FIELD TESTING OF WASTE FORMS USING LYSIMETERS[a]

J. W. McConnell, Jr., R. D. Rogers
Idaho National Engineering Laboratory
EG&G Idaho, Inc.
Idaho Falls, ID 83415

ABSTRACT

The Low-Level Waste Data Base Development--EPICOR-II Resin/Liner Investigation Program funded by the U.S. Nuclear Regulatory Commission is obtaining information on performance of radioactive waste in a disposal environment. Waste forms manufactured from ion exchange resins used to clean up water from the accident at Three Mile Island Nuclear Power Station are being examined in field tests. This paper presents a description of the field testing and results from the first year of operation.

INTRODUCTION

Ion exchange resin materials from EPICOR-II prefilters used 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 composed of ion exchange media loaded with radionuclides and solidified in matrices of cement and Dow polymer. Emphasis is placed on evaluating the requirements of 10 CFR 61 "Licensing Requirements for Land Disposal of Radioactive Waste" (1) by obtaining data on performance of waste in a disposal environment. The field testing experiment consists of using lysimeter arrays at Argonne National Laboratory (ANL-E) in Illinois and Oak Ridge National Laboratory (ORNL) (2,3).

METHODS AND MATERIALS

The two types of wastes used in the field testing experiment include (a) a mixture of synthetic organic ion exchange resins and (b) a mixture of organic ion exchange resins and an inorganic zeolite. Solidification agents used to produce the 4.8- by 7.6-cm cylindrical waste forms (Fig. 1) examined in the study were Portland Type I-II cement and Dow vinyl ester-styrene (4). Seven waste forms were stacked end-to-end and inserted into each lysimeter (5 lysimeters at ORNL and 5 at ANL-E) to provide a 1-L volume.

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

Lysimeters used in this study were designed to be self-contained units which will be disposed upon termination of the 20-year study. Each is a 0.91- by 3.12-m right-circular cylinder divided into an upper compartment containing fill material, waste forms, and instrumentation, and a lower compartment which collects leachate (Fig. 2). Four lysimeters at each site are filled with soil, while the 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 the Savannah River Laboratory, SC. Instrumentation within each lysimeter includes five porous cup soil-water samplers (moisture cups) and three soil moisture/temperature probes. The probes are connected to an on-site data acquisition and storage system (DAS) which also collects data from a field meteorological station located at each site.

Each month, the data stored on cassette tape are retrieved from the DAS and translated into an IBM PC compatible disk file. At least quarterly, water is drawn from the moisture cups and leachate collector of each lysimeter. Moisture cups are identified first by the lysimeter number and second by the cup location (see Fig. 2); thus the cup directly below the waste form of Lysimeter 5 is 5-3. The water samples are analyzed for beta and gamma emitting radionuclides. Details on formulation of the waste forms; design, installation, and instrumentation of the lysimeters; and acquisition and storage of the data are provided in Ref. 4-7. Highlights of the test results are presented in this paper. A detailed presentation of the data and discussion of results is given in Ref. 8.

Monitoring of soil moisture cups to track migration of radionuclides began with the first collection of liquid samples in September 1985 (three months after the time of placement) and continued every three months thereafter. The monitoring includes sampling and chemical analysis of liquids obtained from locations near the waste forms. Soil moisture/temperature data at three elevations in each

a. Work supported by the U.S. Nuclear Regulatory Commission, Office of Nuclear Materials Safety and Safeguards under DOE Contract No. DE-AC07-76ID01570.

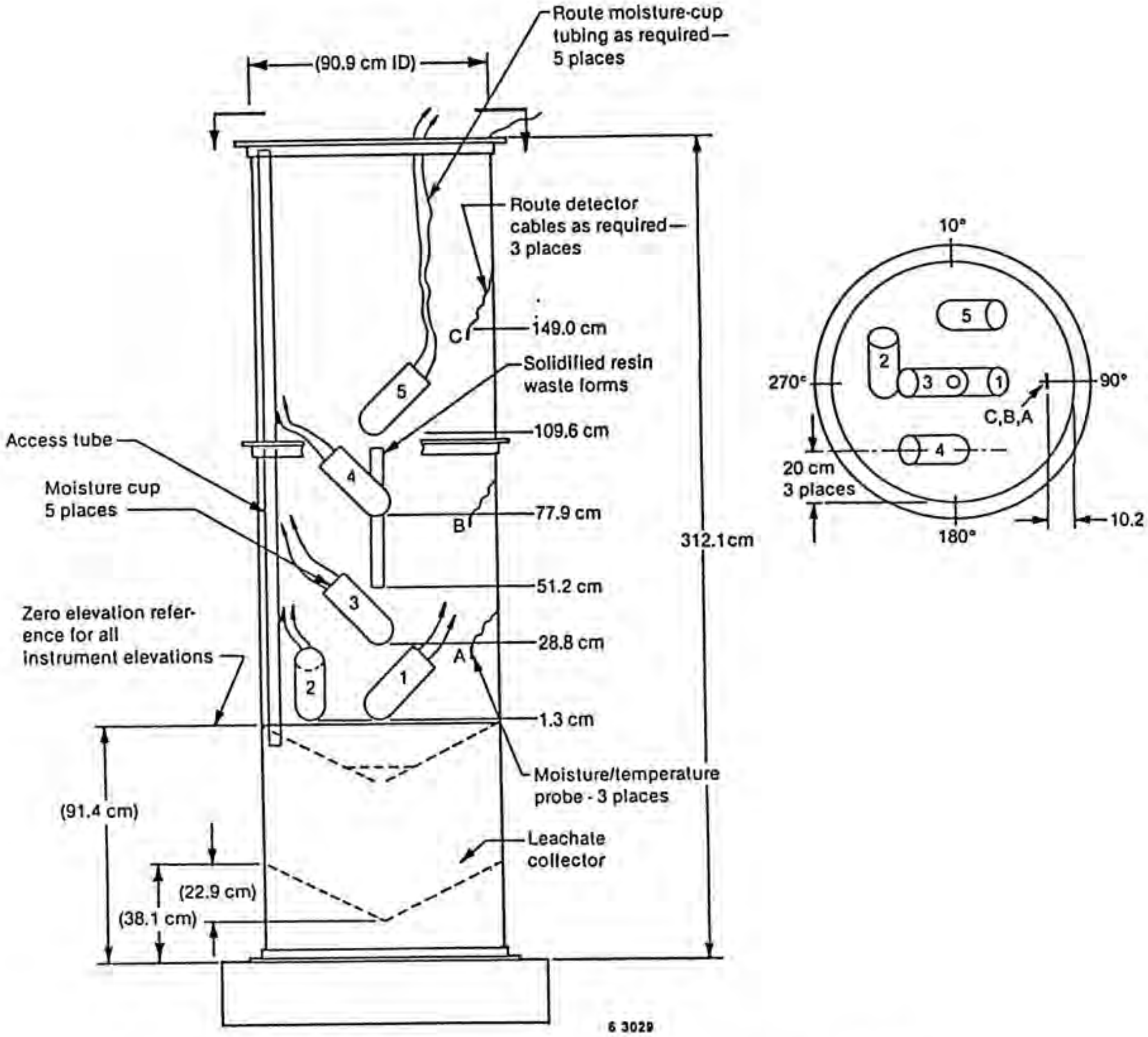

Fig. 2. Component locations in the lysimeter vessel.

lysimeter, along with a complete weather history, are recorded on a continuous basis by the DAS.

RESULTS AND DISCUSSION

Results obtained from the first year of field testing are presented in this section. A few minor problems were experienced with both the ANL-E and ORNL DAS during the first year. There also was a period from late August 1985 until November 1985 when the ORNL system was inoperable, due to the necessity of returning the system to the manufacturer for repairs.

Weather Data

Wind speed, air temperature, relative humidity, and rainfall data are recorded over a 12-month period by the DAS at each site. Air temperature data from ANL-E (Fig. 3) show that there were many days from mid-November 1985 until mid-March 1986 when air temperatures were below freezing, while there were very few days when air temperatures reached 0°C at ORNL (Fig. 4). Rainfall data from the ORNL site appeared to be higher than normal. This trend became apparent during December 1985, and indications were that the Weather Measure tipping bucket rain gauge supplied with the DAS was not capable of accurately responding to periods of intense rainfall. In June 1986, that rain gauge was replaced with a Clima-

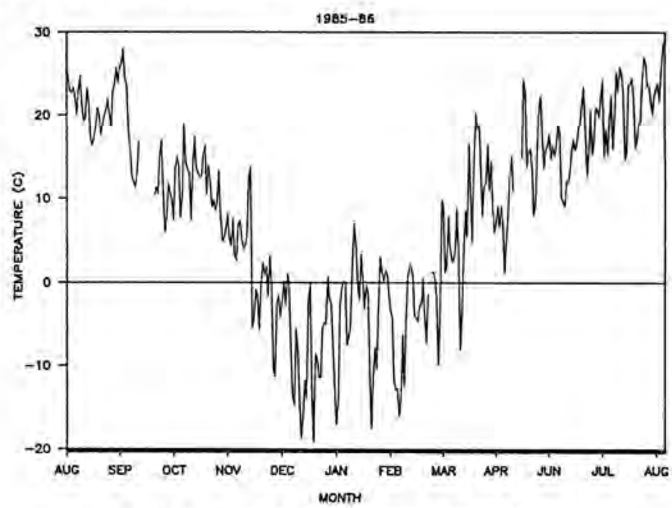

Fig. 3. ANL-E air temperature data.

tronics tipping bucket gauge designed for episodic high-intensity rainfall. Data from the replacement gauge appear to be accurate most of the time; however, the rainfall data recorded by the DAS contain occasional, erroneously high data points. Corrective measures for determining the source of the spurious data are ongoing. The malfunctions have not resulted

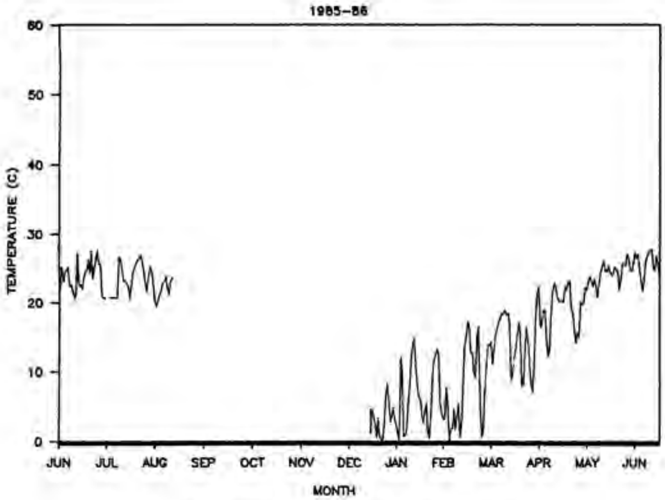

Fig. 4. ORNL air temperature data.

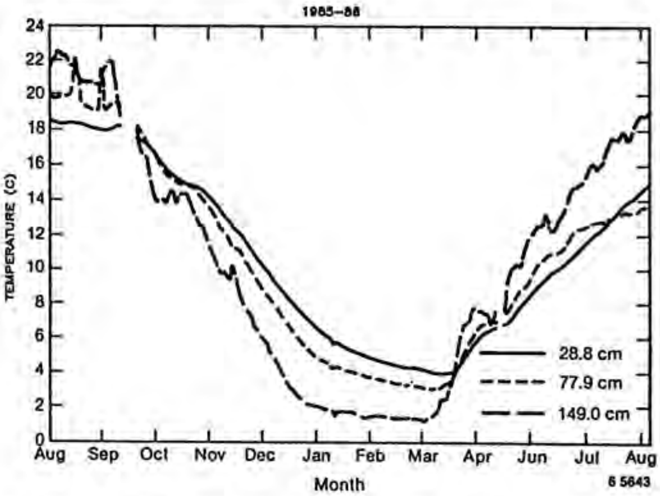

Fig. 5. Soil temperature data from ANL-E Lysimeter 2.

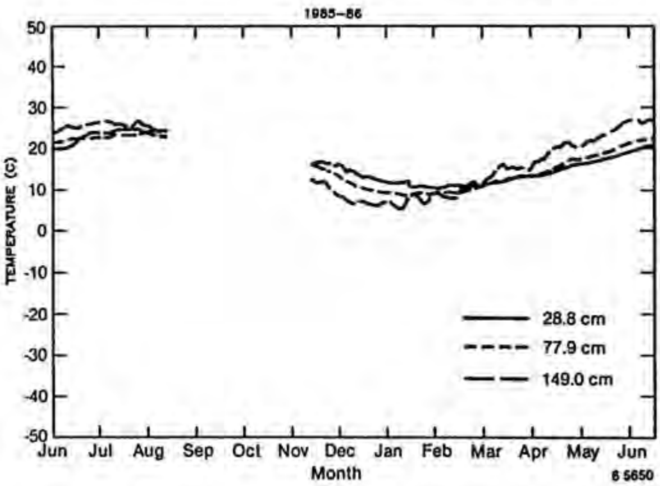

Fig. 6. Soil temperature data from ORNL Lysimeter 2.

in a loss of rainfall data, since both ANL-E and ORNL have mechanical recording rain gauges in close proximity to the lysimeter sites. Data from those nearby rain gauges (Table I) were used to calculate the total precipitation received at each site.

TABLE I

Yearly Precipitation at ANL-E and ORNL as Measured by Back-up Instrumentation--July 1985 through July 1986

Month	Precipitation (cm) ANL-E	ORNL
July[a]	--	13.3
August[b]	5.6	23.1
September[c]	6.3	4.3
October	11.6	7.6
November	18.9	10.2
December	1.7	5.3
January	0.6	3.1
February[c]	6.4	10.4
March	7.6	7.1
April[c]	4.0	5.1
May	7.7	7.7
June[c]	11.1	2.6
July	8.6	
Total	93.5	99.0

a. ORNL precipitation measurements initiated in July.

b. ANL-E lysimeter experiment initiated in August.

c. Months that leachate was retrieved for analyses.

Lysimeter Soil Temperature Data

Data from temperature probes located in all ten lysimeters at the depth of the waste forms (77.9 cm) indicate that at no time were the waste forms exposed to freezing temperatures (see Figs. 5 and 6 for Lysimeter 2 data). The soil (or sand) temperature data further show (as would be expected) that the near-surface soil temperatures (elevation 149.0 cm, 66.7 cm below the soil surface) fluctuated more than those of the intermediate (elevation 77.9 cm) or bottom (elevation 28.8 cm) soils. It also is noted from the data that the frost line in the soil did not move as deep as the first probe (66.7 cm below the soil surface).

Some abnormally low soil temperature readings were observed from the intermediate and bottom probes in ANL-E Lysimeter 3 in January 1986 and Lysimeter 4 in June 1986. There were no such occurrences with near-surface probes. One possible explanation for the malfunction is related to an average soil subsidence of 30 cm in all ANL-E soil-filled lysimeters. That subsidence was manifested as a general settling of the soils and waste forms. It is hypothesized that subsiding soil may have caused damage to the lead wires connecting the lower probes to the DAS. Those probes are being replaced with new units, and recent data from the replacement probes show they are functioning normally.

The bottom temperature probes in ORNL Lysimeters 3 and 5 have consistently indicated high temperatures compared with temperatures measured by other probes in those and nearby lysimeters. Because the abnormal readings began close to the time of lysimeter installation, it is possible that the probes or wiring were damaged during installation. In any case, they are to be replaced.

Lysimeter Soil Moisture Data

Moisture probe data from the two sites show that two (ORNL) to three (ANL-E) months were required after the lysimeters were filled with soil for the soil to reach saturation (Figs. 7 and 8). As a precaution, the accuracy of the probes in the soil-filled lysimeters was determined by comparing their data with the gravimetric water content of soil cores retrieved from all four ORNL soil-filled lysimeters and one ANL-E lysimeter. From those comparisons it was apparent that the probes are overestimating the soil moisture content. Corrective action is ongoing and consists of recalculating the polynomial equation that transforms probe input into percent moisture using laboratory and, if necessary, field soil gravimetric measurements.

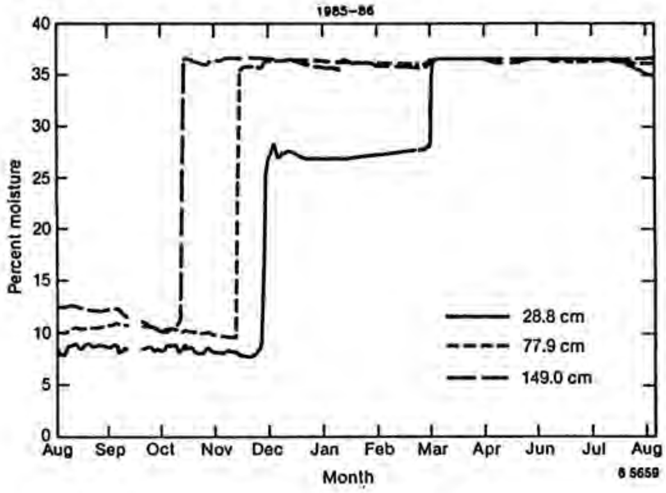

Fig. 7. Soil moisture data from ANL-E Lysimeter 2.

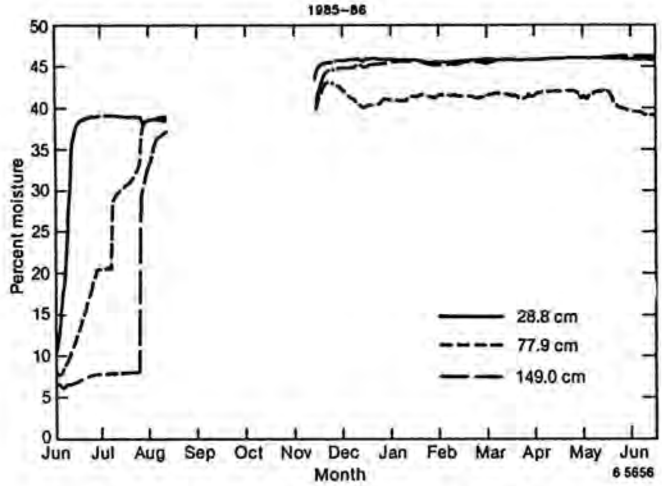

Fig. 8. Soil moisture data from ORNL Lysimeter 2.

During a 12-month period, the ANL-E site received 93.5 cm of precipitation. ORNL received 99.0 cm, well below the norm of 134 cm. Using these values and the exposed surface area of the lysimeter (6489.5 cm^2), it was calculated that the ANL-E and ORNL lysimeters received 607 L and 643 L of water, respectively. The total water retrieved from the leachate collectors of each lysimeter is shown in Table II. The leachate collectors of the soil-filled lysimeters at ANL-E contained an average of 129 L; those at ORNL contained an average of 442 L. The leachate collectors of the sand-filled lysimeters at ANL-E and ORNL contained 338 L and 528 L, respectively.

The two sites received comparable volumes of precipitation, however the water moved through the lysimeters at those sites in unequal amounts due to differences in soil texture and weather conditions. The soil used at ANL-E is heavier and contains more fine material such as silts and swelling clay than that used at ORNL (5). Therefore, infiltration and percolation of water through the ANL-E soil would be reduced in comparison with ORNL soil. The effect of weather is especially apparent when comparing the sand-filled control lysimeters at the two sites. At ANL-E, 55% of the volume of precipitation passed through the lysimeter, versus 82% at ORNL.

TABLE II

Total Quantities of Leachate Retrieved from Lysimeters during a 12-Month Period[a]

	Quantity of Water (L)				
	Lysimeter				
Site	1	2	3	4	5
ANL-E	113	132	160	112	338
ORNL	415	438	449	465	528

a. August 1985 through July 1986 for ANL-E; July 1985 through June 1986 for ORNL.

Based on the amount of water retrieved from the lysimeters, 0.18 pore volumes of water passed through the ANL-E soil-filled lysimeters, while 0.62 pore volumes passed through the similar ORNL lysimeters. Pore volumes for the control (sand-filled) lysimeters were 0.57 at ANL-E and 0.94 at ORNL. Theoretically, 18% of the water held in the soil pore space of the ANL-E soil-filled lysimeters was replaced during the year, while 62% was replaced in the ORNL lysimeters. Similarly, 57% and 94% was replaced in the ANL-E and ORNL sand-filled lysimeters, respectively. Therefore, if radionuclides were in the water surrounding the waste forms, the greatest opportunity for detection would be found in water from the ORNL site. [This is based on two assumptions: (a) the radionuclide is water soluble and (b) the soil column does not interfere with radionuclide movement.] It is noted that Co-60 and Sr-90 move through soils freely, while Sb-125 and Cs-137 are readily retained by most soils. The inert sand used in Lysimeter 5 at each site was selected because it exhibits a low exchange capacity, and thus does not interfere with the movement of the radionuclides being investigated. Results of the difference in water volume throughput are discussed in the following section.

Radionuclide Analysis

Gamma emitting radionuclides were not found in the first two samplings from the lysimeters. However, in April 1986, Co-60 was discovered in water samples from ANL-E moisture cup 3-3 (Table III); Cs-137 was found in the leachate of ANL-E Lysimeter 5 (the sand-filled lysimeter); and Sb-125 was found in ORNL moisture cup 5-3 (Table IV) (also a sand-filled lysimeter). In addition, Sr-90 was found in significant quantities in the moisture cups of ANL-E Lysimeters 4 and 5 and in the leachate of all ANL-E and ORNL lysimeters during the April sampling. The concentration of Sr-90 in the ORNL lysimeter leachate was almost two orders of magnitude higher than in the ANL-E samples, as predicted by the soil moisture data analysis in the previous section.

TABLE III

Results of Beta and Gamma Analysis of ANL-E Soil Moisture and Leachate Samples Obtained in 1986

Sample Identification	Concentration[a] (pCi/L)					
	Co-60		Cs-137		Sr-90	
	April	June	April	June	April	June
Composite[b]	--	<5	--	<5	--	<1
1-1[c]	--	<5	--	<5	--	<1
1-3	<5	<5	<5	<5	<1	<1
2-3	<5	<5	<5	<5	<1	<1
3-3	11 ± 7	13 ± 7	<5	<5	1.1 ± 1.0	11.3 ± 1.4
4-3	<5	<5	<5	<5	2.7 ± 1.8	<1
5-1	--	<5	--	<5	--	349.6 ± 11.3
5-3	<5	<5	<5	<5	55.6 ± 3.1	127.6 ± 6.7
1[d]	<5	<1	<5	<1	0.5 ± 0.3	<1
2	<5	<1	<5	<1	0.5 ± 0.2	<1
3	<5	<1	<5	<1	0.4 ± 0.1	<1
4	<5	<1	<5	<1	0.6 ± 0.3	<1
5	<5	<1	5.4 ± 1.1	<1	1.0 ± 0.4	5.8 ± 0.3

a. Concentration ± 2 sigma.

b. Composite of water from moisture cups 2-5, 3-5 and 4-5.

c. Moisture cup identification number.

d. Leachate collector identification number.

TABLE IV

Results of Beta and Gamma Analysis of ORNL Soil Moisture and Leachate Samples Obtained in 1986

Sample Identification	Concentration[a] (pCi/L)							
	Co-60		Cs-137		Sb-125		Sr-90	
	April	June	April	June	April	June	April	June
Composite[b]	--	<19	--	<19	--	<27	--	8.6 ± 10.2
1-3[c]	<16	<27	<14	<22	<27	<27	2.7 ± 4.9	0.5 ± 7.6
2-3	<16	<81	<14	<54	<27	<27	7.0 ± 13.5	15.4 ± 18.1
3-3	<16	<54	<11	<54	<27	<27	5.9 ± 12.9	32.4 ± 18.9
4-3	<14	<27	<11	<16	<27	<27	<11	<5
5-3	<14	89.2 ± 32.4	<14	<3	351 ± 5	540 ± 81	6.2 ± 18.1	17.6 ± 12.2
1[d]	<2	<5	<1	<5	<3	<3	62.2 ± 8.1	9.2 ± 3.5
2	<1	<8	<1	<6	<3	<0.3	27.0 ± 5.4	2.4 ± 2.7
3	<3	<8	<2	<6	<5	<0.3	4.9 ± 2.7	1.1 ± 2.7
4	<2	<8	<2	<5	<3	<0.3	54.1 ± 8.1	11.6 ± 4.6
5	<2	<8	<1	<5	<5	<37	45.9 ± 8.1	2.4 ± 2.9

a. Concentration ± 2 sigma.

b. Composite of water from moisture cups 1-5, 2-5, 3-5, and 4-5.

c. Moisture cup identification number.

d. Leachate collector identification number.

Analysis of water samples obtained in June 1986 showed that Co-60 still persisted in ANL-E moisture cup 3-3 and the Sb-125 had increased substantially in ORNL moisture cup 5-3. The origin of the Sb-125 is not known, but the radionuclide is assumed to come from the waste forms. The original analysis of the radionuclide content of the prefilters from which this resin was taken identified Sb-125 in quantities of 0.1% of the total radionuclide content, although later resin analysis did not find any. Co-60 also was found for the first time in ORNL moisture cup 5-3 in June. Also, Sr-90 was detected in moisture cups 3-3 and 5-3 at ORNL and in two additional cups (3-3 and 5-1) at ANL-E, while there was none detected in the June sampling of ANL-E moisture cup 4-3. The concentration of Sr-90 in ANL-E moisture cup 5-3 was more than twice that found in the April sample. ANL-E moisture cup 5-1 is located directly below 5-3, and the water from this moisture cup was analyzed for the first time in June in an attempt to detect movement of Sr-90 through the silica sand profile. The concentration of Sr-90 in ANL-E moisture cup 5-1 was almost three times that of 5-3. In general, occurrence of Sr-90 in the leachate collector samples at both sites was down significantly from the April sampling, with measurable amounts being found only in ANL-E Lysimeter 5 and ORNL Lysimeters 1 and 4.

Occurrence of radionuclides in water samples from both the soil- and sand-filled lysimeters after such a short period of time (months rather than years) was unexpected. While Sr-90 is known to be soluble in soil solution and does move through the soil column almost unhindered by the soil matrix, it appears that leaching and movement of the radionuclides is occurring at a more accelerated rate in the soil than was thought possible. The appearance of Sr-90 indicates that small quantities were readily leached from the waste forms, possibly due to abrasion of waste form surfaces during installation into the lysimeters. The higher initial concentrations of Sr-90 in the ORNL leachate collectors could reflect the greater pore volume of water that has passed through these lysimeters. Comparing Sr-90 data from the April and June samplings, it appears that Sr-90 moved through the lysimeters as an initial slug which, in the case of ORNL, has been washed out of the soil into the leachate collectors or, as at ANL-E, is in the process of being flushed into the collectors. Data from ANL-E Lysimeter 5 would support this hypothesis, since it appears that a plume of Sr-90 movement has been detected in this lysimeter, with the trailing edge showing up in the area of moisture cup 5-3, the bulk near 5-1, and a leading edge moving into the leachate collector. Although Sr-90 has been detected, the total quantity leached is only a small fraction of that available in the waste forms (<0.0001%).

CONCLUSIONS

Lysimeter operations at ANL-E and ORNL have been successful the first year. Despite some operational problems, the environmental detectors and DAS have provided useful data for portraying the year-long environmental conditions experienced by each lysimeter. The radionuclide monitoring/liquid collection portion of the experiment is functioning better than anticipated. It was noted that Sr-90 is leaching from the waste forms into the soil of the lysimeters much sooner than expected. This was especially apparent at ORNL, where Sr-90 was found in all leachate collector samples. It was seen that the characteristics of the ORNL environment and the Savannah River Laboratory soil resulted in more movement of Sr-90 at ORNL than ANL-E by allowing a greater quantity of precipitation to infiltrate and percolate through the soil. ANL-E, however, has had a persistent occurrence of Co-60 in water obtained from one moisture cup. Since cobalt is not "fixed" by soil (like cesium), its movement is expected to continue.

REFERENCES

1. 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste," Code of Federal Regulations, Office of Federal Register.
2. R. C. SCHMITT and H. W. RENO, "Program Plan of the EPICOR and Waste Research and Disposition Program of the Technical Support Branch," EGG-TMI-0521, EG&G Idaho, Inc. (revised December 1983).
3. J. W. McCONNELL, Jr., "EPICOR-II Resin/Liner Research Plan," EGG-TMI-6198, EG&G Idaho, Inc. (1983).
4. R. M. NEILSON and J. W. McCONNELL, Jr., "Solidification of EPICOR-II Resin Waste Forms," GEND-INF-055, EG&G Idaho, Inc. (1984).
5. R. D. ROGERS, J. W. McCONNELL, Jr., E. C. DAVIS, M. W. FINDLEY, "Field Testing of Waste Forms Containing EPICOR-II Ion Exchange Resins Using Lysimeters," NUREG/CR-4498, U.S. Nuclear Regulatory Commission (1986).
6. H. W. RENO, J. W. McCONNELL, Jr., R. C. SCHMITT, "EPICOR-II Research and Disposition Program: FY-1983 Annual Report," EGG-2287, EG&G Idaho, Inc. (1984).
7. J. W. McCONNELL, Jr., H. W. RENO, A. L. AYERS, JR., R. C. SCHMITT, "EPICOR and Waste Research and Disposition Program: FY-1984 Annual Report," EGG-2353, EG&G Idaho, Inc. (1985).
8. R. D. ROGERS, J. W. McCONNELL, Jr., M. W. FINDLEY, E. C. DAVIS, "Lysimeter Data from EPICOR-II Waste Forms--Fiscal Year 1986," EGG-TMI-7417, EG&G Idaho, Inc. (1986).

NOTICE