RESULTS OF FIELD TESTING OF WASTE FORMS USING LYSIMETERS*

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ABSTRACT

The Field Lysimeter Investigation: Low-Level Waste Data Base Development Program 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 (TMI) 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 presents field testing results of those waste forms through FY-1990. The results from the lysimeter experiments are presented and the use of lysimeter data to determine input parameters used in performance assessment is discussed.

INTRODUCTION

Concerns over the practices associated with the disposal of low level radioactive wastes has resulted in a very real need to obtain accurate data on the long-term field performance of these wastes. Regulations have been enacted by the U.S. Nuclear Regulatory Commission (NRC) that link low-level radioactive waste acceptance criteria to the long-term satisfactory performance of the waste. Under 10 CFR Part 61, "Licensing Requirements for Land Disposal of Radioactive Waste," commercially generated low level radioactive waste is classified as Class A, B, C or greater than Class C (1). Wastes classified as either Class B or Class C must be stabilized; the stability must be ensured for a minimum of 300 years.

To verify the 300-year stability of waste forms, the NRC originally specified the use of short-term standardized tests with the hope that such tests in principle could provide information relevant to near-surface disposal performance objectives. Those tests, which were published in the NRC Branch "Technical Position (TP) on Waste Form," are currently undergoing critical review to determine their applicability to the 300-year stability requirements (2).

A central requirement in LLW disposal site design is the need for a detailed understanding of the waste form as the radionuclide source of the site and the effect of that source on site performance. A major aid in any site design is the site Performance Assessment (PA). It is intended that the PA will predict whether a proposed disposal site will meet the performance objectives. Correct assumptions regarding the performance of the buried waste form have a direct bearing on the outcome of the PA analyses. Conservative assumptions regarding rates of radionuclide release will result in over-design of containment, while under estimating the release of especially mobile constituents will over-predict containment performance.

A useful experimental device that will provide hard data for determining waste form stability is the lysimeter. Lysimeters are ideal systems for obtaining actual field test data because, when properly designed and operated, they can be used to isolate soil/waste systems under actual environmental conditions. Such conditions cannot be duplicated by standard laboratory testing. Lysimeters lend themselves to instrumentation for the acquisition of environmentally related data and the collection and storage of migrating precipitation.

The Field Lysimeter Investigation: Low-Level Waste Data Base Development Program, funded by the NRC, is obtaining information on performance of radioactive waste in a disposal environment. The requirements of 10 CFR 61 are also being investigated under this program. Waste forms are being field-tested to (a) develop a LLW data base, and (b) obtain information on survivability of waste forms in a disposal environment.

The purpose of this paper is to present the experimental results of two instrumented, operational lysimeter arrays over 5 years of operation. This study was developed to perform field testing of waste forms composed of solidified

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ion-exchange resin materials from EPICOR-II* prefilters used in the cleanup of the TMI Nuclear Power Station Unit 2 (3,4). Waste used in the study is significant because it has a high loading of radionuclides and is comprised of ion-exchange media that is used by the nuclear industry.

METHODS AND MATERIALS

Wastes used in the experiment were taken from EPICOR-II prefilters used to decontaminate water at TMI-2 and include a mixture of synthetic organic ion-exchange resins from prefilter PF-7 and a mixture of organic-exchange resins and an inorganic zeolite from prefilter PF-24. 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). 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. The PF-7 waste contained 89% of the radionuclides as Cs-137 while PF-24 contained 94% Cs-137. The PF-7 waste also contained 5% Sr-90 and PF-24 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 at the termination of the study. Each lysimeter is a 0.91- x 3.12-m right-circular cylinder divided into an upper compartment that 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; a fifth, used as a control, is filled with inert silica oxide sand. The lysimeters at ANL-E contain soil indigenous to the site,

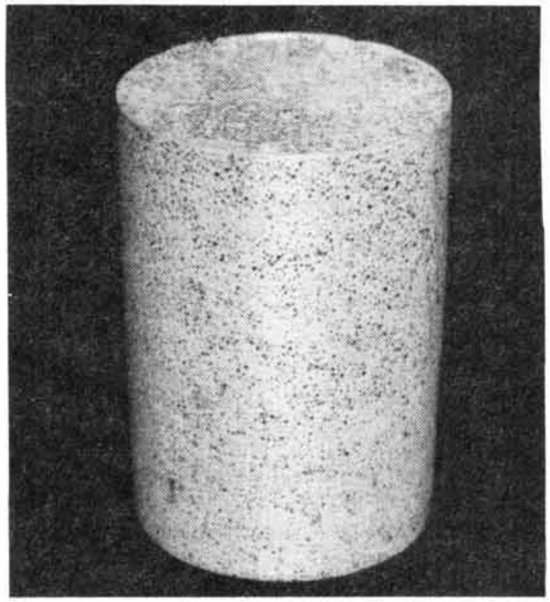

Fig. 1. Example of an EPICOR-II waste form.

TABLE I

Lysimeter Waste-Form Composition

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

* Mention of specific products and/or manufacturers in this document implies neither endorsement of preference nor disapproval by the U.S. Government, any of its agencies, or EG&G Idaho, Inc., of the use of a specific product for any purpose.

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 on-site data acquisition and storage system (DAS) that also collects data from a field meteorological station located at each site. Each month data stored on a cassette tape are retrieved from the DAS and translated into an IBM-PC-compatible disk file. On a quarterly basis, water is drawn from the porous cup soil-water samplers and the lysimeter leachate collection compartments. These water samples are analyzed for chemical species and beta- and gamma-producing nuclides. Details on waste form formulations are given in Ref. 5. Lysimeter design, installation, instrumentation, operation, and data acquisition are provided in Ref. 6.

Monitoring of moisture cups began with collecting liquid samples in September 1985 (2 to 3 months from the time of placement). Each month, data stored on a cassette tape are retrieved from the DAS and translated into a IBM-PC-compatible disk file. 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. 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. Results of testing are presented in Refs. 7 through 15 as well as this paper.

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 life the experiment (7-15). Figure 3 shows the cumulative precipitation for each site since the initiation of field work. Examples of the lysimeter soil temperature data recorded over 1-year periods and examples of data from the moisture probes at ANL-E and ORNL can be found in Refs. 7 through 15. Data recorded in FY-1990 indicate that the lysimeter soil columns at both sites have remained moist during the last reporting period. The average soil moisture of ANL-E lysimeter soils was 56.1% of the soil moisture holding capacity; for ORNL, this value was 39.2%.

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 cm^2). The cumulative volume of precipitation received by each ANL-E lysimeter was 2769 L; at ORNL, this value was 4139 L. It appears that the throughput is

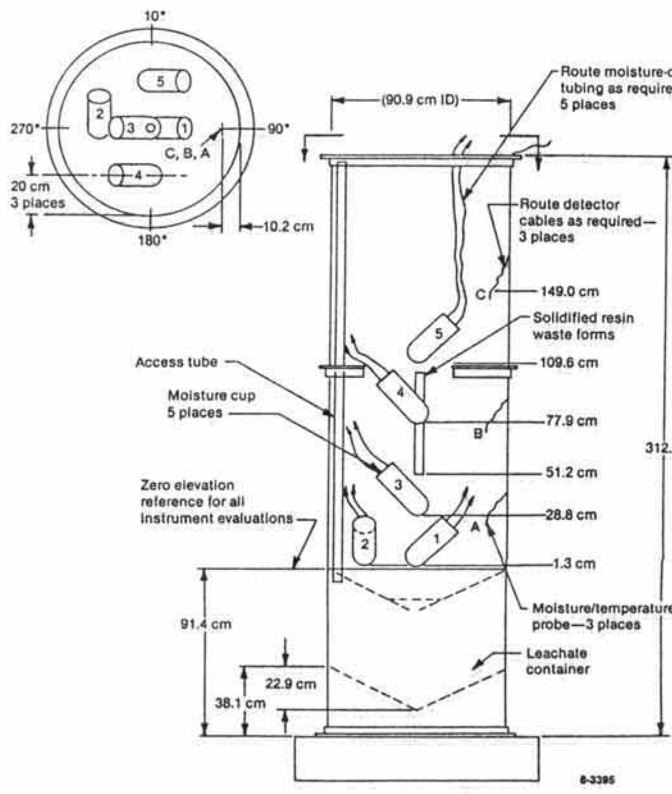

Fig. 2. Lysimeter vessel component locations.

dependent on site conditions (period of time soil surface was frozen, amount of precipitation received as snow, etc.) and lysimeter fill material. The total volumes of precipitation that have moved through the lysimeters represent an *average* 1.71 pore volumes for the ANL-E soil lysimeters and 4.94 pore volumes for soil lysimeters at ORNL, while the control lysimeters at ANL-E and ORNL were 4.70 and 7.29 pore volumes, respectively.

Radionuclide Analysis

Four samplings from moisture cups and leachate collectors were taken at each site during the 12-month period. The cumulative amounts of nuclides as determined in water samples obtained from number 3 moisture cups and leachate collectors for all sampling periods are displayed graphically in Figs. 4 through 12.

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. However, consistent significant occurrences have been observed in all the number 3 cups at ANL-E and ORNL, except cup 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 - 7). There are standout amounts of Sr-90 retrieved from moisture cup samples at both sites. Those include 452,840 pCi from cup 3-3 at ANL-E (Fig. 4) and 37,286 pCi *from cup* 1-3 at ORNL (Fig. 5).

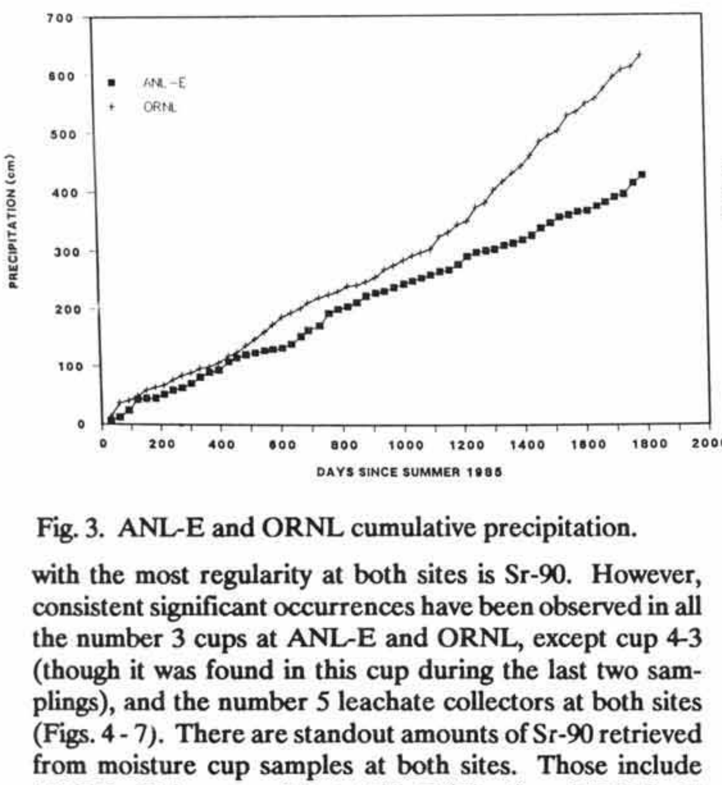

Fig. 3. ANL-E and ORNL cumulative precipitation.

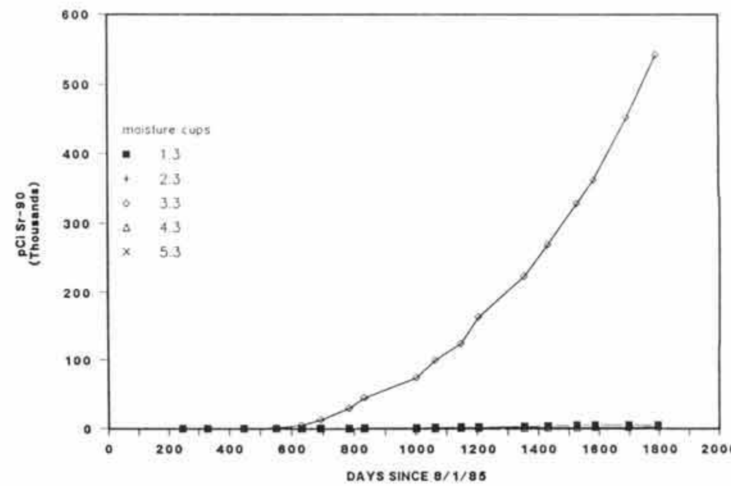

Fig. 4. ANL-E cumulative Sr-90 collected in moisture cup number 3.

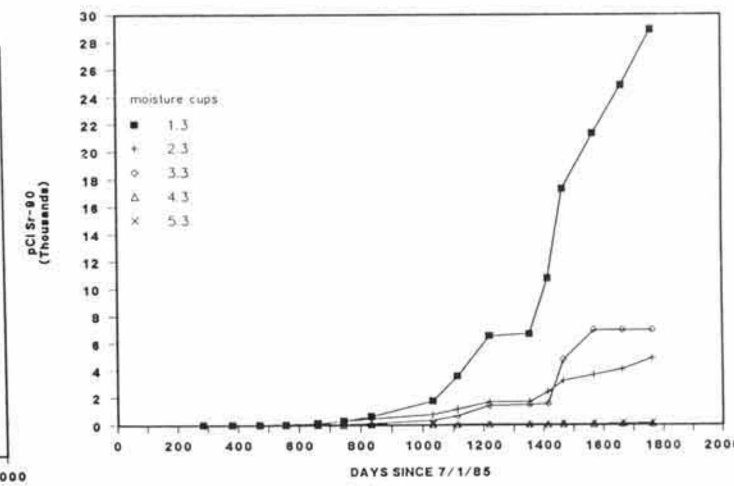

Fig. 5. ORNL cumulative Sr-90 collected in moisture cup number 3.

During the past 12 months, only the leachate collector water from the control lysimeters at each site contained significant amounts of Sr-90 (Figs. 6 and 7). This is comparable to the previous years findings and is in sharp contrast to the number 3 cups data, which continue to demonstrate that substantial amounts of Sr-90 are still being released from the waste forms (7-15). The percent of total Sr-90 being measured in the leachate collectors and number 3 cups is inconsistent (11). This could indicate differences in waste form performance at the two sites. However, there is still a comparable percentage of total Sr-90 in the leachate water of the control lysimeter for the two sites (Figs. 6 and 7). These data are interesting, because the waste forms at each site have been experiencing similar exposure to moisture and temperature (11).

Gamma-producing nuclides have occurred with regularity at ANL-E and are again present at ORNL. ANL-E cup 2-3, below a cement waste form containing large amounts of Cs-137, has received Cs-137, with a significant increase in quantities of this radionuclide appearing this year after initially peaking in the February 1987 sample

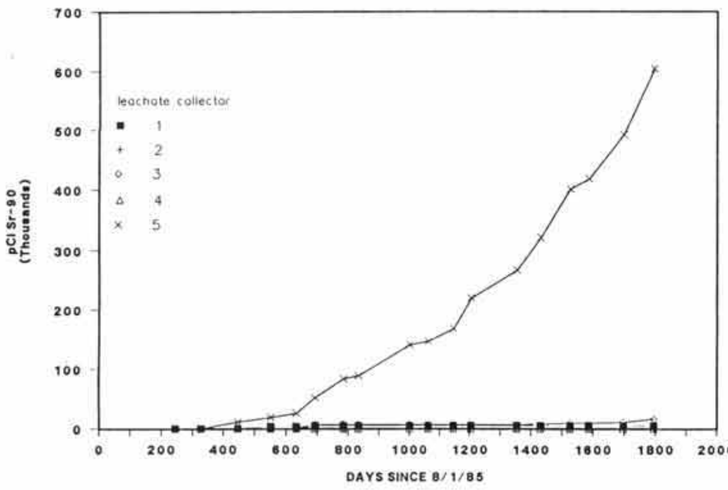

Fig. 6. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

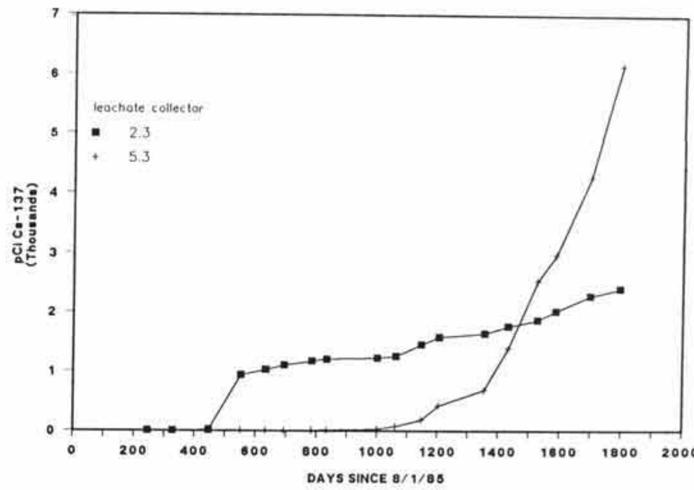

Fig. 8. ANL-E cumulative Cs-137 collected in moisture cup number 3.

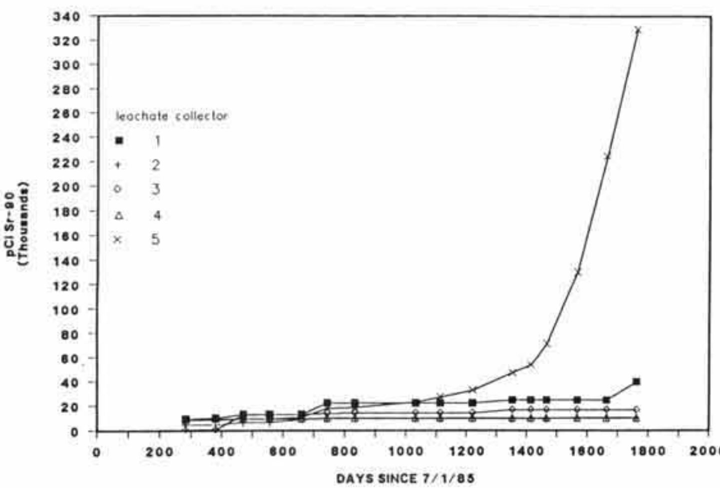

Fig. 7. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

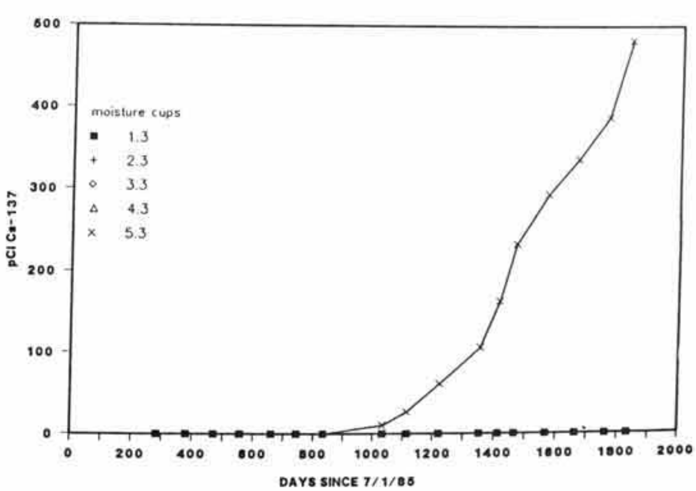

Fig. 9. ORNL cumulative Cs-137 collected in moisture cup number 3.

(Fig. 8). Since June of 1987, Cs-137 has been appearing in ANL-E cup 5-3. The quantity of this nuclide increased in each of the sampling periods during the last year with an abrupt increase during the last sampling period (Fig. 8) (6). There continues to be no sustained occurrence of Cs-137 in any of the ANL-E leachate collector water. Measurable amounts of Cs-137 began to occur in ORNL cup 5-3 during the May 1988 sampling and have continued in subsequent samplings for a total of 480 pCi (Fig. 9). Also Cs-137 has been detected consistently in water from the ORNL lysimeter leachate collectors (Fig. 10). Breakthrough of Cs-137 into the ORNL-5 leachate collector occurred in November 1988 (Fig. 10), and thus far a total of 133,198 pCi have passed through the lysimeter.

The release curve for Sb-125 (Fig. 10) appears to resemble the bench leach results for Sr-90 and Cs-137 indicating that the limiting factor on movement of Sb-125 in this lysimeter could be released from the waste form (9). No

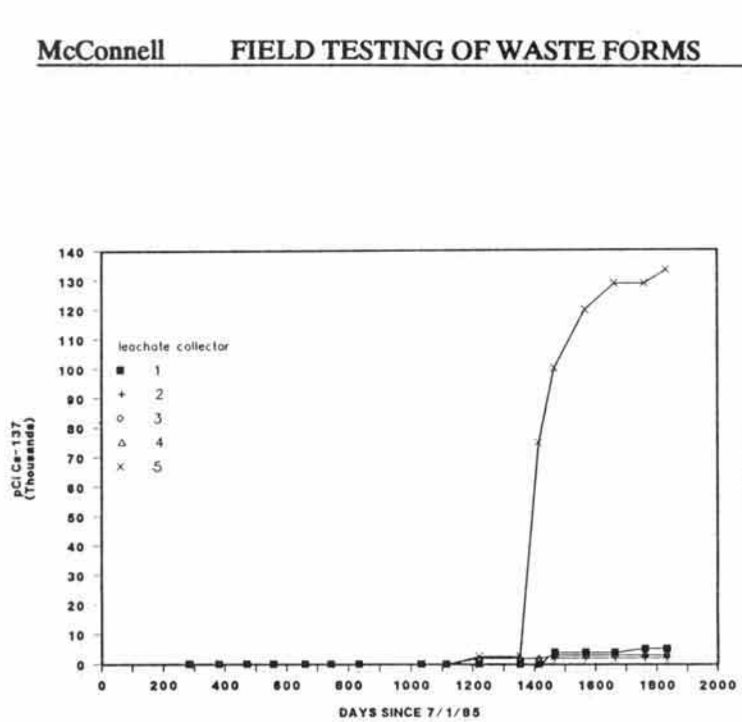

Fig. 10. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

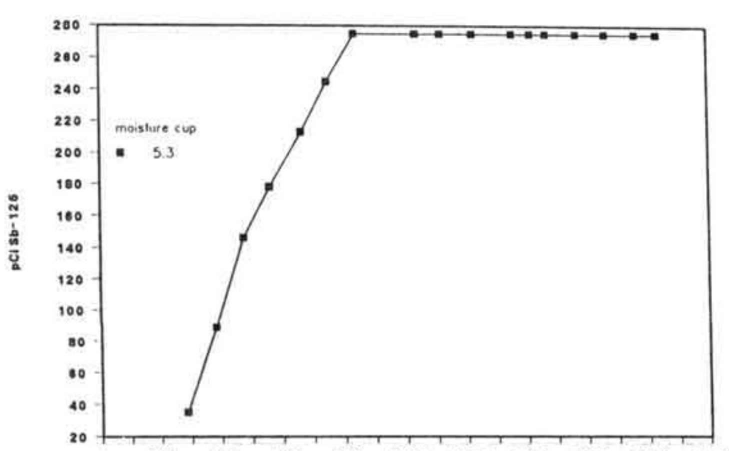

Fig. 11. ORNL cumulative SB-125 collected in moisture cup number 3.

Sb-125 has been found in water samples from ORNL lysimeter 5 since November 1988 (Fig. 12).

On an intrasite comparison (Table II), the conclusion that the VES waste forms (lysimeters 3 and 4,) have released quantities of Sr-90 comparable to those lysimeters containing cement waste forms (lysimeters 1 and 2) is valid, because any movement of Sr-90, or lack thereof, does not appear to correlate to either type of waste form.

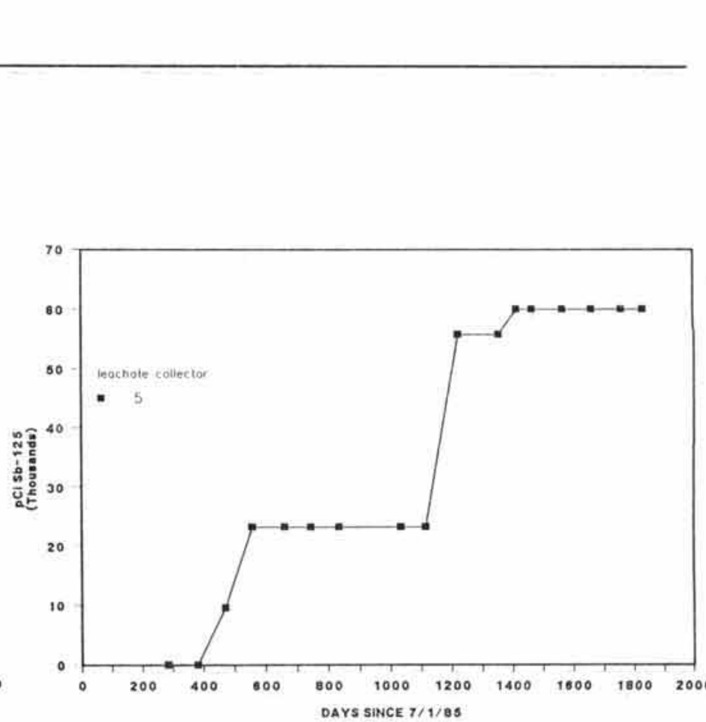

Fig. 12. ORNL cumulative SB-125 collected in lysimeter leachate collector.

At ANL-E, recovery of SR-90 in cup 3 and the leachate collectors continues to indicate a uniformity of waste form performance. More Sr-90 is found in ANL-E cup 3-3 (VES waste form) than in the other ANL-E cups but the effect appears to be moderated by the distance traveled in soil from the waste form to the leachate collector. Movement of the nuclide into the leachate of collector ANL-5 is much greater than that of the other lysimeters and thus provides continued evidence of the moderating effect of soil. Greater quantities of Sr-90 appear to be moving through the ORNL lysimeter than the ANL-E lysimeter. Once again, there appears to be no correlation between the type of waste form and the amount of nuclide recovered in the leachate collector. About 0.1% of the Sr-90 contained in ORNL-5 has now been recovered in leachate from the lysimeter. Recovery of Sr-90 in the ORNL cups has been greater in those lysimeters containing the cement waste forms but any conclusions drawn based on those data must be tempered with data from ANL-E cups 3-5, which shows greater recovery of Sr-90 from a VES waste form.

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 5 years that lysimeters are a valuable resource for data applicable to calculating parameters used in developing site-specific performance assessments. The operational lysimeters are providing continuous data from the near-field (that area comprised of waste form and surrounding soil) in a number of areas that directly relate to waste form stability.

TABLE II

Comparison of Total Sr-90 and Cs-137 Inventory per Lysimeter to Total Amounts in Lysimeter Water

		Percent Total Inventory Sr-90 x 10^6				Percent Total Inventory Cs-137 x 10^6			
		Moisture Cups		Leachate Water		Moisture Cups		Leachate Water	
Lysimeter Number	Solidifying Agent	ANL	ORNL	ANL	ORNL	ANL	ORNL	ANL	ORNL
1	Cement	29	205	27	235	--	--	--	2.0
2	Cement	70	174	49	313	0.2	--	--	0.1
3	VES	1981	5	60	64	--	--	--	0.7
4	VES	47	2	6	219	--	--	--	0.1
5	Cement	32	6	3322	10000	10.0	0.3	--	9.0

The input parameters used in codes have been calculated using data collected from the operation of the NRC lysimeters at ANL-E and ORNL (10). The most important information on waste form performance is derived from the cumulative release curves and is represented by the maximum cumulative release value at the time release ends; the point where the curve flattens as in Figs. 11 and 12. 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 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-E cup 3). It is also apparent that Sr-90 is able to move more freely through the SRL soil at ORNL. It seems 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 number 3 cups), but rather it is the soil characteristics (including soil and quantity of soil water) that limit movement.

Data on waste-form performance presented in this paper suggest that VES is comparable to cement in its ability to retain Sr-90. These data differ from those obtained at SRL, which show that cement minimizes the release of Sr-90 (7-15). Both data reported herein and by SRL agree that Cs-137 is more readily released from cement than VES.

Data provided by these lysimeter experiments have been shown to be useful in computing many parameters used as input to PA codes. The utility of this reliable source of data will be demonstrated through continued operation of the lysimeters. NRC planning recommends that these lysimeters be augmented by experiments containing solidified decontamination wastes from commercial plants.

The U.S. Department of Energy (DOE) at the Idaho National Engineering Laboratory is investigating use of lysimeter systems based on the design presented here to provide data for calculating parameters to be used in PA of LLW disposal sites and for verification of the codes used in those performance assessments in accordance with DOE Orders governing the disposal of LLW. Consideration is also being given to using these lysimeters as waste form performance monitors at DOE disposal sites.

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