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-1988, presents results of waste form leach-testing, and compares those results to the releases of radionuclides from these lysimeter waste forms.

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. 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 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 ten lysimeters, five at ORNL and five 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- 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 porous cup soil-water samplers an soil moisture/temperature probes. The probes are connected to an on-site data acquisition system (DAS),

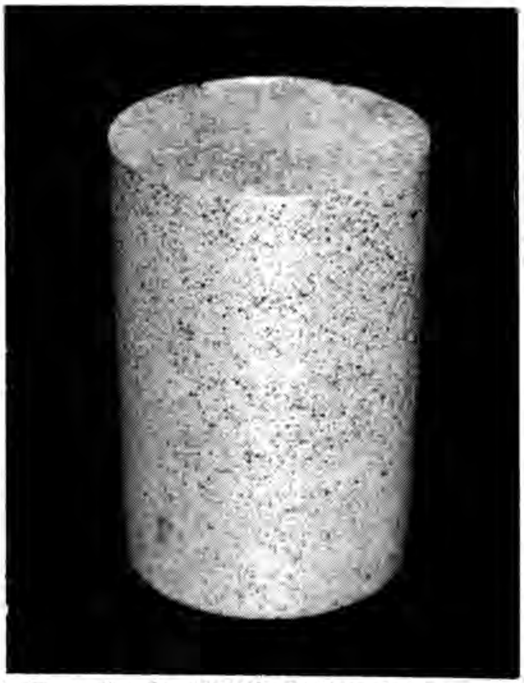

Fig. 1. Example of an EPICOR-II Waste Form.

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* Work supported by the U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, under DOE Contract No. DE-AC07-76ID01570.

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. On approximately a quarterly basis, water is drawn from the porous cup soil-water samplers and the leachate collection compartments to track the migration of radionuclides. (Because of funding constraints, ORNL did not make a January sampling in 1988.) The water samples are analyzed for beta- and gamma-producing nuclides. Details on waste-form formulation and testing, lysimeter design, installation, instrumentation, operation, and data acquisition are provided in Refs. 5 through 12.

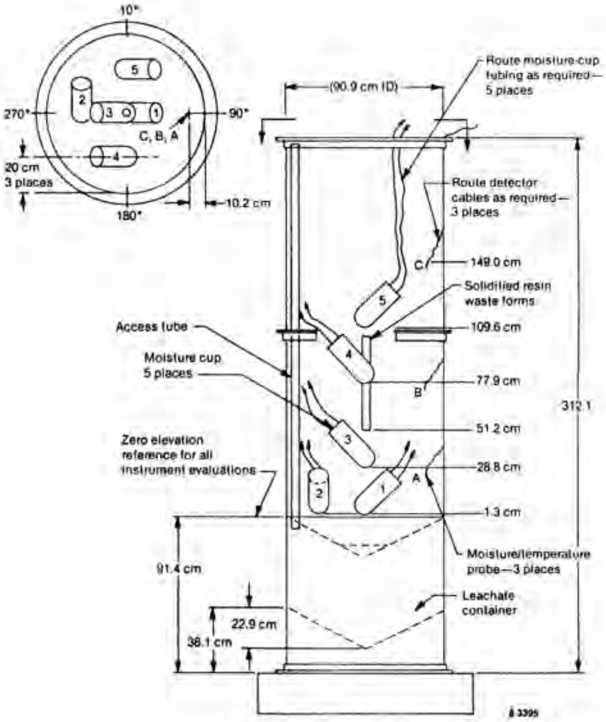

Fig. 2. Lysimeter Vessel Component Locations.

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

Monitoring of moisture cups began with collecting liquid samples in September 1985 (two to three months from the time of placement) and has continued about 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 DAS.

RESULTS AND DISCUSSION

Waste-Form Leachability

A number of EPICOR-II waste forms were subjected to the tests specified in the Technical Position on Waste Form. Leachate samples taken during leachability testing with demineralized water were analyzed for Sr-90. Those samples had been obtained during tests on waste forms containing cement binder or fabricated with VES. Both types of waste forms had been irradiated in the Advanced Test Reactor (ATR) radiation facility, using spent fuel as the radiation source (6).

The data of cumulative fractional release with time for an irradiated cement waste form (Type 1 resin) and an irradiated VES waste form (Type 1 resin) in demineralized water are plotted in Fig. 3 for Sr-90 and Cs-137. The total fractional releases were nearly identical for the two radionuclides from a specific waste form. It is noted that the cement waste form exhibited the higher fractional release of both Sr-90 and Cs-137, about 8% of the total inventories, while the VES fractional releases were about 4.5% of the inventories. The leach indices for the waste forms are also given. The cement leach indices were comparable for Sr-90 and Cs-137 (9.0 and 9.3) and lower than those of the VES (9.7). Also, the Sr-90 leached more rapidly from both types of waste forms than did Cs-137. This was particularly evident in the case of the VES waste-form, where nearly all the leachable Sr-90 had been removed in the first five days of leaching.

Figure 4 presents fractional release of Cs-137 over time in demineralized water from unirradiated Portland Type I-II cement and VES waste forms containing Type 1 and Type 2 resins. These data illustrate the lower leachability (higher leachability index) of VES compared with cement

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for the EPICOR-II resin waste forms. The waste forms containing Type 2 resins exhibited better leach characteristics for Cs-137, probably because those resins contained inorganic zeolite which does not degrade with the radiation doses observed in the EPICOR-II prefilters.

A brief comparison of the information of Figs. 3 and 4 shows that the leachability of the waste forms was higher with a higher irradiation dose. This effect was more pronounced with VES waste forms.

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 88.8 cm at ANL-E and 82.6 cm at ORNL. ANL-E was very close to their normal annual rainfall of 85.2 cm (10); while, for a third year, ORNL was low at about 40% of their normal annual rainfall of 138.8 cm (12). Fig. 5 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 1987 until near mid-March 1988. ORNL experienced several days where there was an air temperature as low as 0°C (typical data shown in Refs. 8, 9 and 10).

Examples of the lysimeter soil temperature data recorded over one-year periods at ANL-E and ORNL can be found in Refs. 8, 9 and 10. At no time during the FY-1988 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.

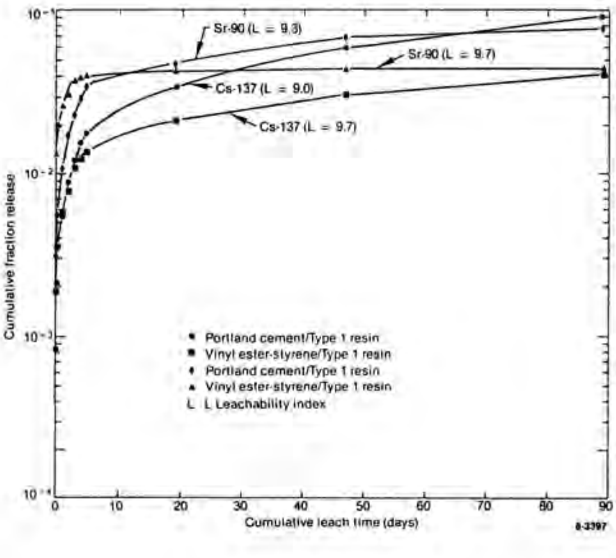

Fig. 3. EPICOR-II Waste Form Radionuclide Cumulative Release After Irradiaiton.

Examples of data from the moisture probes at both ANL-E and ORNL can be found in Refs. 8, 9, and 10. Data recorded in FY-1988 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 all 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. 5), it is possible to calculate the volume of water which has been received by the exposed lysimeter surfaces (6489.5 cm2). The cumulative volume of precipitation received by each ANL-E lysimeter was 1639.8 L; at ORNL, this value was 1965.0 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 783.5 + 182.9 L, or 47.8% of total precipitation, passed through the soil lysimeters; while for the control lysimeter, this value was 1567 L, or 95.6% of available precipitation. For ORNL, the values were 1598 ± 21.2 L (82.7%) for the soil-filled lysimeters and 1931 L (97.4%) for the control lysimeter. Not only does precipitation have Fig. 5. ANL-E and ORNL cumulative precipitation.

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

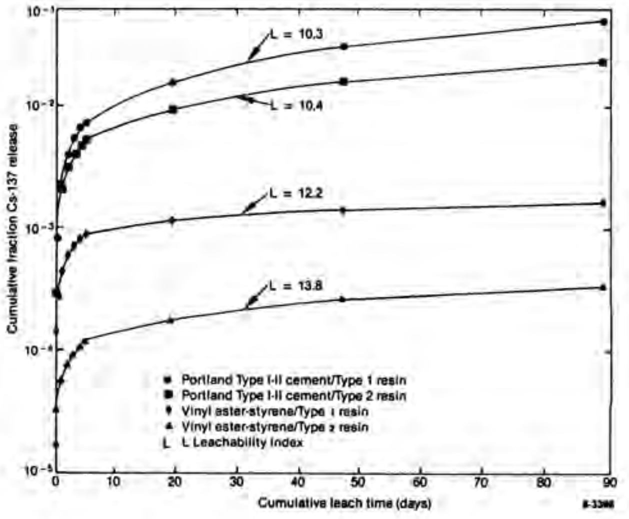

Fig. 4. Cumulative Fractional Release of Cs-137 From Unirradiated Waste Forms.

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 has been drained from the surface of these lysimeters and later replaced. It was determined that the soil structure of the top several inches of soil had been compacted. Fresh soil was used to replace the compacted soil in FY-1987, which reduced the ponding problem significantly. The problem reoccurred in FY-1988 and was thought to be caused by precipitation impact. The soil was screened and again replaced, and a polyester mesh and pea gravel were placed over it. The lack of infiltration was again relieved.

The total volumes of precipitation that have moved through the lysimeters represent an average 1.10 pore volumes for the ANL-E soil lysimeters and 2.25 pore volumes for soil lysimeters at ORNL, while the control lysimeters at ANL-E and ORNL were 2.21 and 2.71 pore volumes, respectively. Thus, since the beginning of the study, 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 twice 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 ANL, while only three were obtained at ORNL due to a delay in FY-1988 funding. The cumulative amounts of nuclides as determined in water samples obtained from lysimeter cups 3 and leachate collectors for all sampling periods are displayed graphically in Figs. 6 through 12.

It is apparent from these data that not all nuclides are appearing consistently in the water obtained from the cups and leachate collectors. The nuclide which appears with the most regularity at both sites is Sr-90. Consistent significant occurrences, however, have been in all the cups 3 at ANL, the cups 3 (except 4-3) at ORNL, and the number 5 leachate collectors at both sites (Figs. 6 through 9). There are standout amounts of Sr-90 retrieved from cup samples at both sites. Those include 100,190 pCi from 3-3 at ANL (Fig. 6) and 3,555 pCi from 1-3 at ORNL (Fig. 7). As noted in the Waste-Form Leachability section of this report, Sr-90 leachates 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. 6 and 7, respectively) when plotted with time form what appear to be the beginnings of typical leach rate curves, indicating that

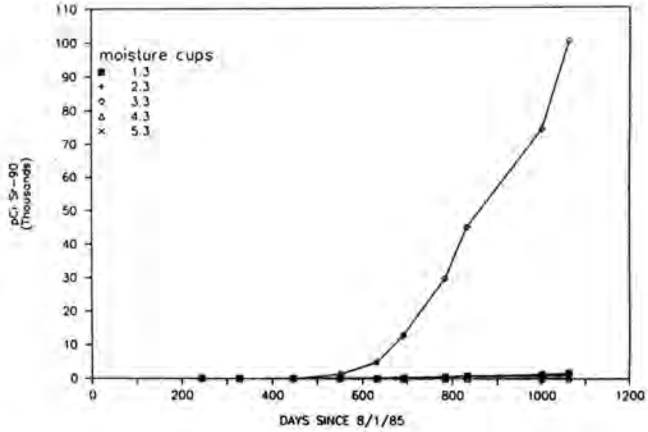

Fig. 6. ANL-E Cumulative Sr-90 Collected in Moisture Cup Number 3.

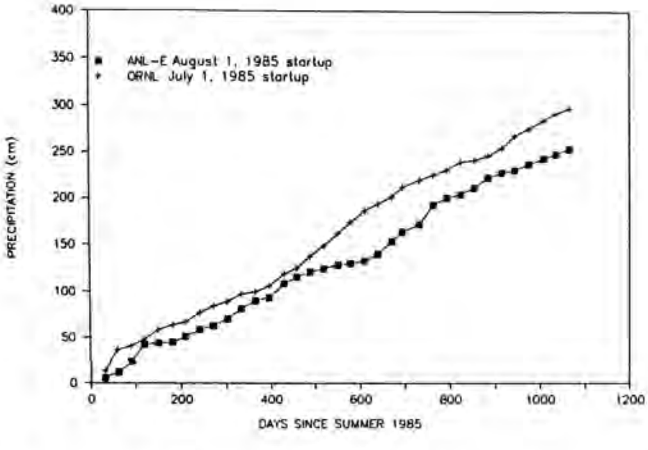

Fig. 5. ANL-E and ORNL Cumulative Precipation.

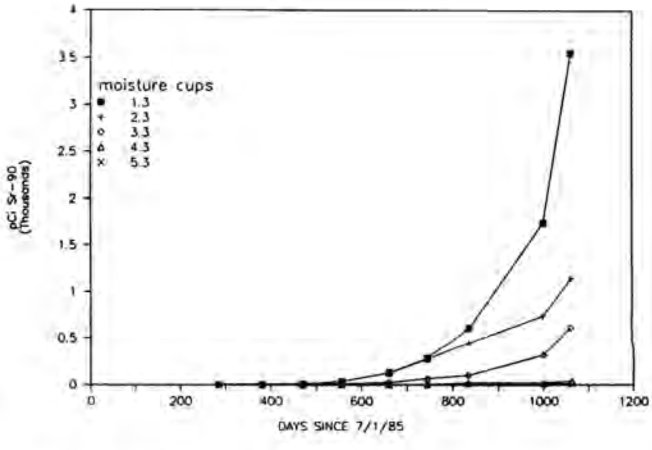

Fig. 7. ORNL Cumulative

TABLE II

Results of BETA and GAMMA Analysis of ANL-E Soil Moisture and Leachate Samples--Year 3--1987-88.

	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
Sample Number	September 1987	November 1987	April 1988	June 1988	September 1987	November 1987	April 1988	June 1988	September 1987	November 1987	April 1988	June 1988
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	<1
LYS 4	<1	<1	<1	<1	<1	2 ± 1	<1	<1	<1	<1	<1	<1
LYS 5	<1	<1	<1	<1	39 ± 3	2 ± 1	<1	3 ± 1	126 ± 1	99 ± 4	187 ± 7	139 ± 2
LYS 1-3[c]	<5	<5	<5	<5	6 ± 5	<5	9 ± 7	8 ± 6	2886 ± 68	5173 ± 195	6423 ± 259	6680 ± 135
LYS 2-3	<5	<5	<5	<5	666 ± 46	317 ± 35	191 ± 25	255 ± 15	1350 ± 18	1588 ± 26	1281 ± 30	1182 ± 26
LYS 3-3	<5	<5	<5	<5	<5	<5	9 ± 12	<5	1.7E5 ± 1648	1.9E5 ± 2894	3.6E5 ± 601	2.6E5 ± 3720
LYS 4-3	<5	<5	<5	<5	<5	<5	<5	33 ± 8	99 ± 24	142 ± 25	133 ± 9	1140 ± 38
LYS-5-3	<5	<5	<5	<5	45 ± 24	69 ± 14	200 ± 25	521 ± 27	2870 ± 318	4089 ± 502	187 ± 7	6283 ± 151

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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.

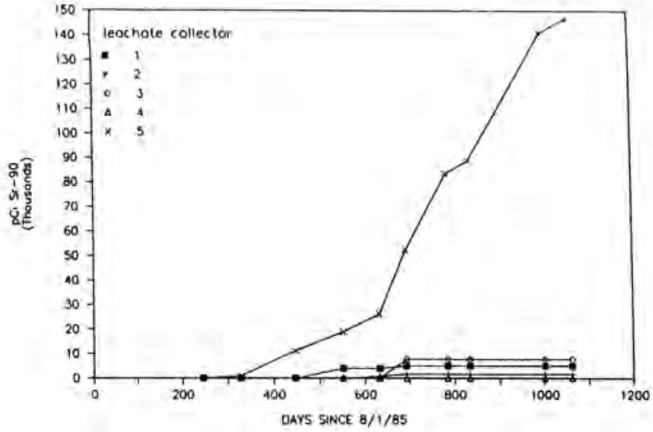

Fig. 8. ANL-E Cumulative Sr-90 Collected in Lysimeter Leachate Collectors.

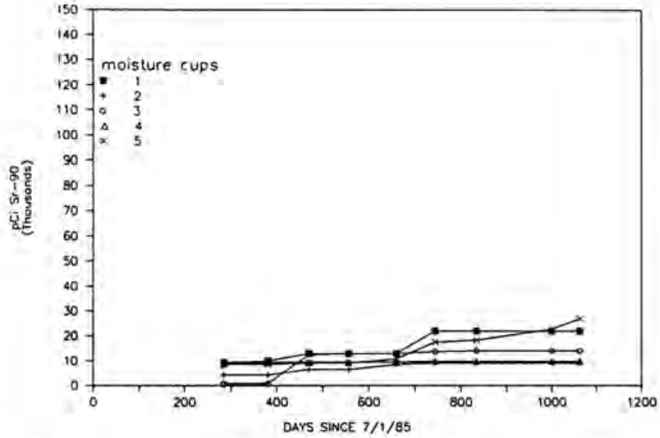

Fig. 9. ORNL Cumulative Sr-90 Collected in Lysimeter Leachate Collectors.

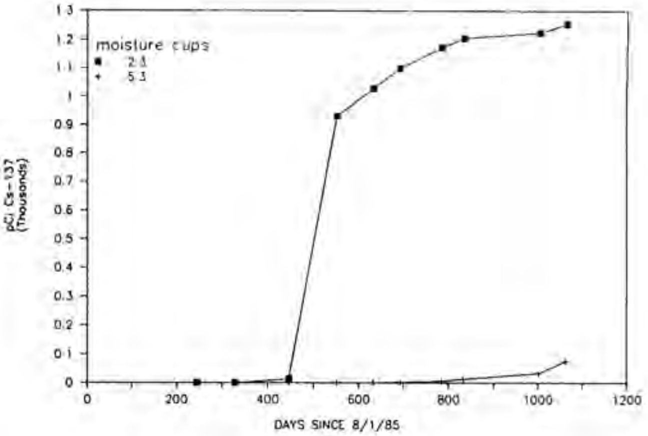

Fig. 10. ANL-E Cumulative Cs-137

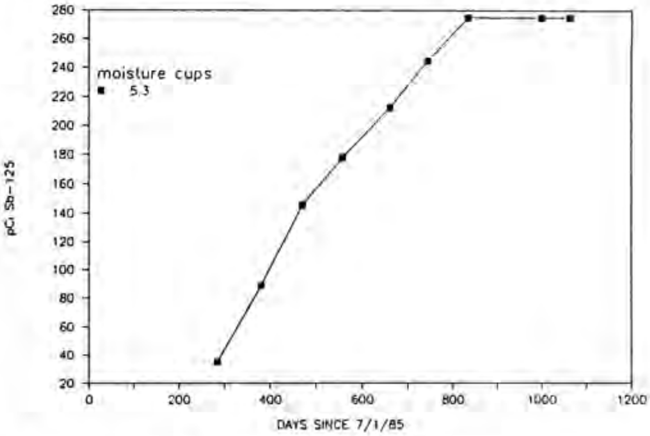

Fig. 11. ORNL Cumulative Sb125 Collected in Moisture Cup Number 3.

and 3,555 pCi from 1-3 at ORNL (Fig. 7). As noted in the Waste-Form Leachability section of this report, Sr-90 leachates 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. 6 and 7, 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 3-3 cup represents about 0.0003% of the waste form inventory (12).

At ANL, Sr-90 retrieved from the cups 3 of the soil lysimeters range from 25% to 1000% of that found in the leachate collectors, while at ORNL these values range from 0.2% to 8%.

It is of interest that during this past 12 months only the leachate water from the control lysimeters and none from the soil lysimeters at both sites contained significant amounts of Sr-90. This is different from the previous years findings (Refs. 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 (except ORNL-4). Except for ANL 3-3, the percent of total Sr-90 being measured in the cups 3 is practically the same (12). This indicates that the waste forms are performing much the same at both sites. These data are interesting,

TABLE III

Results of BETA and GAMMA Analysis of ORNL Soil Moisture and Leachate Samples--Year 3--1987-88

	Concentration (pCi/L)[a]											
	Co-60			Cs-137			Sb-125			Sr-90		
Sample Number	October 1987	April 1988	July 1988	October 1987	April 1988	July 1988	October 1987	April 1988	July 1988	October 1987	April 1988	July 1988
LYS 1[b]	<27	<8	<8	<27	<8	<8				0.08 ± 4.0	2.97 ± 3.5	<2.7
LYS 2	<16	<8	<5	<22	<8	<5				0.73 ± 2.7	1.35 ± 2.7	<2.7
LYS 3	<19	<8	<5	<19	<5	<5				7.3 ± 5.1	0.00 ± 2.7	2.7 ± 3.2
LYS 4	<19	<8	<5	<24	<8	<5				0.57 ± 2.8	3.24 ± 4.8	<2.7
LYS 5	<19	<8	<5	<22	<5	<5	<54	<24	<13	8.65 ± 4.3	12.2 ± 4.8	18.9 ± 5.4
LYS 1-3[c]	<19	<54	23.5 ± 18.7	<24	<54	<54				3243 ± 270	11350 ± 270	18123 ± 541
LYS 2-3	<16	<54	<27	<27	<54	<54				1675 ± 54	2970 ± 270	4057 ± 270
LYS 3-3	35 ± 32	<54	<27	<27	<54	<19				378 ± 27	2160 ± 270	2975 ± 270
LYS 4-3	<16	<54	<27	<16	<27	<21				167 ± 205	2.16 ± 14.1	186 ± 78
LYS 5-3	51 ± 24	<54	<27	<24	100 ± 40	162 ± 32	297 ± 81	<108	<81	13.5 ± 5.5	32.4 ± 21.6	8.3 ± 19.2
LYS 1-1	<19	<54	<57	24 ± 24	<81	<27				3.5 ± 3.8	<13	<2.7
LYS 2-1	<27	<54	<27	<27	<54	<27				1.1 ± 3.3	20.3 ± 18.4	<2.7
LYS 3-1	<19	<54	<27	<22	<27	<27				2.9 ± 3.2	10.8 ± 17.0	<2.7
LYS 4-1	<22	<54	<24	<22	<54	<21				5.9 ± 6.2	<13	4.8 ± 15.7
LYS-5-1	<19	<54	<21	<22	<27	<21				4.0 ± 5.4	16.8 ± 15.1	32.5 ± 18.9

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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.

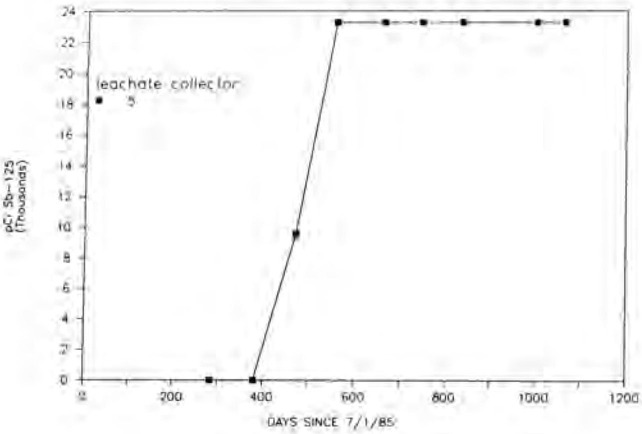

Fig. 12. ORNL Cumulative Sb-125 Collected in Lysimeter Leachate Collectors.

since the waste forms at each site have been experiencing similar exposure to moisture and temperature (12).

In contrast to the last report (11), gamma-producing nuclides have occurred with regularity only at ANL-E. ANL 2-3, below a cement waste form containing large amounts of Cs-137, continues to receive Cs-137 (Table II), though the quantity of this nuclide appears to be decreasing after peaking in the February 1987 sample (Fig. 10). The shape of that curve is very similar to those produced by the bench leaching experiments (shown in Figs. 3 and 4), that is, an initial rapid rise in Cs-137 followed by a flattening. It would appear that Cs-137 release from that waste form is controlling movement of that nuclide through the soil. Since June of 1987, Cs-137 has begun appearing in ANL 5-3. TABLE II

The quantity of this nuclide appears to be increasing each sampling period (Fig. 10). No sustained occurrence of Cs-137 has been detected in water from the ORNL lysimeters. A measurable amount of Cs-137 was found in ORNL 5-3 during the April 1988 sampling. As was concluded in the last report (11), since Cs-137 has only been found at ANL-E and not at the ORNL soil lysimeter containing the same cement waste form, there is some indication that characteristics of the ANL-E environment and/or soil are promoting release of Cs-137 from that waste form and/or facilitating movement through the soil column.

Antimony-125 has been found consistently at ORNL in cup 3 of the control lysimeter ORNL-5 until April of 1988 (Fig. 11 and Table III). The total quantity of Sb-125 recovered from those water samples is 275 pCi (Fig. 12). No further Sb-125 has been recovered from leach water since January 1987, when it was calculated that approximately 0.001% of the Sb-125 inventory from the waste form had been recovered. These curves also resemble the bench leach results for Sr-90 and Cs-137 (Fig. 3), indicating that the limiting factor on movement of Sb-125 in this lysimeter is release from the waste form.

On an intrasite comparison (Figs. 7 and 8), the conclusion 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, since none of those lysimeters released additional Sr-90 to the leachate collectors. Data from the cups 3 also tend to support the evidence that VES is no better at retaining Sr-90 than cement (Figs. 6 and 7). Based on percent of total inventory release to the cups 3, ANL-1 and -2 (cement) have received 26 to 7 E-6%, respectively, of the Sr-90, while ANL-3 and -4 (VES) have received 207 to 1 E-6%, respectively. Comparable data at ORNL for the cement are 10 to 22 E-6%, while those for VES are 0.4 to 1 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 as well as Sb-125 from ORNL-5 continue to be of interest, but lack of occurrence in other lysimeters with the same type waste forms make it difficult to draw conclusions. Since Cs-137 has continued to appear in ANL 5-3, this 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.

CONCLUSIONS

The leaching of Sr-90 from both Portland Type I-II cement and VES waste forms containing EPICOR-II ion exchange resins occurred early in the bench leachability tests of irradiated waste forms.

The fractional releases of Sr-90 and Cs-137 from the Portland Type I-II cement waste form in bench tests were comparable as were those from the VES waste form.

In the lysimeter experiments, strontium-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. However, this movement appears to be only of importance initially, since Sr-90 movement through these soils has not been noted during the last year. During the past 12 months, Sr-90 continued to be found in leachate water in the control lysimeters at both sites. It appears then that the limiting step in receiving Sr-90 in the leachate is not release of the nuclide from the waste forms (since Sr-90 is found in cups 3), but rather it is the soil profile (including soil and quantity of soil water) which limits movement. This conclusion is supported by data from ongoing 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). This interesting difference should be studied further. Both data reported herein and data reported by SRL and Hanford agree that Cs-137 is more readily released from cement than VES. A comparison of release of Cs-137 and Sb-125 from lysimeter waste forms with bench leachability results from similar waste forms indicates that the release of these two radionuclides is being controlled by leaching from the waste forms.

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