acta*, Vol. 55, 1991, pp. 433–440.

32. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, T. M. Sullivan, R. M. Neilson, Jr., and L. D. Hilton, *Field Lysimeter Investigations—Test Results: Low-Level Waste Data Base Development Program: Test Results for Fiscal Years 1994 and 1995*, NUREG/CR-6256, INEL-95/0073, Vol. 3, Idaho National Engineering Laboratory, Lockheed Idaho Technologies Company, June 1996.

33. C. W. Francis, "Radiostrontium Movement in Soils and Uptake in Plants," *DOE Critical Review Series*, TID-27564, Technical Information Center, U.S. Department of Energy, Oak Ridge, TN, 1978.

34. T. M. Sullivan, J. W. McConnell, Jr., R. D. Rogers, and I. T. Fitzgerald, "Modeling Field Lysimeter Release Data Using the Computer Code DUST," *Waste Management '93, Proceedings of the Symposia on Waste Management, Tucson, Arizona, February 28 - March 4, 1993*, Vol. 1, pp. 781–785.

35. T. M. Sullivan, *Selection of Models to Calculate the LLW Source Term*, NUREG/CR-5773, Brookhaven National Laboratory, October 1991.

36. T. M. Sullivan, *DUST, Disposal Unit Source Term: Data Input Guide*, NUREG/CR-6041, BNL-NUREG-52375, Brookhaven National Laboratory, 1993.

37. J. W. McConnell, Jr., R. D. Rogers, M. W. Findlay, E. C. Davis, J. D. Jastrow, R. M. Neilson, Jr., and L. D. Hilton, *Field Lysimeter Investigations—Test Results: Low-Level Waste Data Base Development Programs: Test Results for Fiscal Years 1986, 1987, 1988, and 1989*, NUREG/CR-6256, INEL-95/0073, Vol. 1, Idaho National Engineering Laboratory, Lockheed Idaho Technologies Company, May 1995.

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P. R. Reed, NRC Project Manager

11. ABSTRACT (200 words or less)

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program, funded by the U.S. Nuclear Regulatory Commission, is (a) studying the degradation effects in organic ion-exchange resins caused by radiation, (b) examining the adequacy of test procedures recommended in the Branch Technical Position on Waste Form to meet the requirements of 10 CFR 61 using solidified ion-exchange resins, (c) obtaining performance information on solidified ion-exchange resins in a disposal environment, and (d) determining the condition of liners used to dispose the ion-exchange resins.

During the field testing experiments, both portland type I–II cement and Dow vinyl ester-styrene waste form samples were tested in lysimeter arrays located at Argonne National Laboratory-East (ANL-E) in Illinois and at Oak Ridge National Laboratory (ORNL). The study was designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over an extended period. Those experiments have been shut down and are to be exhumed. This report discusses the plans for removal, sampling, and analysis of waste form and soil cores from the lysimeters. Results of partition coefficient determinations are presented, as well as application of a source term computer code using those coefficients to predict the lysimeter results. A study of radionuclide-containing colloids associated with the leachate waters removed from these lysimeters is described. An update of upward migration of radionuclides in the sand-filled lysimeter at ORNL is included.

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