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PATENT CLEARED

FIELD TESTING OF WASTE FORMS USING LYSIMETERS

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ABSTRACT

The Low-Level Waste Data Base Development--EPICOR-II Resin/Liner Investigation Program funded by the U.S. Nuclear Regulatory Commission (NRC) is obtaining information on performance of radioactive waste in a disposal environment. This paper presents a description of the field testing and gives preliminary findings. Solidified 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 martices of cement and Dow polymer. Emphasis is placed on evaluating the requirements of 10 CFR 61 "Licensing Requirements for Land Disposal of Radioactive Waste" by obtaining data on performance of waste in a disposal environment using lysimeter arrays at Oak Ridge National Laboratory (ORNL) and Argonne National Laboratory in Illinois (ANL).

INTRODUCTION

Lysimeters are used in field testing because when properly designed they can be used to isolate and manipulate soil systems under actual environmental conditions. Lysimeters have been used for many years for field tests to determine the amount of leaching of various elements from soil due to percolation of water. In fact, the word lysimeter comes from the two Greek roots lysi, meaning loosening, and meters, to measure.

The shape and size of lysimeters are determined by the imagination and experimental requirements of the investigator. They can range from small volumes of field soil isolated from surrounding areas by impervious dividers to concrete or metal tanks. If tanks are used they can consist of an upper and lower compartment and are placed into the field so that the open end is level with the soil surface. The upper compartment serves as a containment for soil while the lower level serves as a collection and storage compartment for water which has pasted through the overlying soil. Those lysimeters without bottom compartments will have some porous material in the bottom such as gravel to separate percolating water from soil.

Lysimeters lend themselves to instrumentation. They can be placed on top of weighing devices so that water content can be determined gravimetrically and they can have various water sampling, temperature, and moisture sensing devices implanted as they are filled with soil. The amount of sophistication depends on experimental objectives.

Lysimeters are the obvious tool to use in the field testing of solidified waste forms. In the study reported here, the waste forms are composed of solidified ion exchange resin materials from EPICOR-II prefilters used in the cleanup of the Three Mile Island Nuclear Power Station. They are being subjected to long term testing 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 has been 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.

METHODS AND MATERIALS

Waste forms used in the experiment contain a mixture of synthetic organic ion exchange resins or the organic exchange resins mixed with an inorganic zeolite. Solidification agents which were used to produce the 4.8-by-7.6-cm cylindrical waste forms (Figure 1) examined in the study were Portland Type I-II cement and Dow vinyl ester-syrene (VES).[2] Seven of these waste forms were stacked end-to-end in each lysimeter[3] (5 lysimeters at ORNL and 5 at ANL-E)[4,5] to provide a 1-L volume. The inventory and approximate nuclide content of waste forms used in each lysimeter is found in Table 1.

Lysimeters used in this study were designed to be self-contained units which will be disposed 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 a lower compartment, which collects leachate (Figure 2). Four lysimeters at each site are filled with soil, while a fifth (used as a control) is filled with inert silica oxide sand. Figure 3 shows the placement of the lysimeters. Instrumentation within each lysimeter includes 5 Teflon porous cup soil-water samplers and 3 soil moisture/temperature probes[3] (Figure 2). 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[3].

Each month, data stored on a cassette tape are retrieved from the DAS and translated into an IBM PC compatible disk file. These data are reduced using LOTUS[R] for tabular and graphic displays. At least quarterly, water is drawn from the porous cup soil-water samplers and the lysimeter leachate collection compartment and selected samples are analyzed for beta and gamma producing nuclides. Soil moisture/temperature at three elevations in each lysimeter, along with a complete weather history, are recorded on a continuing basis by the DAS. Highlights of the results of testing are presented in this paper. A more detailed presentation of the data is given in Reference 8.

RESULTS AND DISCUSSION

Results obtained from 17 months of field testing are presented in this section. There was a period from late August 1985 until November 1985 when the ORNL DAS was inoperable, due to the necessity of returning the system to the manufacturer for repairs.

Instrument Operation and Data

Wind speed, air temperature, relative humidity, and rainfall, are recorded over a 12-month period by the DAS systems for the ANL and ORNL sites. Air temperature data from ANL (Figure 4) show that there were days of freezing temperatures from mid-November 1985 until mid-March 1986, while there were very few days with air temperatures of 0°C at ORNL (Figure 5). Rainfall data from the ORNL site appeared to be higher than normal. This trend became apparent during December 1985, and indications were than the Weather Measure tipping bucket rain gauge supplied with the DAS system was not capable of accurately responding to period of intense rainfall. In June 1986, that rain gauge was replaced with a Climatronics tipping bucket gauge designed for episodic high-intensity rainfall. Data from this gauge appear to be accurate most of the time; however, the rainfall data recorded by the DAS contain occasional, erroneously high data points. The malfunctions have not resulted 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 2) were used to calculate the total quantities of precipitation received by each site.

Temperature probes located in all ten lysimeters at the depth of the waste forms (77.0 cm) indicate that at no time were the waste forms exposed to freezing temperatures (Figures 6 and 7). 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) fluctuate more than the intermediate (elevation 77.9 cm) or bottom (elevation 28.8 cm) soils. It is also 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 lysimeter ANL-3 in January 1986 and in ANL-4 by 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 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 system. These probes were replaced with new ones, and recent data from the replacements shows that they are functioning normally.

The bottom temperature probes in ORNL-3 and -5 has consistently indicated high temperatures as compared to temperatures measured by probes in those and nearby lysimeters. Because the abnormal readings began close to the time of lysimeter installation, it is possible that probes or wiring were damaged during installation.

Moisture probes at the two sites show that two (ORNL) and three (ANL) months were required after the lysimeters were filled with soil for the soil to reach saturation (Figures 8 and 9). As a precaution, the accuracy of the probes in the soil-filled lysimeters was determined by comparing their data against the gravimetric water content of soil cores retrieved from all four ORNL lysimeters and one ANL lysimeter. From those comparisons it was apparent that the probes are over-estimating the soil moisture content.

Corrective action consisted of recalculation of the polynomial equation which transforms probe input into percent moisture using laboratory and field soil gravimetric measurements.

Lysimeter Water Balance

During a 12-month period, the ANL site had 93.5 cm of precipitation while ORNL received 99.0 cm, well below the normal of 134 cm. Using these values and the area of exposed lysimeter (6489.5 cm^2), it was calculated that the ANL and ORNL lysimeters received 607 L and 643 L of water, respectively. Total water retrieved from the leachate collectors of each lysimeter during the period from July 1985 to July 1986 is shown in Table 2. On the average, the leachate collectors of the soil-filled ANL lysimeters contained 128.7 $\pm$ 22.6 L; those at ORNL, 441.7 $\pm$ 20.9 L. The leachate collectors of the sand-filled lysimeters at ANL and ORNL contained 337.9 L and 528.0 L, respectively.

The two sites received comparable volumes of precipitation, however water moved through the lysimeters at the sites in unequal amounts. Because vegetation is removed from each lysimeter, evapotranspiration did not factor into water loss. It appears that the differences in water movement were due to soil texture and weather conditions. Soil used at ANL is heavier (contains more fine material such as silts and swelling clay) than the soil used at ORNL.[3] Therefore, infiltration and percolation of water through the ANL soil would be reduced in comparing the sand-filled control lysimeters at the two sites. At ANL, 55% of the volume of precipitation passed through the lysimeter versus 82% for ORNL.

Based on the amount of water retrieved from the lysimeters, the ANL soil-filled lysimeters had 0.18 pore volumes of water pass through them while 0.62 pore volumes passed through similar ORNL lysimeters. Pore volume for the control lysimeters (sand filled) were 0.57 for ANL and 0.94 at ORNL. Theoretically then, 18% of the water held in the soil pore space of the ANL lysimeters was replaced during the first 12 months, while 62% was replaced in the ORNL soil lysimeters. Similarly, 57% and 94% was replaced in the ANL and ORNL control lysimeters, respectively. Therefore, if nuclides 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 nuclide is water soluble and (b) the soil column does not interfere with nuclide movement.] It is noted that ^{60}Co and ^{90}Sr move through soils freely while ^{125}Sb and ^{137}Cs are readily retained by most soils. The inert sand used in the number 5 lysimeters was selected because it does not interfere with the movement of the nuclides under investigation.

Radionuclide Analysis

Water samples have been collected from the leachate collectors and moisture cup 3 from each lysimeter on five occasions since experiment initiation. The first two times, water samples were analyzed only for gamma-producing nuclides. The last three water samples, taken at both sites in April, June, and October 1986, were analyzed for both gamma-producing nuclides and the beta-producing nuclide ^{90}Sr.

During June 1986, in addition to obtaining water samples from leachate collectors and moisture cup 3, water samples were taken from moisture cup 5 (the one nearest the soil surface) of each soil lysimeter. Those samples were then combined for use as a composite sample. Because moisture cup 5 is located above the waste forms, the composite water sample serves as a control to detect nuclides which might originate from sources other than the waste forms. Radionuclides were not found above background quantities in those samples and sampling of those cups was not performed after this data.

Gamma-producing nuclides were not found in the first two samplings. However, in April 1986, ^{60}Co was discovered in water samples from the moisture cup of ANL 3 (Table 3); ^{137}Cs was found in the leachate of ANL 5 (the sand-filled lysimeter); and ^{125}Sb was found in the moisture cup of ORNL 5 (Table 4) (also a sand filled lysimeter). In addition, ^{90}Sr was found in significant quantities in the moisture cups of ANL 4 and 5 and in the leachate of all ANL and ORNL lysimeters during the April sampling. The concentration of ^{90}Sr in the ORNL lysimeter leachate was almost two orders of magnitude higher than the ANL samples.

Analysis of water samples obtained in June 1986 (Tables 3 and 4) showed that ^{60}Co still persisted in moisture cup ANL 3-3, with a substantial increase in ^{125}Sb in ORNL moisture cup 5-3. The origin of ^{125}Sb is not know, but it is assumed to be the waste forms. Original evaluation of radionuclide content of the prefilters from which this resin was taken identified ^{125}Sb in quantities of 0.1% of the total nuclide content, although it was absent in a subsequent resin analysis. Cobalt-60 was also found for the first time in ORNL 5-3 in June. Also, ^{90}Sr was detected in moisture cups at ORNL (ORNL 3-3 and 5-3) and in two additional cups at ANL (ANL 3-3 and 5-1), while there was none detected in this sampling of ANL 4-3. The concentration of ^{90}Sr in ANL 5-3 was more than double that found in the April sample. ANL 5-1 is the moisture cup 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 ^{90}Sr through the silica sand profile. The concentration of ^{90}Sr in ANL 5-1 was almost three times that of 5-3. In general, occurrence of ^{90}Sr in the leachate sample at both sites was down sharply from the April 1986 sampling, with measurable amounts being found only in ANL 5 and ORNL 1 and 4.

Results from the analyses of the October 1986 water samples (Tables 3 and 4) showed that ^{60}Co, which was found during the previous two sampling periods in ANL 3-3, was absent. Cesium-137 was discovered in the water samples from ANL 2-3. Cobalt-60 was not found in ORNL 5-3 this sampling period, however, ^{125}Sb not only persisted in ORNL 5-3 but was detected in the leachate collection tank of lysimeter 5 (the sand-filled lysimeters). At ANL, ^{90}Sr was detected only in the moisture cups of lysimeter 5 (ANL 5-1, 5-3, 5-4) and was absent from that lysimeter's leachate collectors. On the other hand, ^{90}Sr was found in all the moisture cups and four of the five leachate collectors of the ORNL lysimeters. There was no ^{90}Sr in ORNL-4 at this sampling.

Occurrence of nuclides in the water samples from both the soil and inert sand lysimeters in such a short period of time (months rather than years) was unexpected. While ^{90}Sr 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 nuclides is occurring at a more accelerated rate in the soil than was thought possible. Appearance of ^{90}Sr in the April leachate of all lysimeters at both sites and at ORNL in October indicates that small quantities were readily leached from all the waste forms irrespective of their formulation (cement or VES) or initial ^{90}Sr concentration. The higher initial concentration at ORNL could reflect the greater pore volume of water that has passed through these lysimeters. This could also be the case for the occurrence in October 1986. When comparing ^{90}Sr data from the April and June 1986 samplings, it appears that ^{90}Sr moved through the lysimeters as an initial slug which, in the case of ORNL, has been washed out or, as at ANL-E, is in the process of being flushed. Data from ANL 5 would support this hypothesis, since it appears that a plume of ^{90}Sr movement has been detected in this lysimeter, with the trailing edge showing up in the area of ANL 5-3, the bulk near ANL 5-1, and a leading edge moving into the leachate collector. The October ORNL data would indicate that there is a source of mobile ^{90}Sr and continued leaching of this element is to be expected. These data are also supported by the October ANL data where it is seen that ^{90}Sr has moved from the location of the waste form (cup 1) to near the bottom of the sand profile (cup 3). Though ^{90}Sr has been detected, the total quantity leached is only a small fraction of that available in the waste forms (Table 1).

Finally, because it is apparent that the soil in the lysimeters has subsided (very evident at ANL), it was decided to determine if the movement had caused a shift in the position of the waste forms. This was accomplished by lowering a radiation-detecting probe down the access tube which leads into the leachate holding tank. Readings were taken every 15.2 cm in all lysimeters, and radiation intensity with depth was recorded. Readings of the soil lysimeters were then compared with readings from the sand-filled controls. At ORNL, the intensity of radiation readings for each lysimeter approximated the known depth of the waste forms (Table 5). However, at ANL, some settling has occurred; radiation readings from the waste forms in the soil-filled lysimeters (1-4) were still high at the 182.9-cm depth, while the activity in the inert control had moderated by that depth, indicating a downward movement of the waste forms of about 7.5 cm in the soil-filled lysimeters. There is no evidence that the movement has impacted the experiment except for minor damage to some moisture/temperature probes.

REFERENCES

1. 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste," Code of Federal Regulations, Office of Federal Register.

2. R. M. Neilson and J. W. McConnell, Jr., Solidification of EPICOR-II Resin Waste Forms, GEND-INF-055, EG&G Idaho, Inc., August 1984.

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5. J. W. McConnell, Jr., EPICOR-II Resin/Liner Research Plan, EG&G-TMI-6198, March 1983.

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TABLE 1. LYSIMETER WASTE FOR INVENTORY

Lysimeter	Waste Form ID	Contact Gamma Dose[a] (R/h)	Lysimeter	Waste Form ID	Contact Gamma Dose[a] (R/h)
ANL 1[b]	C1A-11[d]	2.1	ORNL 1[c]	C1-13	2.2
	C1A-12	2.1		C1-16	2.2
	C1-22	2.1		C1A-13	2.1
	C1-12	2.2		C1A-14	2.2
	C1-15	2.2		C1A-15	2.1
	C1-20	2.1		C1A-16	2.1
	C1-21	2.2		C1A-17	2.1
ANL 2	C2B-17	10.0	ORNL 2	C2B-8	10.0
	C2B-21	10.0		C2B-9	10.0
	C2B-15	10.0		C2B-10	10.0
	C2B-18	10.0		C2B-11	10.0
	C2B-19	10.0		C2B-12	10.0
	C2B-20	10.0		C2B-13	10.0
	C2B-16	10.0		C2B-14	10.0
ANL 3	D1A-3[e]	4.5	ORNL 3	D1A-8	4.5
	D1A-13	4.0		D1A-9	4.5
	D1A-4	4.5		D1A-10	4.5
	D1A-5	4.5		D1A-11	4.5
	D1A-7	4.5		D1A-12	4.5
	D1A-14	4.0		D1A-15	4.0
	D1A-6	4.5		D1A-16	4.0
ANL-4	D2-5	18.0	ORNL 4	D2-7	18.0
	D2-2	18.5		D2-8	18.0
	D2-3	18.0		D2-9	18.0
	D2-6	18.0		D2-10	18.0
	D2-13	17.0		D2-11	18.5
	D2-14	17.0		D2-15	17.0
	D2-4	18.0		D2-16	17.0

TABLE 1. (continued)

Lysimeter	Waste Form ID	Contact Gamma Dose[a] (R/h)	Lysimeter	Waste Form ID	Contact Gamma Dose[a] (R/h)
ANL 5	C1A-19	2.1	ORNL 5	C2B-22	10.0
	C1A-21	2.1		C2B-23	10.0
	C1A-22	2.1		C2B-24	10.0
	C1A-18	2.1		C2B-25	10.0
	C1-14	2.2		C2B-26	10.0
	C1-17	2.2		C2B-27	10.0
	C1-18	2.1		C2B-28	10.0

a. Waste forms have the following average curie contents:

	^{134}Cs	^{137}Cs	^{90}Sr	Total
D1 and D1A	4.38×10^{-3}	66.22×10^{-3}	3.92×10^{-3}	74.52×10^{-3}
D2 and D2A	18.22×10^{-3}	275.45×10^{-3}	0.64×10^{-3}	294.31×10^{-3}
C1 and C1A	2.95×10^{-3}	44.58×10^{-3}	2.64×10^{-3}	50.17×10^{-3}
C2A and C2B	13.53×10^{-3}	204.59×10^{-3}	0.47×10^{-3}	218.59×10^{-3}

b. Argonne National Laboratory, Argonne, IL.

c. Oak Ridge National Laboratory, Oak Ridge, TN.

d. Portland Type II Cement.

e. Dow vinyl ester-syrene.

TABLE 2. YEARLY PRECIPITATION AT ANL AND ORNL AS MEASURED BY BACK-UP INSTRUMENTATION--JULY 1985 THROUGH JULY 1986

	Precipitation (cm)	
Month	ANL	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	7.2
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 lysimeter experiment initiated in July.

b. ANL-E lysimeter experiment initiated in August.

c. Months leachate was retrieved for analyses.

TABLE 5. RADIATION INTENSITY WITH DEPTH IN EPICOR-II FIELD LYSIMETERS

Depth from Soil Surface (cm)	Radiation Intensity (mR/h)									
	ANL-E Lysimeter Number					ORNL Lysimeter Number				
	1	2	3	4	5	1	2	3	4	5
15.2	--	--	--	--	--	--	--	--	--	--
30.5	--	--	--	--	--	--	--	--	--	--
45.7	--	--	--	--	--	--	0.02	0.04	0.01	0.04
61.0	--	--	--	0.3	1.0	0.005	0.04	0.18	0.29	0.21
76.2	--	--	--	1.0	3.0	0.16	0.24	0.65	1.6	1.4
91.4	0.4	--	0.5	1.3	6.0	0.81	1.0	2.7	1.6	1.4
106.7	0.7	2.0	2.5	7.0	10.0	2.9	3.5	13.7	25.8	20.8
121.9[b]	3.5	18.0	5.0	35.0	12.0	6.1	11.2	29.0	52.4	40.3
137.2	6.0	28.0	20.0	43.0	12.0	11.2	16.1	39.5	70.2	48.3
152.4	7.5	39.0	18.0	67.0	12.0	11.2	17.7	40.3	70.9	36.3
167.6[b]	7.0	32.0	17.0	65.0	8.0	8.1	12.9	30.6	54.8	20.9
182.9	3.6	21.0	10.0	38.0	6.0	2.4	4.6	14.5	25.8	5.9
198.1	1.8	10.0	2.5	18.0	5.0	0.73	1.5	3.7	9.7	1.1
213.4	--	--	--	--	--	0.16	0.31	0.89	1.4	0.16

a. Readings were not above background.

b. Location of waste form is indicated by sets of bars.

LIST OF FIGURES