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 (USNRC) is obtaining information on 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 field 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 1987.

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 which were solidified in matrices of cement and DOW polymer.

METHODS AND MATERIALS

Wastes used in the experiment include a mixture of synthetic organic ion exchange resins (herein referred to as Type 1 waste), and a mixture of organic exchange resins and an inorganic zeolite (herein referred to as Type 2 waste). Solidification agents employed to produce the 4.8- by 7.6-cm cylindrical waste forms (Fig. 1) used in the study were Portland Type I-II cement and Dow vinyl ester-styrene (VES) (4). 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 25% of the radionuclides as ^{90}Sr while Type 2 contained about 1% ^{90}Sr. There were also significant amounts of ^{137}Cs and ^{134}Cs and trace amounts of ^{60}Co and ^{125}Sb found in those wastes. There are 10 lysimeters, 5 at ORNL and 5 at ANL-E. Lysimeters used in this study were designed to be self-contained units which will be disposed of at the termination of the 20-year study. Each is a 0.91- by 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 porous cup soil-water samplers and soil moisture/temperature probes. The probes are connected to an on-site data acquisition system (DAS) which 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. At least quarterly water is drawn from the porous 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 formulation, lysimeter design, installation, instrumentation, operation, and data acquisition are provided in Refs. 4, 5, 6, 7, 8, and 9.

Monitoring of moisture cups began with collecting liquid samples in September 1985 (two to three months from the time of placement) and has continued once every three 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 data acquisition system. The DAS systems at both sites continued to function as expected during this reporting period.

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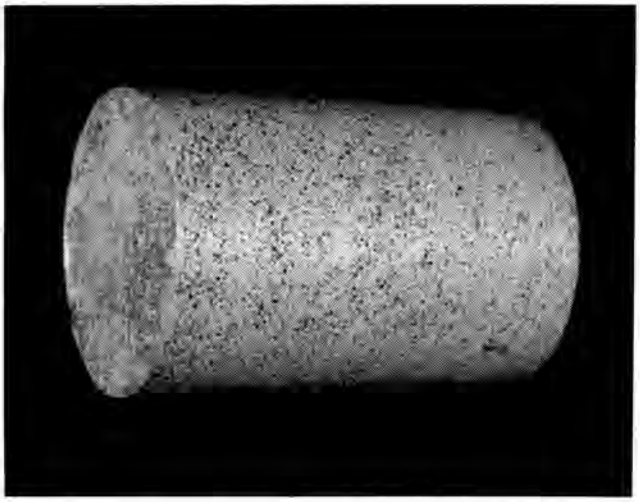

Fig. 1. Example of an EPICOR-II Waste Form.

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 86.4 cm at ANL-E and 120.4 cm at ORNL. ANL-E was very close to their normal annual rainfall of 85.2 cm (10) while for a second year, ORNL was below their normal annual rainfall of 138.8 cm (11). Figure 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 the first of November 1986 until near mid-April 1987. ORNL experienced very few days where there was an air temperature as low as 0°C (typical data shown in Refs. 8 and 9).

Examples of the lysimeter soil temperature data recorded over a one-year period at ANL-E and ORNL can be found in Refs. 8 and 9. At no time during the FY 1987 reporting period was a freezing temperature recorded at any depth within a lysimeter.

Examples of data from the moisture probes at both ANL-E and ORNL can be found in Refs. 8 and 9. Data recorded in FY 1987 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 lysimeter over time (as determined by averaging the outputs of the three probes in each lysimeter) showed that moisture data for these lysimeters at each site were relatively similar.

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 which has been received by the exposed lysimeter surfaces (6489.5 cm^2). The cumulative volume of precipitation received by

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
5 ANL-E	Silica Oxide	Cement with Type 1 waste
5 ORNL	Silica Oxide	Cement with Type 2 waste

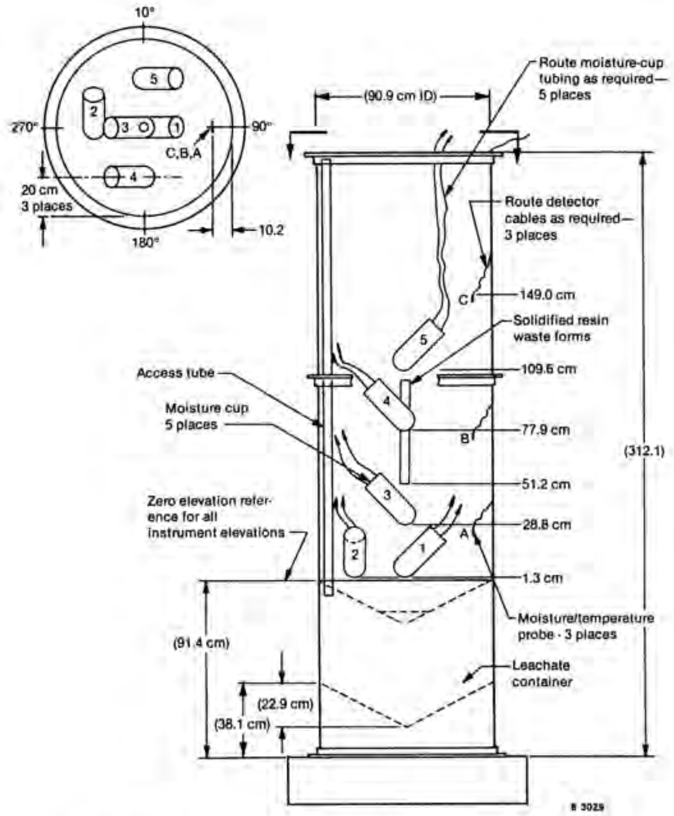

Fig. 2. Lysimeter Vessel Component Locations.

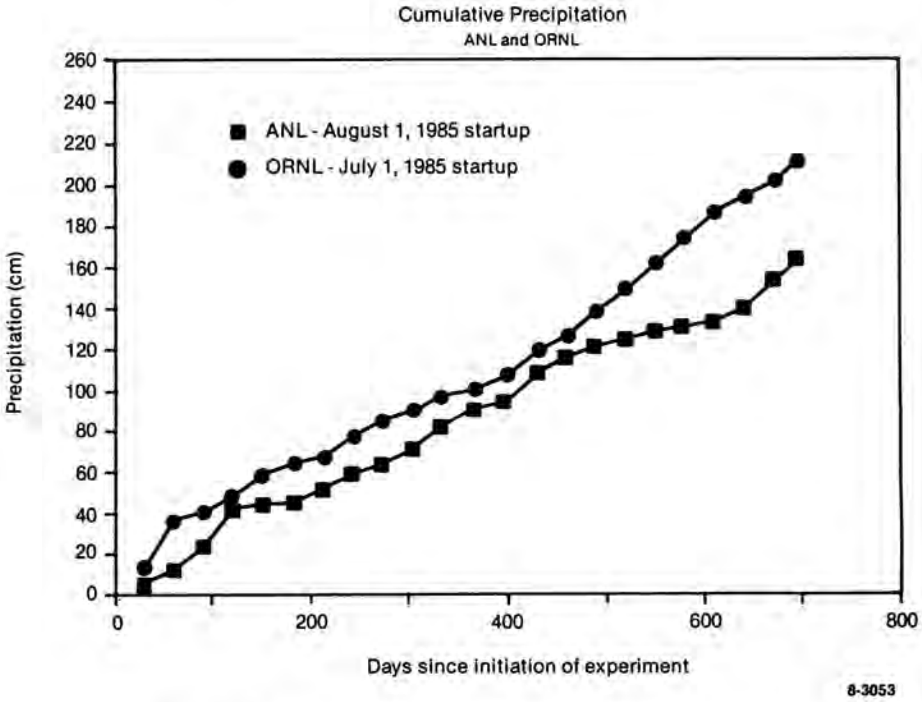

Fig. 3. ANL-E and ORNL Cumulative Precipitation.

each ANL-E lysimeter was 1085.8 L; at ORNL, this value was 1424.1 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 441.5 ± 81.2 L, or 40.6% of total precipitation, passed through the soil lysimeters; while for the control lysimeter, this value was 945 L, or 87.0% of available precipitation. For ORNL, the values were 1135 ± 22.5 L (79.7%) for the soil-filled lysimeters and 1342 L (94.2%) 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).

ANL-E lysimeters 1 and 2 have experienced water ponding during periods of heavy rainfall. To prevent loss of precipitation, some water was drained from the surface of these lysimeters and later replaced. It was determined that the top several inches of soil had been overcompacted during installation. Fresh soil was used to replace the compacted soil which reduced the ponding problem significantly.

The total volumes of precipitation that have moved through the lysimeters represent an average 0.62 pore volumes for the ANL-E soil lysimeters and 1.60 pore volumes for soil lysimeters at ORNL; while the control lysimeters at ANL-E and ORNL were 1.61 and 2.28 pore volumes, respectively. Then, since the beginning of the study, 62% of the water held in pore spaces of the soil column in the ANL-E lysimeters has been replaced. In the ORNL lysimeters, 160% of the water held in pore spaces of the soil column has been replaced. The ANL-E and ORNL control lysimeters had 161% and 228% of the water held in pore spaces of the sand columns respectively replaced.

Radionuclide Analysis

Water samples were collected on a quarterly basis from leachate collectors and moisture cups of each of the lysimeters during the last 12-month period. At each sampling, water from the leachate collectors (1 L of the collected sample) and cups (0.1 L of the collected sample) closest to the waste forms were analyzed for gamma-producing nuclides and the beta-producing nuclide ^{90}Sr. *Moisture* cup 3 samples are taken first. The analysis protocol, however, triggers the analysis of water from additional cups in a sequential manner if nuclides are found in a cup 3 sample. For example, when nuclides were found in cup 3 of a lysimeter, water from cup 1 (directly below cup 3), then cup 4, followed by cup 2 were analyzed (see Fig. 2 for cup placement).

Tabular data for the last four samplings are found in Table II and III. Nuclides which are appearing consistently at the sites include ^{90}Sr and ^{137}Cs at ANL-E and ^{90}Sr and ^{125}Sb at ORNL. Strontium-90 has occasionally occurred in measurable concentrations in all cup 3 and leachate samples. Consistent occurrence of ^{90}Sr, however, has only been in cups ANL 1-3, ANL 3-3, ANL 5-3, leachate collector ANL-5, ORNL 1-3, ORNL 2-3, and ORNL 3-3 . Strontium 90 occurrences in ORNL cup 1 and leachate collector samples have been sporadic. A possible conclusion from these data is that movement of ^{90}Sr is being controlled by some site-specific phenomenon, such as precipitation or soil type.

Further definition of ^{90}Sr behavior can be seen by examining cumulative totals for ^{90}Sr over time (Figs. 4, 5, 6, and 7). The release of ^{90}Sr from the control lysimeters at both sites was observed in most samples after 245 days at ANL-E and 284 days at ORNL. The movement of ^{90}Sr through the soil lysimeters at ANL-E was very different from the movement through the ANL-E control lysimeter (Fig. 6), while those at ORNL have been very similar to the ORNL control lysimeter (Fig. 7). An above average amount of ^{90}Sr has been detected on the ANL-E 3-3 moisture cup. At both sites similar quantities of the nuclide are passing through the soil columns into the leachate collectors except ANL-5, which has consistently accumulated above average amounts of ^{90}Sr. These data lead to the conclusion that the soil must be a determining factor in the occurrence of ^{90}Sr in leachate water. What is not known is whether the soil environment contributes to both the release of ^{90}Sr from the waste form as well as its subsequent movement.

The ^{90}Sr has moved at least the 22 cm distance from the waste form to moisture cup 3 in both ANL-E and ORNL lysimeters. However, Strontium 90 has not been found in other ANL-E soil moisture cups except 5-1. This is not the case at ORNL where detectable amounts have been found sporadically in all cup 1 samples (Table II and III). It has been noted that the ANL-E soil lysimeters have had a much reduced movement of water through them as compared with similar lysimeters at ORNL (0.62 versus 1.60 pore volumes). At ORNL, ^{90}Sr was seen in at least one cup 3 after 0.24 pore volumes and in all cup 3 samples by 0.70 pore volumes. Increasing quantities of ^{90}Sr were found in all leachate water by 0.70 pore volume. So while it is not possible to specifically determine why ANL-E soils have restricted the movement of ^{90}Sr in most lysimeters, we believe that both physical and chemical mechanisms can be contributing factors (physical in the sense that the soil impedes the movement of water).

Each site has had gamma-producing nuclides occurring with regularity in some lysimeter water samples. At ANL-E, ^{137}Cs has been found in ANL 2 cup 3 samples for the past four samplings (Table II and Fig. 8). The concentration of

TABLE II

Results of BETA and GAMMA Analysis of ANL-E Soil Moisture and Leachate Samples--Year2--1986-87

	Concentration (pCi/L)[a]											
	Cobalt-60				Cesium-137				Strontium-90			
Sample Identification	October 86	February 87	April 87	June 87	October 86	February 87	April 87	June 87	October 86	February 87	April 87	June 87
Lys 1[b]	<5	<5	<5	<1	<5	<5	3 ± 2	<1	3.0 ± 18.6	51.3 ± 27.0	<1	26.2 ± 21.3
Lys 2	<5	<5	<5	<1	<5	<5	<5	<1	8.1 ± 18.1	20.3 ± 25.4	25.7 ± 21.1	22.1 ± 20.5
Lys 3	<5	<5	<5	<1	<5	18 ± 1	<5	<1	7.0 ± 17.0	0.4 ± 16.6	<1	75.6 ± 32.4
Lys 4	<5	<5	<5	<1	<5	<5	<5	<1	11.9 ± 16.2	<27	<1	8.6 ± 19.4
Lys 5	<5	<5	<5	<1	<5	<5	<5	<1	40.5 ± 21.6	82 ± 2	85.8 ± 0.5	148.5 ± 40.5
Lys 1-3[c]	<5	<5	<5	<5	<5	<5	<5	<5	118.8 ± 37.8	13.5 ± 51.3	535 ± 18	1341 ± 15
Lys 2-3	<5	<5	<5	<5	109 ± 19	9183 ± 116	969 ± 28	723 ± 27	11.9 ± 23.6	22.1 ± 22.1	2970 ± 54	1370 ± 16
Lys 3-3	<5	<5	<5	<5	<5	<5	<5	<5	1198 ± 135	13500 ± 270	36100 ± 471	79200 ± 672
Lys 4-3	<5	<5	<5	<5	<5	<5	<5	<5	NA	16.2 ± 24.8	52 ± 7	15.7 ± 21.9
Lys 5-3	<5	<5	<5	<5	<5	<5	<5	21 ± 11	647 ± 10	950 ± 54	198 ± 34	2293 ± 35
Lys 2-1		FL[d]	<5	<5		FL	<5	<5		FL	<5	
Lys 3-1	<5	NA[e]			<5	NA			NA			NA
Lys 5-1	<5	<5	<5	<5	<5	<5	<5	<5	99 ± 2	190 ± 2	198 ± 11	NA
Lys 2-4			<5				<5				<1	
Lys 5-4	<5	NS[f]	<5		<5	NS	<5		6 ± 5	NA	<1	NA
Lys 2-2				<5			<5					NA
Lys 5-2	NA			<5	<5			<5	<5		24 ± 3	NA

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a. Concentration ±2 sigma.

b. Leachate sample from 1 L sample size.

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

d. Sample not taken due to frozen lines.

e. Sample not analyzed.

f. No sample taken.

TABLE III

Results of BETA and GAMMA Analysis of ORNL Soil Moisture and Leachate Samples--Year 2--1986-87

	Concentration (pCi/L)[a]															
	Cobalt-60				Cesium-137				Antimony-125				Strontium-90			
Sample Identification	October 86	January 87	April 87	July 87	October 86	January 87	April 87	July 87	October 86	January 87	April 87	July 87	October 86	January 87	April 87	July 87
Lys 1[b]	<5	<5	<5	<11	<8	<5	<5	<5					29.7 ± 5.4	1.3 ± 1.6	2.5 ± 4.3	89.2 ± 13.5
Lys 2	<8	<8	<0.5	<8	<8	<5	<0.5	<8					20.8 ± 5.1	1.3 ± 1.6	8.4 ± 5.1	6.49 ± 4.05
Lys 3	<8	<8	<8	<11	<5	<5	<8	<11					108 ± 10.8	2.2 ± 1.8	3.0 ± 4.8	7.84 ± 3.78
Lys 4	<5	<8	<5	<11	<5	<5	<5	<8					1.9 ± 2.5	1.4 ± 1.5	3.2 ± 4.0	4.32 ± 3.78
Lys 5	<5	<8	<5	<11	<5	<5	<5	<8	64.9 ± 24.3	54.0 ± 18.9	ND[d]	ND	3.5 ± 2.7	0.7 ± 1.4	6.5 ± 4.9	45.9 ± 8.11
Lys 1-3[c]	<54	<22	<22	<27	<27	<19	<19	<54					64.9 ± 10.8	297 ± 27	946 ± 54	1540 ± 108
Lys 2-3	<27	<24	<22	<54	<27	<22	<19	<54					64.9 ± 27.0	270 ± 27	919 ± 81	1459 ± 108
Lys 3-3	<54	<24	<27	<54	<54	<19	<19	<27					24.9 ± 7.8	45.9 ± 13.5	165 ± 27	405 ± 54.0
Lys 4-3	<54	<27	<22	<54	<54	<24	<19	<54					6.2 ± 3.2	<5	18.9 ± 11.3	10.0 ± 14.3
Lys 5-3	<54	<24	<16	<27	<27	<27	<16	<54	568 ± 108	324 ± 81	351 ± 108	ND	16.5 ± 10.3	5.1 ± 6.5	3.8 ± 7.6	11.3 ± 18.6
Lys 1-1	<24	<27	<3	<81	<22	<19	<3	<81					16.2 ± 10.8	1.9 ± 6.2	<5.4	9.19 ± 12.4
Lys 2-1	<81	<27	<27	<54	<54	<22	<27	<54					0.5 ± 5.7	4.3 ± 7.3	67.6 ± 27.0	8.65 ± 11.6
Lys 3-1	<19	<22	<14	<54	<19	<22	<16	<54					5.1 ± 6.8	16.2 ± 14.9	2.2 ± 7.3	4.86 ± 10.8
Lys 4-1	<19	<22	<19	<81	<19	<22	<19	<81					3.0 ± 6.2	2.4 ± 5.9	4.0 ± 7.8	26.7 ± 23.8
Lys 5-1	<54	<27	<22	<54	<27	<22	<19	<54					70.3 ± 16.2	10.0 ± 8.6	<8.1	8.65 ± 13.8

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a. Concentration ±2 sigma.

b. Leachate sample from 1 L sample size.

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

d. No isotope detected.

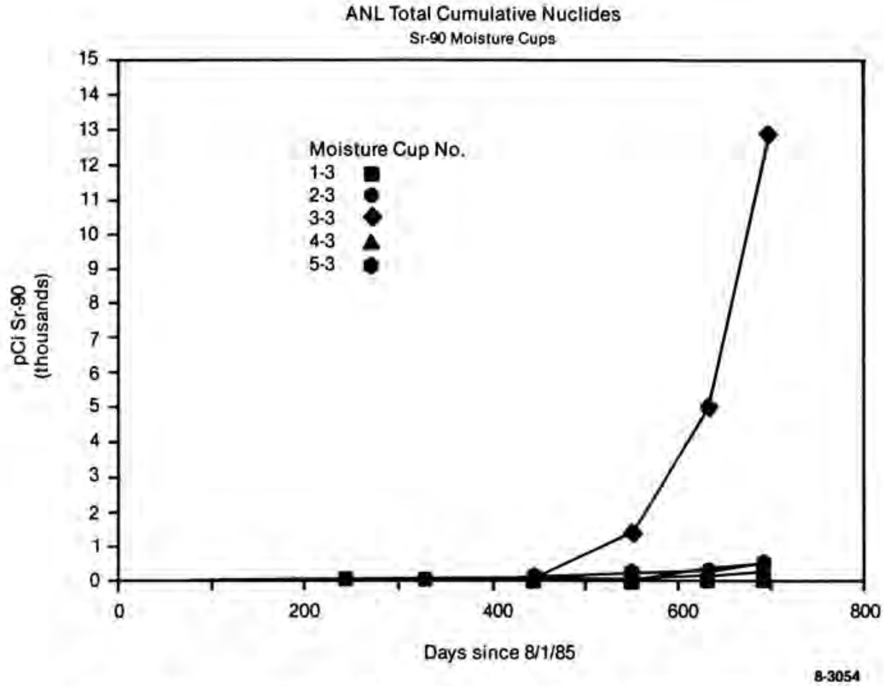

Fig. 4. ANL-E Cumulative ^{90}Sr Collected in Moisture Cups Number 3.

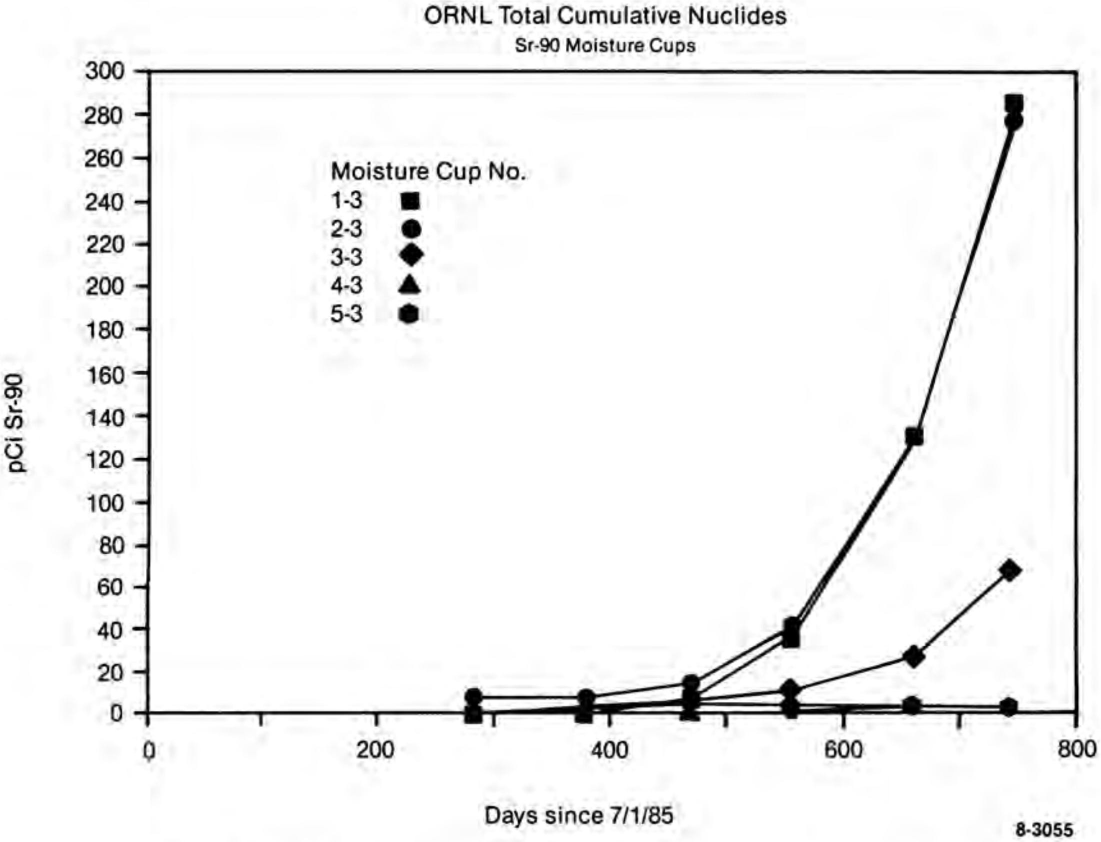

Fig. 5. ORNL Cumulative ^{90}Sr Collected in Moisture Cups Number 3.

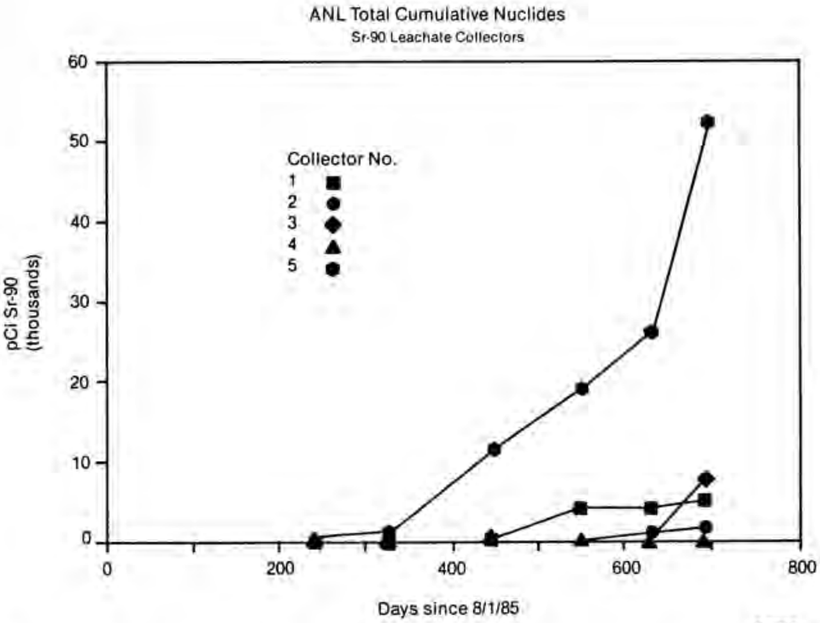

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Fig. 6. ANL-E Cumulative ^{90}Sr Collected in Lysimeter Leachate Collectors.

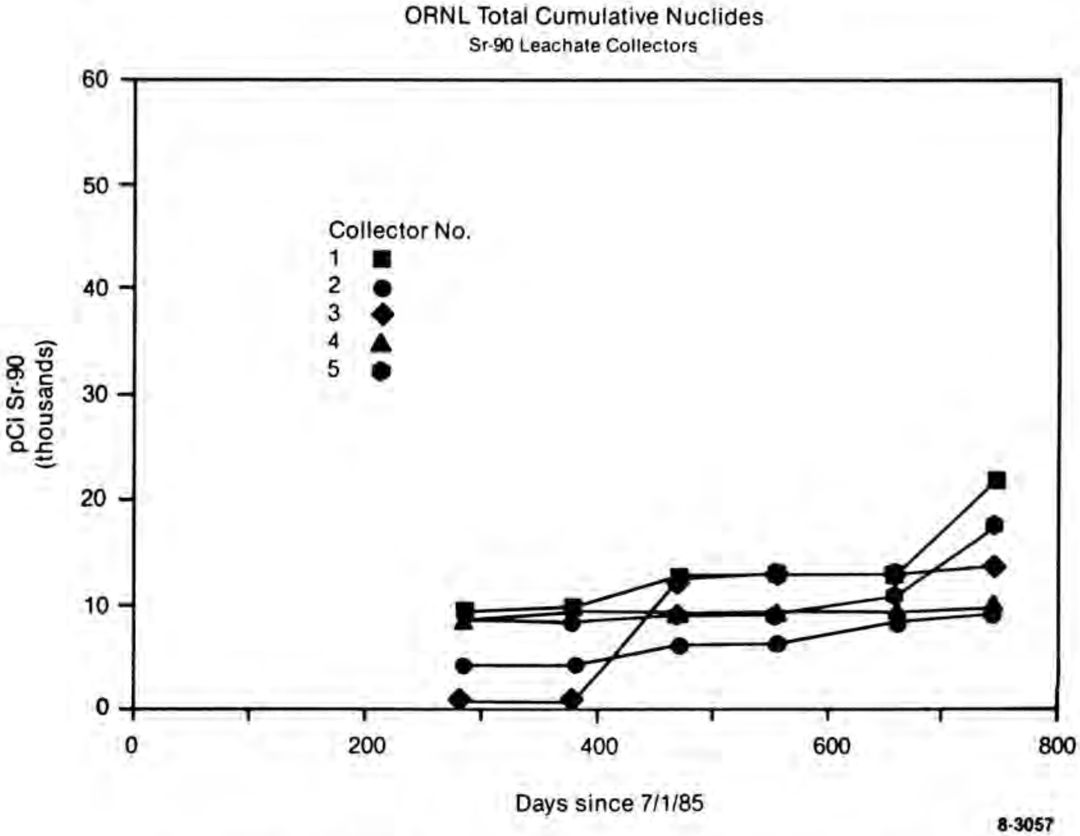

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Fig. 7. ORNL Cumulative ^{90}Sr Collected in Lysimeter Leachate Collectors.

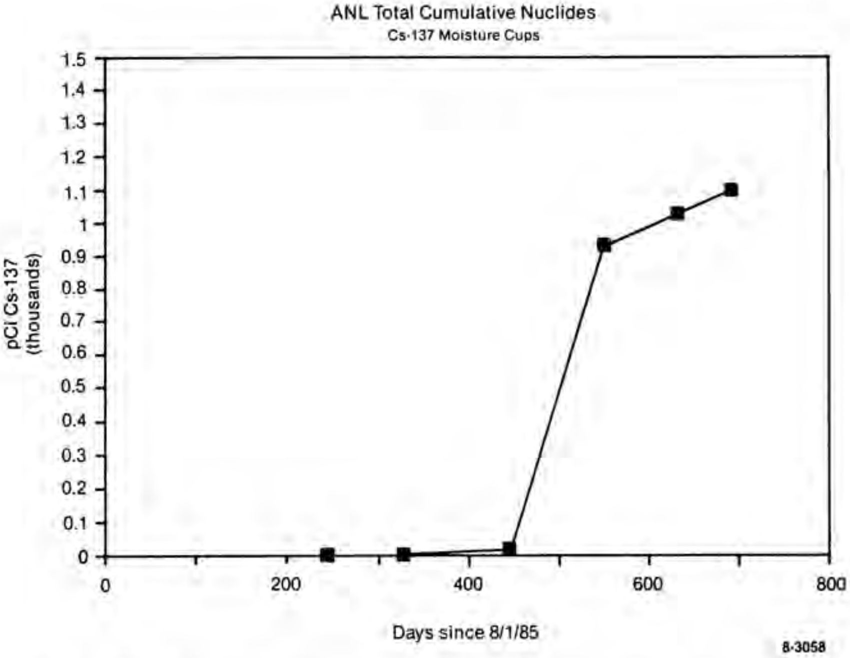

Fig. 8. ANL-E Cumulative ^{137}Cs Collected in Moisture Cup Number 5-3.

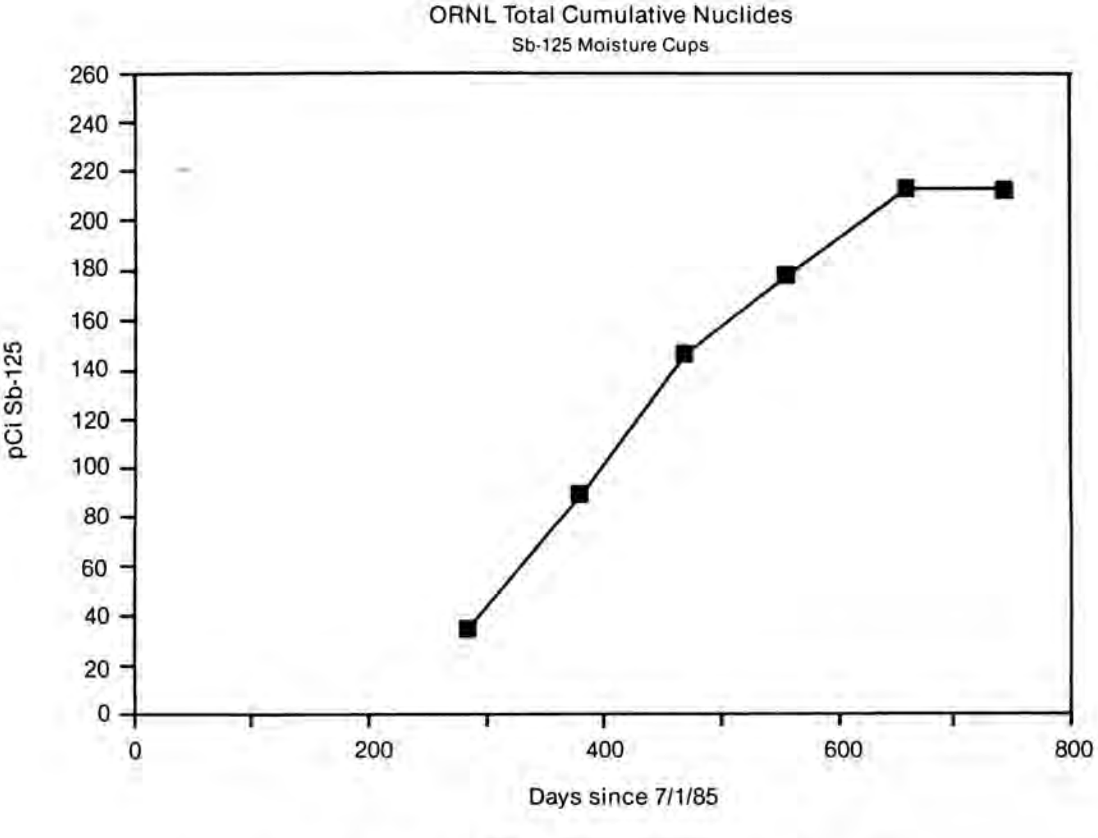

Fig. 9. ORNL Cumulative ^{125}Sb Collected in Moisture Cup 5-3.

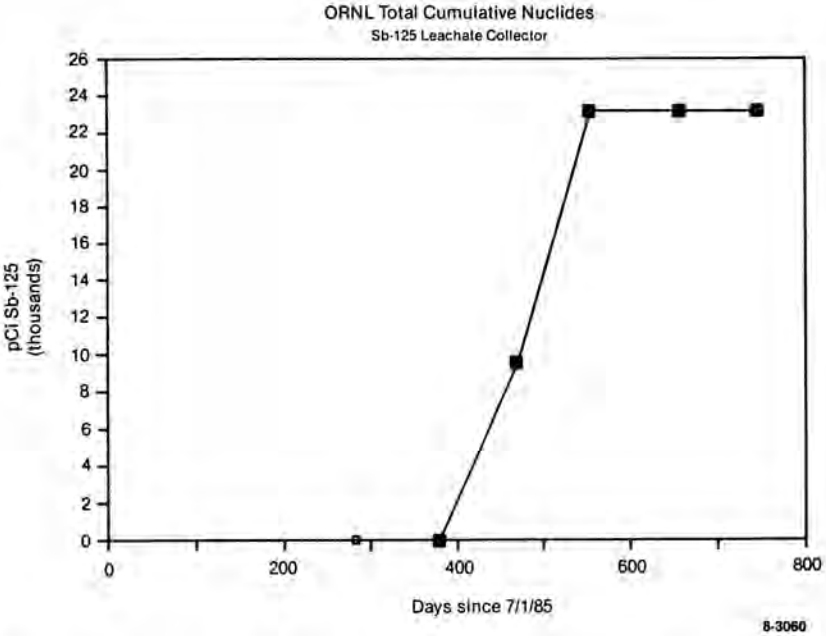

Fig. 10. ORNL Cumulative ^{125}Sb Collected in Lysimeter Leachate Collector Number 5.

this nuclide appears to have peaked in the February 1987 sample. Total quantity of ^{137}Cs recovered from cup 2-3 samples over time is 1098 pCi (Fig. 8). The amount of ^{137}Cs recovered does not represent a total for this nuclide but can be viewed as reflective of nuclide content in the soil at this depth. The rate of movement of ^{137}Cs to another detection point (cup 1 or leachate collector) will provide additional information on what facilitates movement. The fact that ^{137}Cs has only been found at ANL-E, and not at the ORNL lysimeters containing the same composition cement waste form, indicates that some characteristic of the ANL-E environment or soil or both is promoting release of ^{137}Cs from that waste form, thus facilitating movement through the soil column.

Antimony-125 has been found consistently at ORNL in cup 3 of the control lysimeter ORNL-5 (Table III). The total quantity of ^{125}Sb recovered from those liquid samples is 213 pCi (Fig. 9) which, combined with the 23,256 pCi that have passed through the control lysimeter via leach water (Table III and Fig. 10) represent approximately 0.001% of the calculated ^{125}Sb inventory of the waste form. The reason for the appearance of ^{125}Sb in only this lysimeter is not known. Because this is a control lysimeter, the quantity and timing of precipitation or the particular characteristic of the waste form is resulting in release and movement of ^{125}Sb.

Since ^{90}Sr is present in the moisture cups and leachates of all lysimeters, some initial comparisons of the four different types of waste forms (see Table I) are possible. While no conclusions can be drawn on waste form performance from the available data, some generalized observations can be made. On an intrasite comparison of leachates (Figs. 6 and 7), the soil lysimeters with vinyl ester-styrene waste forms (lysimeters 3 and 4; see Table I) have released total quantities of ^{90}Sr comparable to those lysimeters containing the cement waste form (lysimeters 1 and 2). These early observations suggest that VES is not superior to cement in its ability to retain ^{90}Sr. This conclusion is supported by the quantity of ^{90}Sr that occurred in water from ANL 3-3 (Table II) in comparison to the cement wasteform releases. The amount of ^{90}Sr that has been released and transported, however, is slight when compared with the total waste form inventory (4,9) and thus real trends may not be detectable as yet. It is also of interest to note that the quantity of ^{90}Sr recovered appears to be correlated with inventory quantities (i.e., in the case of both cement and VES waste forms, the ones with the higher inventory, lysimeters 1 and 3, generally had more ^{90}Sr appear in the leachate). The Type 1 waste form in the ANL-E control lysimeter is also releasing more ^{90}Sr than in the Type 2 control waste form at ORNL. These data suggest that the occurrence of ^{90}Sr in leachate water is dependent on release rates from the waste forms rather than transport mechanisms in the soil.

In the cases of ^{137}Cs found in ANL 2-3 water and ^{125}Sb found in ORNL 5-3 and ORNL 5 water, evaluation of the performance of the waste forms is difficult. What is noticeable is that both ANL 2 and ORNL 5 contain the same type waste form, cement with Type 2 waste. It is apparent that gaining a clear picture of waste form performance will require sampling over extended periods of time.

CONCLUSIONS

Lysimeter operation during the second year at ANL-E and ORNL has been successful. Analyses of data collected during the past 24 months is beginning to show patterns in nuclide availability and movement. Strontium-90 is still the most prevalent nuclide in collected liquid samples. Because this nuclide has been found more consistently in the leachate collectors of ORNL lysimeters (Figs. 6 and 7), there is an indication that its movement is being controlled by site-specific soil types. This conclusion is supported by data from ongoing lysimeter work at the Savannah River Laboratory (SRL) and at Hanford.

SRL has found that ^{90}Sr will move from a buried waste form, migrate through the soil column, and appear in collected leachate water (12). It is not surprising, then, that ^{90}Sr moves through soil in the ORNL lysimeters, since that soil originated at SRL (12). On the other hand, lysimeter work with waste forms at Hanford has shown that ^{90}Sr does not move in those soils (13).

Data on waste form performance presented in this paper suggest that VES is comparable to cement in its ability to retain ^{90}Sr. That is different than the results obtained at SRL. Data from that experiment show that cement minimizes the release of ^{90}Sr (12). This interesting difference should be studied further. Both data reported herein and data reported by SRL and Hanford agree that ^{137}Cs is more readily released from cement than VES.

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NOTICE

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