

RESULTS OF FIELD TESTING OF WASTE FORMS USING LYSIMETERS

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ABSTRACT

The TMI-2 EPICOR-II Resin/Liner Low-Level Waste Data Base Development Program*, funded by the U.S. Nuclear Regulatory Commission (NRC), is obtaining information on the performance of radioactive waste in a disposal environment. Waste forms fabricated using ion-exchange resins from EPICOR-II prefilters employed in the cleanup of the Three Mile Island Nuclear Power Station are being tested to: (a) develop a low-level waste data base, and (b) obtain information on survivability of waste forms in a disposal environment. This paper updates field testing of those waste forms during FY-1989. The results of the lysimeters are presented and the use of lysimeter data in performance assessment is discussed.

INTRODUCTION

The purpose of the field testing task is to expose samples of solidified resin waste to the actual physical, chemical, and microbiological conditions of a disposal environment.(1,2) Emphasis is placed on investigating the requirements of 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste."(3) The waste forms are composed of radionuclide-loaded ion-exchange media that were solidified in matrixes of cement and DOW polymer. Waste forms fabricated with the same formulations were also subjected to the tests specified in the "Technical Position on Waste Form,"(4) issued by the U.S. Nuclear Regulatory Commission (NRC).

METHODS AND MATERIALS

Wastes used in the experiment were taken from EPICOR-II prefilter liners and include a mixture of synthetic organic ion-exchange resins from prefilter PF-7 (herein referred to as Type 1 waste) and a mixture of organic-exchange resins and an inorganic zeolite from prefilter PF-24 (herein referred to as Type 2 waste). Solidification agents employed to produce the 4.8 x 7.6 cm cylindrical waste forms (Fig. 1) used in the study were Portland Type I-II cement and DOW vinyl ester-styrene (VES).(5) Seven of the waste forms were stacked end-to-end and inserted into each lysimeter to provide a 1-L volume. Table I lists waste form description by lysimeter number. Waste Type 1 contained 71% of the radionuclides as Cs-137 while Type 2 contained 94% Cs-137. Type 1 waste also contained 25% Sr-90 and Type 2 contained 1% Sr-90. There were also significant amounts of Cs-134 and trace amounts of Co-60 and Sb-125 found in those wastes.

There are 10 lysimeters, 5 at Oak Ridge National Laboratory (ORNL) and 5 at Argonne National Laboratory-East (ANL-E). Lysimeters used in this study were designed to be self-contained units that will be disposed of at the

termination of the 20-year study. Each lysimeter is a 0.91- x 3.12-m right-circular cylinder divided into an upper compartment, which contains fill material, waste forms, and instrumentation, and an empty lower compartment for collecting leachate (Fig. 2). Four lysimeters at each site are filled with soil, while a fifth (used as a control) is filled with inert silica oxide sand. The lysimeters at ANL-E contain soil indigenous to the site, while the ORNL lysimeters contain soil taken from Savannah River Laboratory, SC. Instrumentation in each lysimeter includes moisture cup soil-water samplers and soil moisture/temperature probes. The probes are connected to an onsite data acquisition system (DAS), which also collects data from a field meteorological station located at each site.

TABLE I
LYSIMETER WASTE-FORM COMPOSITION

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

Each month, data stored on a cassette tape are retrieved from the DAS and translated into a PC-compatible disk file. On approximately a quarterly basis, water is drawn from the moisture cup soil-water samplers and the leachate collection compartments to track the migration of radionuclides. The water samples are analyzed for beta- and gamma-producing nuclides. Details on waste-form formu-

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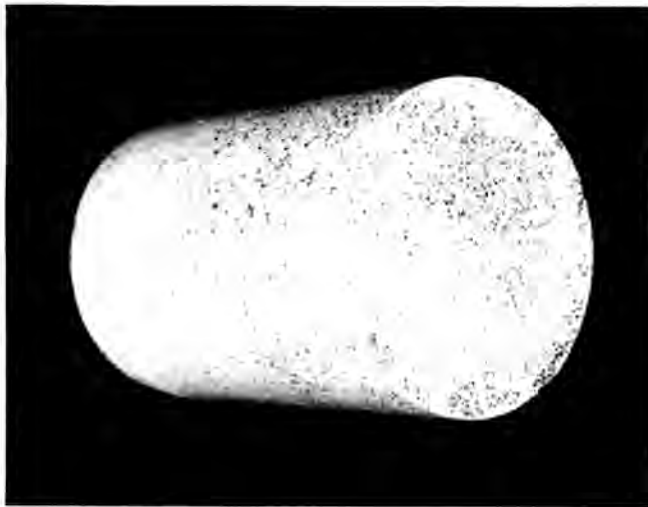


Fig. 1. An Example of an EPICOR-II Waste Form.

lation and testing, lysimeter design, installation, instrumentation, operation, and data acquisition are provided in Refs. 5 through 12.

Monitoring of moisture cups began with collecting liquid samples in September 1985 (2 to 3 months from the time of placement) and has continued about once every 3 months thereafter. The monitoring includes sampling of liquids from locations near the waste form and radiochemical analysis of those liquids. Soil moisture and temperature at three

elevations in each lysimeter, along with a complete weather history, are recorded on a continuing basis by the DAS.

RESULTS AND DISCUSSION

Weather and Soil Data

Precipitation, air temperature, wind speed, and relative humidity were recorded by the ANL-E and ORNL DAS during the 12-month reporting period. Total precipitation for the period was 72.9 cm at ANL-E and 157.4 cm at ORNL. ANL-E was below their normal annual rainfall of 85.2 cm;(13), while ORNL was at about 113% of their normal annual rainfall of 138.8 cm.(14) Fig. 3 shows the cumulative precipitation for each site since the initiation of field work.

Air temperature data from ANL-E indicate periods of freezing temperatures from mid-November 1988 until near mid-March 1989. ORNL experienced some days where there was an air temperature as low as 0°C (typical data shown in Refs. 8 through 12).

Examples of the lysimeter soil temperature data recorded over 1-year periods at ANL-E and ORNL can be found in Refs. 9 through 12. At no time during the FY-1989 reporting period was a freezing temperature recorded by a properly functioning detector at any depth within a lysimeter. A number of detectors have failed at ANL-E, but redundancy has prevented any loss of information.

Examples of data from the moisture probes at both ANL-E and ORNL can be found in Refs. 9 through 12. Data recorded in FY-1989 indicate that the lysimeter soil columns at both sites have remained moist during the reporting period. The moisture content of the soil column of each

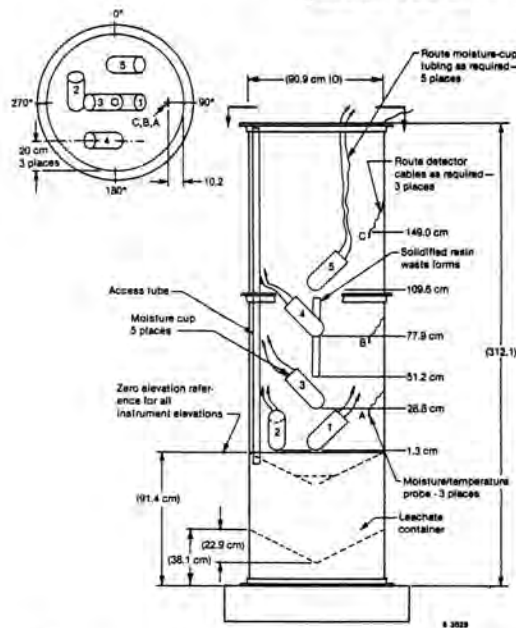


Fig. 2. Lysimeter Vessel Component Locations.

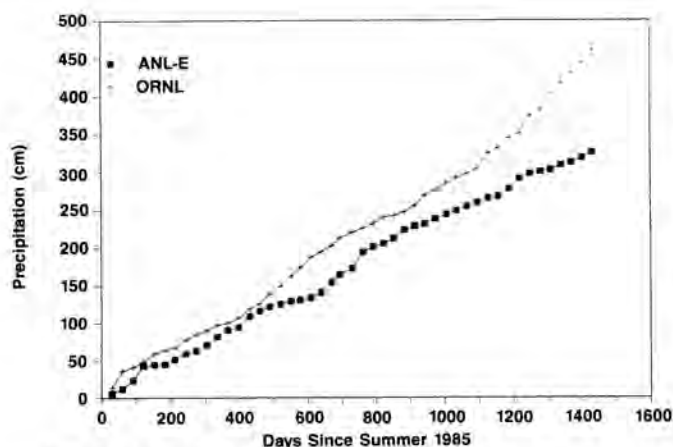


Fig. 3. ANL-E and ORNL Cumulative Precipitation.

lysimeter over time (as determined by averaging the outputs of the all probes in each lysimeter) showed that variation in moisture data for lysimeters at each site were relatively similar. The average soil moisture of ANL-E lysimeter soils was 55.7% of the soil moisture holding capacity; for ORNL, this value was 35.9%.

By using the cumulative rainfall data from each site since the time the lysimeters were placed in operation (Fig. 3), it is possible to calculate the volume of water that has been received by the exposed lysimeter surfaces (6489.5 cm²). The cumulative volume of precipitation received by each ANL-E lysimeter was 2111.7 L; at ORNL, this value was 2991.6 L. It appears that the throughput is dependent on site conditions (period of time soil surface was frozen, amount of precipitation received as snow, etc.) and lysimeter fill material. At ANL-E, an average of 998.9 ± 255.6 L, or 47.3% of total precipitation, passed through the soil lysimeters; for the control lysimeter, this value was 2080 L, or 98.5% of available precipitation. For ORNL, the values were 2558 ± 28.5 L (85.5%) for the soil-filled lysimeters and 2991 L (100%) for the control lysimeter. Not only does precipitation have more of an opportunity to move into the ORNL lysimeters (an observation made by comparing the control lysimeters at each site), but the ORNL soils were more permeable than the ANL-E soils (an observation made by comparing the control lysimeter at each site with that site's soil lysimeters).

The uniformity of soil moisture in the ANL-E lysimeters is somewhat surprising given the long-term decrease in water infiltration in ANL 1 and 2, and now 3. Action to improve drainage of these two lysimeters was begun in July 1987; however, it has now become obvious that the initial rate of drainage cannot be restored. This is due to the present structureless condition of the soils. The present conditions are now thought to be indicative of what would be found if a disposal trench were constructed in this soil. No further effort to improve drainage of these lysimeters is anticipated. Instead, water will no longer be allowed to pond

on the soil surface. At some time interval after a rain event, water in excess of 2 to 3 cm in depth will be removed. Records of the amounts of water removed will be maintained.

The total volumes of precipitation that have moved through the lysimeters represent an average 1.40 pore volumes for the ANL-E soil lysimeters and 3.60 pore volumes for soil lysimeters at ORNL, while the control lysimeters at ANL-E and ORNL were 3.57 and 5.34 pore volumes, respectively. Theoretically, by this time, all of the water held in pore spaces of the soil column in the ANL-E lysimeters has been replaced. In the ORNL lysimeters, more than three times the original amount of water held in pore spaces of the soil column has been replaced.

Radionuclide Analysis

Tabulated results of beta and gamma analysis of samples taken during the period are found in Tables II and III. Four samples were taken at each site during the 12-month period. The cumulative amounts of nuclides as determined in water samples obtained from moisture cups 3 and leachate collectors for all sampling periods are displayed graphically in Figs. 4 through 10.

It is apparent from these data that not all nuclides are appearing consistently in the water obtained from the moisture cups and leachate collectors. The nuclide that appears with the most regularity at both sites is Sr-90. Consistent significant occurrences, however, have been observed in all the cups 3 at ANL, the cups 3 at ORNL, except 4-3 (though it was found in this cup during the last two samplings), and the number 5 leachate collectors at both sites (Figs. 4 through 7). There continue to be standout amounts of Sr-90 retrieved from moisture cup samples at both sites. Those include 270,090 pCi from cup 3-3 at ANL (Fig. 4) and 17,285 pCi from cup 1-3 at ORNL (Fig. 5).

As noted in the Waste-Form Leachability section of last year's report,(11) Sr-90 leachates appear to move from these waste forms more rapidly than Cs-137. The cumulative amounts of Sr-90 collected by ANL-E cup 3-3 and ORNL cup 1-3 (Figs. 4 and 5, respectively) when plotted with time form what appear to be the beginnings of typical leach rate curves, indicating that nuclide release to cups 3 in these two lysimeters is limited by release from the waste forms. Also, while these cumulative totals appear large when compared to other lysimeter experiments, the total in the ANL cup 3-3 represents about 0.001% of the waste form inventory.(12)

At ANL, Sr-90 retrieved from the cups 3 of the soil lysimeters range from 74% to 3200% of that found in the leachate collectors, while at ORNL these values range from 0.1% to 70%. These increases are the results of both an increased quantity of Sr-90 moving into the area near the

TABLE II
Results of Beta and Gamma Analysis of ANL-E Soil Moisture and Leachate Samples--Year 4--1988-89

Sample Identif- ication	Concentration (pCi/L) ^a											
	Co-60				Cs-137				Sr-90			
	September 88	November 88	April 89	June 89	September 88	November 88	April 89	June 89	September 1988	November 1988	April 1989	June 1989
LYS 1 ^b	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
LYS 2	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
LYS 3	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	9.1 ± 1.3
LYS 4	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
LYS 5	<1	<1	<1	<1	2 ± 1	2 ± 1	<1	<1	274 ± 25	387 ± 10	276 ± 5	417 ± 17
LYS 1-3 ^c	<5	<5	<5	<5	<5	20 ± 18	<5	<5	10E4 ± 115	1.2E4 ± 75	8479 ± 90	1.09E4 ± 171
LYS 2-3	<5	<5	<5	<5	2171 ± 59	1294 ± 51	625 ± 36	1309 ± 48	1324 ± 32	2000 ± 31	1720 ± 154	2031 ± 36
LYS 3-3	<5	<5	<5	760 ± 45	<5	<5	<5	3638 ± 86	3.1E5 ± 4466	5.4E5 ± 9740	5.8E5 ± 5017	6.12E5 ± 3554
LYS 4-3	<5	<5	<5	<5	<5	<5	<5	<5	1818 ± 38	1108 ± 28	1968 ± 65	2578 ± 49
LYS-5-3	<5	<5	<5	<5	1539 ± 58	3501 ± 94	2572 ± 70	9389 ± 924	6972 ± 73	5088 ± 119	8042 ± 55	1.28E4 ± 305

a. Concentration ± 2 sigma.

b. Leachate sample from 1-L sample size.

c. Moisture cup sample from ~0.1-L sample size.

TABLE III
Results of Beta and Gamma Analysis of ORNL Soil Moisture and Leachate Samples--Year4--1988-89

Sample Identif- ication	Concentration (pCi/L) ^a															
	Co-60				Cs-137				Sb-125				Sr-90			
	Nov 88	Mar 89	May 89	June 89	Nov 88	Mar 89	May 89	June 89	Nov 88	Mar 89	May 89	June 89	Nov 89	Mar 89	May 89	June 89
LYS 1 ^b	<3	<3	<3	<5	<3	<3	<5	20 _{±4}	<8	<8	<8	<8	<1	5.8 _{±5.1}	<3	<3
LYS 2	<3	<3	<3	<5	<3	<3	<5	10 _{±4}	<5	<8	<8	<8	<1	<5	1.6 _{±3.8}	<3
LYS 3	<3	<3	<5	<3	8.6 _{±4.5}	<5	<5	6 _{±4}	<8	<11	<8	<8	1.6 _{±3.2}	5.7 _{±4.1}	1.9 _{±3.8}	0.3 _{±3.5}
LYS 4	<3	<3	<5	<3	13 _{±2.7}	<3	<5	4 _{±2}	<3	<8	<11	<8	<1	<3	<3	2.4 _{±3.5}
LYS 5	2.2 _{±4.3}	<3	<3	<5	15 _{±3.5}	<3	378 _{±27}	108 _{±5}	18 _{±5.1}	<8	22 _{±14}	<11	32 _{±8.1}	32 _{±8.1}	35 _{±5.4}	73 _{±10.8}
LYS 1-3 ^c	<24	<27	<27	<27	<25	<22	<24	<27	<54	<54	<54	<54	3E4 _{±3000}	1300 _{±54}	4.1E4 _{±2700}	6.5E4 _{±2700}
LYS 2-3	<22	<22	<27	30 _{±22}	<19	<25	<25	<22	<54	<54	<54	<54	5100 _{±540}	220 _{±19}	7300 _{±2700}	8100 _{±540}
LYS 3-3	<22	<27	NA ^d	<27	<16	<22	NA	20 _{±20}	<54	<54	<54	<54	7800 _{±540}	570 _{±27}	NA	3.2E4 _{±2700}
LYS 4-3	38 _{±35}	<22	<19	<27	32 _{±24}	<22	<19	<24	<54	<54	<54	<54	<11	<3	29.7 _{±21.6}	67.6 _{±32.4}
LYS 5-3	<27	<19	<19	<22	351 _{±27}	405 _{±27}	568 _{±27}	703 _{±27}	<54	<54	<54	<54	68 _{±22}	4.3 _{±20}	135 _{±32.4}	205 _{±43.4}
LYS 1-1	<27	<27	<54	<27	<24	35 _{±41}	<24	<54	<54	<54	<54	<54	14 _{±17}	<3	13.8 _{±16}	9.5 _{±17.6}
LYS 2-1	<19	<27	<27	<24	<19	<22	<19	<54	<54	<54	<54	<54	<1	1.7 _{±2.2}	6.5 _{±13.8}	11.9 _{±19.2}
LYS 3-1	<22	<19	<24	<27	<19	<24	<24	<54	<54	<54	<54	<54	<1	<3	3 _{±14.1}	<13
LYS 4-1	<19	<22	<22	<27	<27	<27	<27	<54	<54	<54	<54	<54	<1	3.0 _{±4.3}	<14	2.4 _{±17.3}
LYS 5-1	<27	<22	<22	<27	<27	<27	<27	<54	<54	<54	<54	<54	70.3 _{±21.6}	<3	43.2 _{±21.6}	23.8 _{±24.6}

a. Concentration ± 2 sigma.

b. 1-L subsample from leachate collection.

c. Total moisture cup sample ~0.1-L sample site.

d. Sample not analyzed.

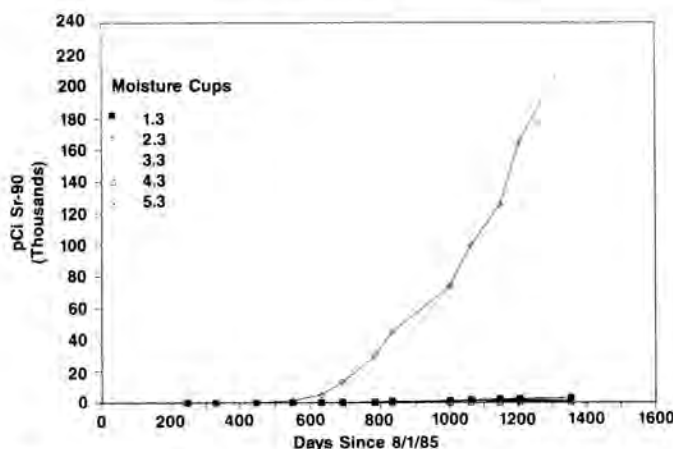


Fig. 4. ANL-E Cumulative Sr-90 Collected in Moisture Cup Number 3.

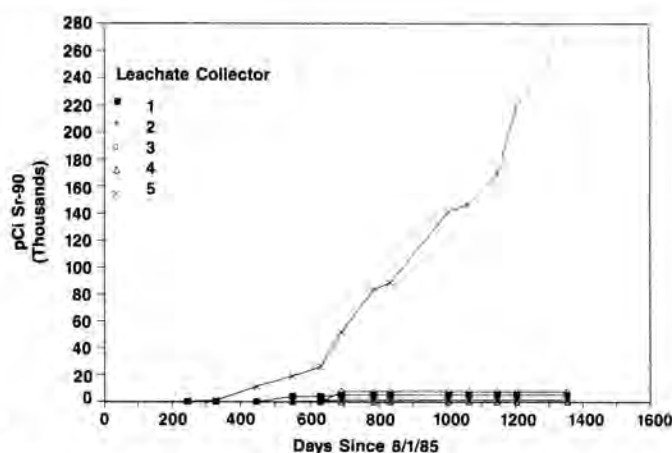


Fig. 6. ANL-E Cumulative Sr-90 Collected in Lysimeter Leachate Collectors.

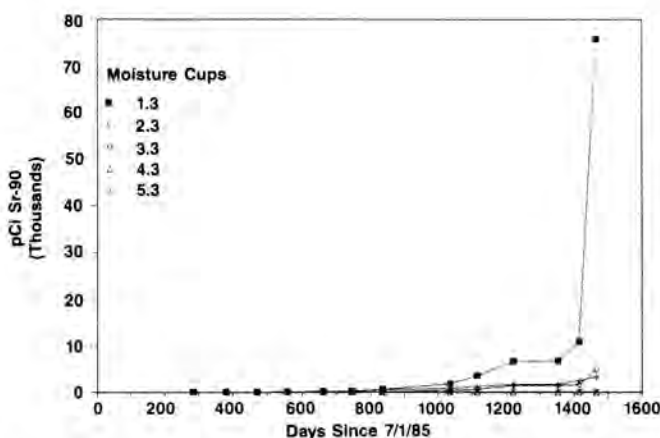


Fig. 5. ORNL Cumulative Sr-90 Collected in Moisture Cup Number 3.

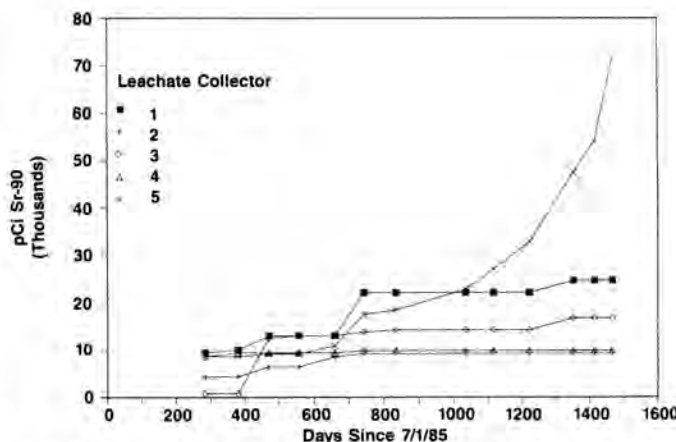


Fig. 7. ORNL Cumulative Sr-90 Collected in Lysimeter Leachate Collectors.

cups and a decrease in movement of that radionuclide through the entire soil profile into the leachate collectors.

During the past 12 months, only the leachate water from the control lysimeters and none from the soil lysimeters at each site contained significant amounts of Sr-90. This is comparable to the previous years findings (Refs. 9, 10, and 11) and is in sharp contrast to the cups 3 data, which continue to demonstrate that substantial amounts of Sr-90 are still being released from the waste forms. The percent of total Sr-90 being measured in the leachate collectors and cups 3 is inconsistent.(12) This could indicate differences in waste form performance at the two sites. These data are interesting, because the waste forms at each site have been experiencing similar exposure to moisture and temperature.(12)

Gamma-producing nuclides have occurred with regularity at ANL-E and are again present at ORNL. ANL cup

2-3, below a cement waste form containing large amounts of Cs-137, continues to receive Cs-137 (Table II), with a significant increase in quantities of this radionuclide appearing this year after peaking in the February 1987 sample (Fig. 8). The quantity of this nuclide increased in each of the sampling periods during the year with an abrupt increase during the last sampling period. There continues to be no sustained occurrence of Cs-137 in any of the ANL leachate water. In sharp contrast to last year, Cs-137 has been detected consistently in water from the ORNL lysimeters. Measurable amounts of Cs-137 began to occur in ORNL cup 5-3 during the April 1988 sample and have continued in subsequent samplings for a total of 233 pCi (Table III). Breakthrough of Cs-137 into the ORNL 5 leachate collector occurred in November 1988 some 7 months after its occurrence in the moisture cup ORNL 5-3, and thus far a total of 100,096 pCi have passed through the lysimeter. In addition,

TABLE III
Results of Beta and Gamma Analysis of ORNL Soil Moisture and Leachate Samples--Year 4--1988-89

Sample Identif- ication	Concentration (pCi/L) ^a															
	Co-60				Cs-137				Sb-125				Sr-90			
	Nov 88	Mar 89	May 89	June 89	Nov 88	Mar 89	May 89	June 89	Nov 88	Mar 89	May 89	June 89	Nov 89	Mar 89	May 89	June 89
LYS 1 ^b	<3	<3	<3	<5	<3	<3	<5	20±4	<8	<8	<8	<8	<1	5.8±5.1	<3	<3
LYS 2	<3	<3	<3	<5	<3	<3	<5	10±4	<5	<8	<8	<8	<1	<5	1.6±3.8	<3
LYS 3	<3	<3	<5	<3	8.6±4.5	<5	<5	6±4	<8	<11	<8	<8	1.6±3.2	5.7±4.1	1.9±3.8	0.3±3.5
LYS 4	<3	<3	<5	<3	13±2.7	<3	<5	4±2	<3	<8	<11	<8	<1	<3	<3	2.4±3.5
LYS 5	2.2±4.3	<3	<3	<5	15±3.5	<3	378±27	108±5	18±5.1	<8	22±14	<11	32±8.1	32±8.1	35±5.4	73±10.8
LYS 1-3 ^c	<24	<27	<27	<27	<25	<22	<24	<27	<54	<54	<54	<54	3E4±3000	1300±54	4.1E4±2700	6.5E4±2700
LYS 2-3	<22	<22	<27	30±22	<19	<25	<25	<22	<54	<54	<54	<54	5100±540	220±19	7300±2700	8100±540
LYS 3-3	<22	<27	NA ^d	<27	<16	<22	NA	20±20	<54	<54	<54	<54	7800±540	570±27	NA	3.2E4±2700
LYS 4-3	38±35	<22	<19	<27	32±24	<22	<19	<24	<54	<54	<54	<54	<11	<3	29.7±21.6	67.6±32.4
LYS 5-3	<27	<19	<19	<22	351±27	405±27	568±27	703±27	<54	<54	<54	<54	68±22	4.3±20	135±32.4	205±43.4
LYS 1-1	<27	<27	<54	<27	<24	35±41	<24	<54	<54	<54	<54	<54	14±17	<3	13.8±16	9.5±17.6
LYS 2-1	<19	<27	<27	<24	<19	<22	<19	<54	<54	<54	<54	<54	<1	1.7±2.2	6.5±13.8	11.9±19.2
LYS 3-1	<22	<19	<24	<27	<19	<24	<24	<54	<54	<54	<54	<54	<1	<3	3±14.1	<13
LYS 4-1	<19	<22	<22	<27	<27	<27	<27	<54	<54	<54	<54	<54	<1	3.0±4.3	<14	2.4±17.3
LYS 5-1	<27	<22	<22	<27	<27	<27	<27	<54	<54	<54	<54	<54	70.3±21.6	<3	43.2±21.6	23.8±24.6

a. Concentration ± 2 sigma.

b. 1-L subsample from leachate collection.

c. Total moisture cup sample ~0.1-L sample site.

d. Sample not analyzed.

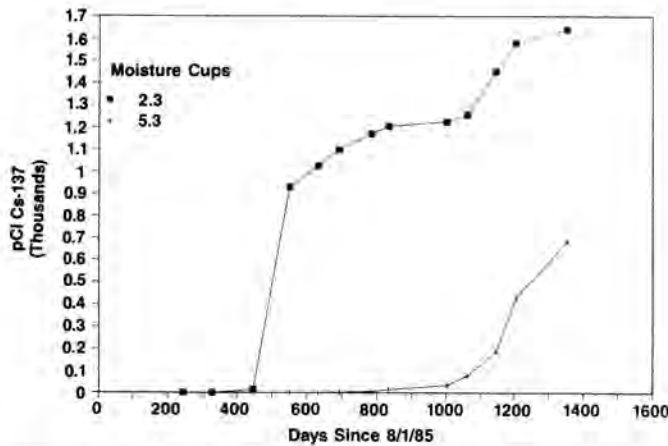


Fig. 8. ANL-E Cumulative Cs-137 Collected in Moisture Cup Number 3.

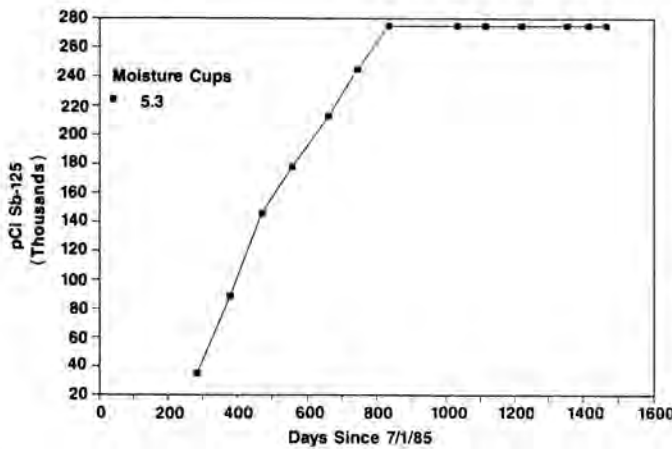


Fig. 9. ORNL Cumulative Sb-125 Collected in Moisture Cup Number 3.

both ORNL 3 and 4 have had Cs-137 occur in leachate collector water.

Although Sb-125 has not been found in water samples from ORNL lysimeter 5 since October 1987, it appeared again in ORNL 5 leachate collector samples beginning with the November 1988 sampling (Table III). It is now calculated that approximately 0.004% of the Sb-125 inventory from the ORNL 5 waste form has been recovered with 0.003% of that occurring during the last year. The release curve for Sb-125 (Fig. 9) appears to resemble the bench leach results for Sr-90 and Cs-137 (11), indicating that the limiting factor on movement of Sb-125 in this lysimeter could be released from the waste form.

On an intrasite comparison (Figs. 6 and 7), the conclu-

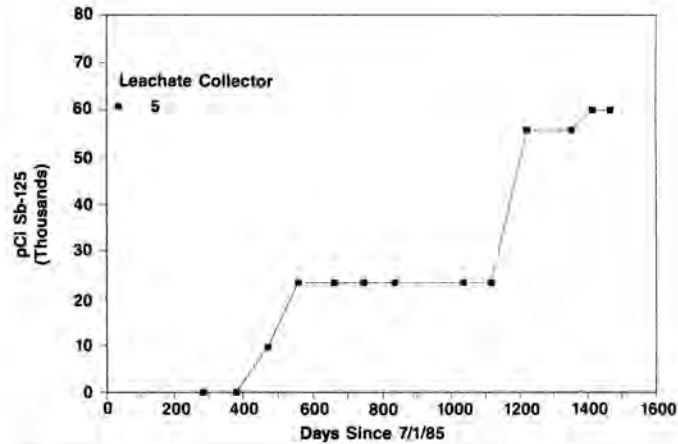


Fig. 10. ORNL Cumulative Sb-125 Collected in Lysimeter Leachate Collectors.

sion that the VES waste forms (lysimeters 3 and 4, Table I) have released quantities of Sr-90 comparable to those lysimeters containing cement waste forms (lysimeters 1 and 2) is still a valid conclusion, because any movement of Sr-90, or lack thereof, does not appear to correlate to either type of waste form. Data from the cups 3 still tend to support the evidence that VES is no better at retaining Sr-90 than cement (Figs. 4 and 5). Based on percent of total inventory release to the cups 3, ANL-1 and -2 (cement) have received 16 to 46 E-6%, respectively, of the Sr-90, while ANL-3 and -4 (VES) have received 16 to 800 E-6%, respectively. Comparable data at ORNL for the cement are 94 to 96 E-6%, while those for VES are 1 to 17 E-6%. These data are initial output and suggest what may be occurring.

The data for Cs-137 found in cups ANL 2-3 and 5-3, and ORNL 5-3 as well as Sb-125 from ORNL-5, continue to be of interest. However, continued lack of occurrence in other lysimeters with the same type waste forms make it difficult to draw conclusions. The continued appearance of Cs-137 in ANL 2-3 and ORNL 5, as well as reappearance in ANL 5-3, would indicate that this occurrence is not an artifact. These data, as well as those for Sr-90, continue to demonstrate the need for long-term field testing of the present waste forms.

Use of Lysimeter Data for Performance Assessment

It is becoming apparent through operational experience and cumulative data provided by the NRC lysimeter array during the past 4 years that lysimeters are a valuable resource for data used in developing site-specific performance assessments. The operational lysimeters are providing continuous data from the near-field (that area

TABLE IV
Relationship Between Performance Assessment Code Parameters and Lysimeter Data.

Code Parameters	Data Collected from Lysimeters
Q = Inventory	Known inventory is introduced by experimental design
P = Annual percolation	Amount of rainfall on lysimeter Amount of evapotranspiration
S = Fraction of saturation	Soil moisture content
V_v = Water velocity	Mass or volume of effluent water per unit time
R = Retardation factor	Mass or volume of effluent solute per unit time relative to V_v
d_s = Soil bulk density	From experimental design of lysimeter
P_s = Effective soil porosity	Can be estimated for saturated conditions from mass of effluent water, volume of soil, soil bulk density
I_r = Inventory released	Radionuclide concentrations in soil pore water and in effluent
V_w = Trench volume	From experimental design of lysimeter
C_w = Radionuclide concentration	Radionuclide concentration in effluent
M_i = molality	Effluent concentrations
MIN = Minerals dissolved or precipitated	From mineralogical characterization of soil at end of experiment

TABLE V
Performance Assessment Code Parameters Derived from ANL and ORNL Data.

Code Parameters	ANL					ORNL				
	1	2	3	4	5	1	2	3	4	5
Annual percolation (P) M/y	0.297	0.346	0.520	0.422	0.817	0.969	0.977	0.983	0.995	1.148
Vertical water velocity (V_V) M/y	1.14	1.33	2.00	1.62	3.89	5.21	5.25	5.28	5.35	5.74
Inventory (Q) pCi Sr-90	18.2E9	3.3E9	27.4E9	4.5E9	18.2E9	18.2E9	3.3E9	27.4E9	4.5E9	3.3E9
Fraction of saturation (S) (ave. of past 3 y)	56.4	56.5	56.4	56.4	50	37.2	37.2	37.2	37.2	50
Soil bulk density G/cm ³ (ds)	1.42	1.39	1.42	1.48	1.55	1.30	1.34	1.30	1.30	1.60
Effective soil porosity (P_S)	0.46	0.48	0.46	0.44	0.42	0.51	0.49	0.51	0.51	0.42
Inventory release (Ir) % Sr-90	27E-6	49E-6	29E-6	1E-6	1500E-6	140E-6	279E-6	60E-6	220E-6	2160E-6
Radionuclide concentration (Cw) Ave pCi Sr-90/L Leachate	6.6	1.9	5.8	0.1	128.2	10.6	3.9	7.1	4.1	25.8

comprised of waste form and surrounding soil) in a number of areas that directly relate to waste form stability.

The relationship between input parameters for codes and data derived from lysimeter operation is compared in Table IV. These parameters have been calculated using data collected during the 48-month operation of the NRC lysimeters at ANL-E and ORNL (Table V). The data could be used in codes to predict the stability of the waste forms for a 300-year period of time.

CONCLUSIONS

In the lysimeter experiments, Sr-90 is still the most prevalent nuclide in collected liquid samples. It appears that waste-form performance is similar with respect to release of Sr-90 (except for a very high release from ANL 3). It is also apparent that Sr-90 is able to move more freely through the SRL soil at ORNL. During the past 24 months, Sr-90 continued to be found in leachate water in the control lysimeters at both sites. It appears that the limiting step in receiving Sr-90 in the leachate is not release of the nuclide from the waste forms (because Sr-90 is found in cups 3), but rather it is the soil characteristics (including soil and quantity of soil water) that limit movement. This conclusion is supported by data from recent lysimeter work at the SRL, where Sr-90 migrated through the soil column,(15) and Pacific Northwest Laboratory (PNL) at Hanford, where it did not.(16)

Data on waste-form performance presented in this paper continue to suggest that VES is comparable to cement in its ability to retain Sr-90. These data still differ from those obtained at SRL, which show that cement minimizes the release of Sr-90.(15) Both data reported herein and data reported by SRL and Hanford agree that Cs-137 is more readily released from cement than VES.

It is fortunate that the NRC has such a data base at a time when the concept of site performance assessment of buried radioactive waste is being developed for implementation. It appears that the data generated from properly designed lysimeter arrays could be used as a tool for performance assessment of solidified radioactive waste, thus assisting NRC in their efforts to develop methods to verify the 300-year stability of waste forms. The preliminary conclusion that Sr-90 movement is soil dependent would indicate that use of laboratory leach test results in performance assessment would not represent actual conditions. In the context of waste-form testing, that is perhaps the most interesting and significant conclusion of this work. These results can serve the NRC as guidelines for new waste-form testing protocol development.

The data provided by these lysimeters experiments has been shown to be useful as input parameters for performance assessments codes. The utility of this reliable source of data will be demonstrated through continued operation

of the lysimeters for a minimum period of time required (20 years) for the validation of codes used to provide predictions on waste-form stability for 300 years and beyond.

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