DP-MS--83-87

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SPECIAL WASTEFORM LYSIMETER PROGRAM
AT THE SAVANNAH RIVER LABORATORY

by

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A paper proposed for presentation at the
DOE Low-Level Waste Management Program
Fifth Annual Participants Information Meeting
Denver, Colorado
August 30 - September 1, 1983
and for publication in the Proceedings of the Meeting

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ABSTRACT

The Special Wasteform Lysimeter project at SRL is designed to measure performance of typical production-line, low-level, solid wasteforms produced at power reactors and emplaced in a humid SLB site. The use of lysimeters permits direct measurement of migration of radioactivity from these wasteforms to provide a technical basis for evaluating how well these forms will perform in an actual burial trench, and additionally allows comparison with unencapsulated defense waste. Cement and polymer wasteforms were placed into 10 lysimeters in March 1982. By March, 1983, ^{60}Co from both types of wasteforms had been detected in the lysimeter sumps. ^{134}Cs , ^{137}Cs , and ^{60}Co were found in porous cup samplers located directly below the wasteforms. Measurements in mid-summer 1983 showed that ^{60}Co levels were 10 to 100 times greater in lysimeters containing the cement forms than in those containing the polymer.

INTRODUCTION

Lysimeter studies at the Savannah River Plant (SRP) are designed to measure radionuclide migration from well-characterized sources under field conditions. The Savannah River Laboratory (SRL) has conducted a variety of lysimeter tests since 1973, in ongoing studies with typical defense low-level wasteforms. In 1981, the SRL lysimeter program was expanded to include special wasteforms typical of those produced at commercial power reactors. The SRL special wasteform lysimeter studies are funded by the DOE Low-Level Waste Management Program (LLWMP).

The purpose of this LLWMP task is to measure radionuclide leaching and migration from encapsulated low-level solid wastes produced at power reactors and emplaced in a humid, shallow-land burial site. The use of lysimeters will permit direct measurement of migration

* The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

of radioactivity from these wasteforms and will provide a technical basis for evaluating how the forms will behave in an actual burial trench. In addition, leaching and migration results can be compared with those from unencapsulated defense wastes, to measure the benefits of encapsulation.

STATUS AND ACCOMPLISHMENTS

The SRL Special Waste Form Lysimeter Program was initiated in April 1981, and construction of 14 lysimeters was completed in October 1981. In late February and early March of 1982, the experiment was started when wasteforms were buried in 10 of the lysimeters. One other lysimeter was filled with soil but no waste, to serve as a control.

The wasteforms were acquired from power reactor sites by Brookhaven National Laboratory (BNL) under subcontract with SRL. BNL was also responsible for establishing source terms and performing full-scale leach tests for each type of wasteform. An initial report (BNL 51613) was published in September, 1982.¹ A final report on the BNL work will be published in Sept, 1983. SRL lysimeter design, loading, operating, and sampling procedures were described at the Fourth Annual LLWMP Participants Meeting.²

Percolate water first appeared in the lysimeter sumps in May, 1982, and the lysimeters have operated properly since then. ⁶⁰Co was first detected in lysimeter sumps in ~~March~~^{Feb}, 1983, by using the preconcentration method described last year.² Subsequent samples from porous cups located directly beneath the wasteforms contained ¹³⁴Cs and ¹³⁷Cs as well as ⁶⁰Co. The information collected so far has allowed preliminary calculation of ⁶⁰Co retardation in soil below the wasteform and estimation of the soil k_d for ⁶⁰Co.

RESULTS

Results from almost ⁷ months of sampling after ⁶⁰Co first appeared in the lysimeter sumps are summarized in Tables 1-3. Table 1 compares ⁶⁰Co measurements for the cement and polymer forms. All wasteforms made from Portland Type III cement or polymer contained typical waste from a single BWR, hence radionuclide concentrations

and isotopic distributions are very similar. The two masonry cement forms were supplied from a PWR.

In all cases the fraction of ^{60}Co migrating from the wasteform, through about 1 *m* of soil beneath the wasteform, and into the lysimeter sump is very small (Table 1, Col 3). Releases from the polymer forms were generally 10 to 100X below those from the comparable Type III cement forms. Fractional releases from the masonry cement are about 10X higher than from the Type III cement. / m

The porous cup samplers positioned directly under the wastes allowed calculation of the "decontamination factor" provided by retention of ^{60}Co on the soil column between the wasteform and sump. Except for one outlying point for the cement + evaporator bottoms + resin, these DF's averaged about 10,000.

Tables 2 and 3 show ^{134}Cs and ^{137}Cs results. In no case has Cs been detected in lysimeter sumps. This is an expected result of the generally high ion exchange capacity clay-containing soils have for Cs. The absence of Cs from any of the porous cup samplers below the polymer wasteforms is consistent with the ^{60}Co data showing better retention by the polymer and is also consistent with the generally poor retention of Cs in cements.

TRANSPORT EQUATION

In addition to estimating DF's through the soil, the porous cup and sump sampling results were compared to a simple one-dimensional equation for radionuclide movement through a soil column. The time of appearance and the concentration of a given radionuclide in the water collected at the bottom of the column will depend primarily on four factors: the leach rate, the water velocity, the distribution coefficient of the radionuclide between the soil and soil water, and the dispersion coefficient. The leach rate and rainfall combine to set the concentration (C_0) of the radionuclide in the soil water just below the waste. This was measured directly by the porous cups.

Radionuclide movement downward through the lysimeter was modeled with a one-dimensional convective-dispersive solute transport equation:

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial X} = D \frac{\partial^2 C}{\partial X^2} - KC \quad (1)$$

Boundary conditions appropriate to the lysimeter system are:

$$C = C_0 e^{-\gamma t}, \text{ at } X = 0$$

$$C = 0, \text{ at } t = 0 \text{ and } X > 0 \quad (2)$$

$$\frac{\partial C}{\partial X} \rightarrow 0, \text{ as } X \rightarrow +\infty$$

The solution to this differential equation is given by Bear ⁴³ and Cleary ⁵⁷ as:

$$C(X, t) = \frac{C_0 e^{-\gamma t}}{2} \left[\exp \left(\frac{Xv}{2D} - \frac{X}{\sqrt{D}} \sqrt{A} \right) \cdot \operatorname{erfc} \left(\frac{X}{2\sqrt{Dt}} - \sqrt{At} \right) + \exp \left(\frac{Xv}{2D} + \frac{X}{\sqrt{D}} \sqrt{A} \right) \cdot \operatorname{erfc} \left(\frac{X}{2\sqrt{Dt}} + \sqrt{At} \right) \right] \quad (3)$$

Notation used in the above equations is summarized below:

A = Collected terms that account for source diminution:

$$A = \frac{v^2}{4D} + K - Y$$

X = Distance in vertical direction (L).

C (X,t) = Concentration of radionuclide at X and t (activity/L³).

C₀ = Initial concentration of radionuclide (activity/L³).

D = Dispersion coefficient (L²/t)

V = Velocity of radionuclide in groundwater (L/t). This is related to the groundwater velocity by

$$V = V_w \left(\frac{1}{1 + K_d R} \right)$$

V_w = Groundwater velocity (L/t)

K_d = Distribution coefficient of soil for radionuclide (L³/mass).

R = Retardation factor = [ρ/η] (Mass/L³)

ρ = Soil bulk density (mass/L³). Assumed = 1.5 g/cm³

η = Effective porosity (dimensionless). Assumed = 0.25

t = Time

K = Radioactive decay constant for radionuclide (1/t)

Y = Source depletion constant (may be a combination of leaching, radioactive decay, etc.) (1/t).

This transport equation is strictly applicable only to situations in which the groundwater velocity is constant. A constant flow velocity would be found only under very simplified saturated flow conditions. In unsaturated flow the hydraulic conductivity (and therefore the flow velocity) is a function of the soil moisture content, which varies in time and space. Therefore, for the equation to be applied to the unsaturated lysimeters, the simplifying assumption is made that the soil moisture content (and therefore the flow velocity) remains constant. Use of the one-dimensional transport equation for modeling soil columns is well established.^{4,5}

For Figure 1, a vertical water velocity of 14 ft/yr was assumed. This is about twice the vertical velocity of water in the unsaturated zone of the SRP burial ground, where there is opportunity for run-off to streams. The lysimeter design does not permit run-off so this water is assumed to percolate downward. The dispersion coefficient (D) is primarily dependent on soil particle size, shape, and uniformity. Because the value of D is not known, the relation $D = \alpha V$ was used, where V is the radionuclide velocity, and α is the dispersivity. The value for dispersivity $\alpha = 1$ was used, based on estimates by Gupta and Greenkorn.⁵

The experimental results shown in Figure 1 are consistent with an effective K_d for ^{60}Co of about 15. A similar calculation assuming a K_d of 100 for ^{137}Cs indicates that about 15 years will be required for detectable amounts of this isotope to reach the lysimeter sump.

FUTURE WORK

During the next year emphasis will be placed on detecting ^{90}Sr in the porous cup samples and in the lysimeter effluent. A method for preconcentrating ^{90}Sr for more sensitive analysis was recently developed for this purpose. K_d for Sr in SRP soils and groundwaters has been estimated to be 15 to 25 so appearance of ^{90}Sr during the next year is likely.

REFERENCES

1. R. M. Neilson, Jr., P. D. Kalb and P. Colombo. Lysimeter Study of Commercial Reactor Waste Forms: Waste Form Acquisition, Characterization and Full-Scale Leaching. BNL 51613, September, 1982 (Revised Feb, 1983).
2. R. H. Hawkins and J. A. Stone. "Special Waste Form Lysimeters." p. 367 in Proceedings of the Fourth Annual Participants' Information Meeting, DOE Low-Level Waste Management Program. ORNL/AFW-82/18, October, 1982.
3. J. Bear. Dynamics of Fluids in Porous Media, American Elsevier Publishing Co., New York, NY (1972).
4. J. N. Lucas. "The Transport of a Radioactive Salt Through a Semi-Infinite Column of Porous Medium: A Physical Model." Water Resources Research, 16 (2) 387-390 (1980).
5. S. P. Gupta and R. A. Green^Khorn. "Determination of Dispersion and Nonlinear Absorption Parameter for Flow in Porous Media." Water Resources Research, 10 (4) 839-846 (1974).

TABLE 1
⁶⁰CO MIGRATION IN THE SWF LYSIMETERS
 (JAN-JUNE, 1983)

Matrix	Waste- form (mCi)	Porous Cup (μ Ci/l)	Sump (nCi)	Fractional Release (Sump $\div$ Wasteform)	Soil Column DF (Cup x Vol $\div$ Sump)
Type III Cement + Bottoms	130	1.4	101	8×10^{-7}	14,000
Type III Cement + Bottoms	139	1.2	208	15×10^{-7}	6,000
Polymer + Bottoms	196	0.04	1.6	8×10^{-9}	25,000
Polymer + Bottoms	196	0.04	16.7	8×10^{-8}	2,000
Type III Cement + Bottoms + Resin	440	0.44	249	6×10^{-7}	2,000
Type III Cement + Bottoms + Resin	402	2.3	9.8	2×10^{-8}	234,000
Polymer + Bottoms + Resin	466	0.17	17.7	4×10^{-8}	9,000
Masonry Cement + Boric Acid	0.9	---	3.2	4×10^{-6}	---
Masonry Cement + Boric Acid	0.9	NS	5.4	6×10^{-6}	---

TABLE 2
¹³⁷CS MIGRATION IN THE SWF LYSIMETERS
 (JAN-JUNE, 1983)

<u>Matrix</u>	<u>Waste- form (mCi)</u>	<u>Porous Cup (μCi/l)</u>
Type III Cement + Bottoms	41	2.2
Type III Cement + Bottoms	43	9.4
Polymer + Bottoms	32	---
Polymer + Bottoms	32	---
Type III Cement + Bottoms + Resin	92	0.8
Type III Cement + Bottoms + Resin	84	9.2
Polymer + Bottoms + Resin	77	---
Masonry Cement + Boric Acid	0.4	0.1
Masonry Cement + Boric Acid	0.4	---

TABLE 3
¹³⁴CS MIGRATION IN THE SWF LYSIMETERS
 (JAN-JUNE, 1983)

<u>Matrix</u>	<u>Waste- form (mCi)</u>	<u>Porous Cup (μCi/l)</u>
Type III Cement + Bottoms	9.2	0.3
Type III Cement + Bottoms	9.8	1.1
Polymer + Bottoms	7.8	---
Polymer + Bottoms	7.8	---
Type III Cement + Bottoms + Resin	23	0.1
Type III Cement + Bottoms + Resin	21	1.3
Polymer + Bottoms + Resin	20	---
Masonry Cement + Boric Acid	0.2	---
Masonry Cement + Boric Acid	0.2	---

SWF LYSIMETERS Co-60

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