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BNL 50583

TRITIUM STORAGE DEVELOPMENT

PROGRESS REPORT #8
APRIL-JUNE 1976

Prepared for the
NUCLEAR FUEL CYCLE AND PRODUCTION DIVISION
UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
WASHINGTON, D.C. 20545

PROCESS TECHNOLOGY DIVISION
DEPARTMENT OF APPLIED SCIENCE

BROOKHAVEN NATIONAL LABORATORY
ASSOCIATED UNIVERSITIES, INC.
UNDER CONTRACT NO. EY-76-C-02-0016 WITH THE
UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

BNL 50583
(Nuclear Waste Management - TID-4500)

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5285 Port Royal Road
Springfield, VA 22161
Price: Printed Copy $3.50; Microfiche $3.00

November 1976 325 copies

Contents

SUMMARY

Lysimeter testing of polymer impregnated tritiated concrete (PITC) specimens at the Savannah River Laboratory (SRL) has continued. After twenty-three weeks, an average tritium fraction release rate of 3.37×10^{-7} day^{-1} has been determined in the percolate water with no measurable release to the air. Data from this experiment is extrapolated to estimate long-term tritium releases. A duplicate PITC specimen is in static leaching in distilled water. After seventy-two days, the average tritium fraction release rate has been 1.96×10^{-5} day^{-1}. An estimation of the increased severity of static leaching in distilled water relative to lysimeter testing is made.

I. Lysimeter Testing of Large-Scale Polymer Impregnated Tritiated Concrete (PITC) Specimens at the Savannah River Plant (SRP)

A. Specimen Preparation

SRP suggested that specimens containing one to ten curies of tritium would provide adequate detectibility in lysimeter testing and that a specimen size of one cubic foot would be convenient. Because of the low rate of tritium release expected in the lysimeter testing of PITC, ten curies of tritium were added to each specimen. The specimens were prepared in five gallon (0.67 ft^3) screw-top polyethylene carboys. These containers represented a standard size that was easily obtainable; this size was acceptable to SRP.

The size of the specimen and its container were not compatible with the end-over-end drum tumbling technique(1) being developed for large-scale PITC composites. It was determined that a conventional dough type mixer (Blakeslee model B-20) when used with a water to cement ratio of 0.22 produced a product of similar density as that obtained by end-over-end tumbling with the same water to cement ratio. Since the resultant polymer loading in a homogeneous cement casting using soak impregnation is dependent upon the initial density, the PITC specimens produced by this method are representative of the product produced by end-over-end tumbling.

The amount of water necessary to give a water to cement ratio of 0.22 for each cement casting was determined. Two millileters of tritiated water with a specific activity of 5,000 Ci/ℓ was added to this water to provide a total of ten curies for each specimen. This water was added to portland type III cement and mixed until a uniform consistancy was obtained. The cement mix was transferred to a five gallon polyethylene carboy while the carboy was being vibrated to provide proper packing of the mix. The carboy was sealed and placed in an oven for five days at 40°C to ensure complete curing of the cement.

After removal from the oven, the casting was allowed to equilibrate to room temperature before polymer impregnation. The soak impregnation technique developed at BNL was employed, in which sufficient monomer is introduced into the void space above the specimen in its container and allowed to soak into the concrete. The specimen was impregnated with styrene monomer containing 0.5 wt.% AIBN (2, 2' - [Azobis-2-methylpropionitrile]) as a polymerization catalyst. After addition of the monomer, the carboy was sealed and the monomer allowed to soak into the concrete overnight. The carboy containing the tritiated concrete was then placed into an oven at 65°C over the weekend for polymerization of the monomer. Subsequent to the equilibration of the PITC to room temperature, the polyethylene carboy was sliced away. Four polymer impregnated trititated concrete specimens were prepared by this technique.

Table 1 contains the formulational data for the four PITC specimens sent to SRP for lysimeter testing. Each specimen contains ten curies of tritium. The specimen geometry is cylindrical with approximate dimensions of 27 cm diameter x 28 cm for a composite volume of 0.566 ft^3. The tritiated concrete had a density of 91.5±0.3 lb/ft^3. Impregnation produced PITC specimens with a density of 106.6±0.2 lb/ft^3 and a polymer loading of 16.5±0.3%.

B. Lysimeter Testing at SRP

The four PITC specimens shipped to SRP were buried in individual lysimeters after removal from their shipping containers as shown in Figure 1. Each lysimeter consists of a steel tank 1.83 m in diameter and 3.05 m deep which is buried in the ground with its open top approximately 15 cm above the soil surface. The PITC specimen is buried 1.5 m below the soil surface and approximately 1.2 m above the bottom of the lysimeter using SRP soil excavated during placement of the steel tank. Rain falling on the exposed soil on the lysimeter surface permeates the soil, contacts the PITC specimen, and accumulates in a gravel layer on the sloping bottom of the lysimeter. A pump is used to collect the percolate water. After the initial time required for soil saturation, percolate water is collected weekly with volume and tritium concentration data compiled.

The tritium concentration in the air above the lysimeter is also measured periodically. This requires the placement of a cover on the open top of the lysimeter for approximately 24 hours. Figure 2 shows the lysimeter with the

TABLE 1

Formulational Data for PITC Specimens for Lysimeter Testing at SRP

Specimen Number	Formulation, Wt.% Cement	Formulation, Wt.% Water	HTO Content ℓ/ft^3	Initial Mass, kg	Impregnated Mass, kg	Polymer Load, %
121-1	82.0	18.0	7.44	23.4	27.3	16.7
121-2	82.0	18.0	7.47	23.5	27.4	16.6
121-3	82.0	18.0	7.47	23.5	27.3	16.2
121-4	82.0	18.0	7.47	23.5	27.4	16.6

Each specimen contains ten (10) curies of tritium.

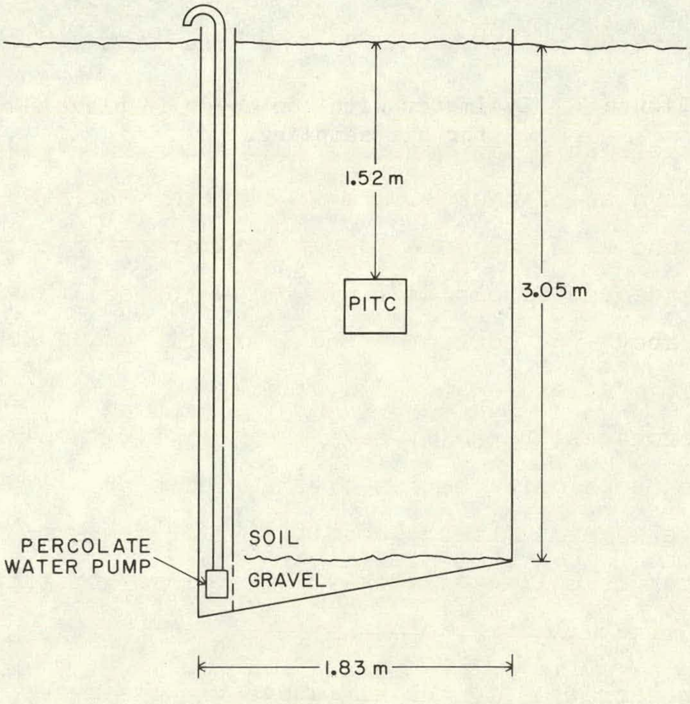

Figure 1. SRP test lysimeter for measurement of the rate of tritium release from polymer impregnated tritiated concrete (PITC).

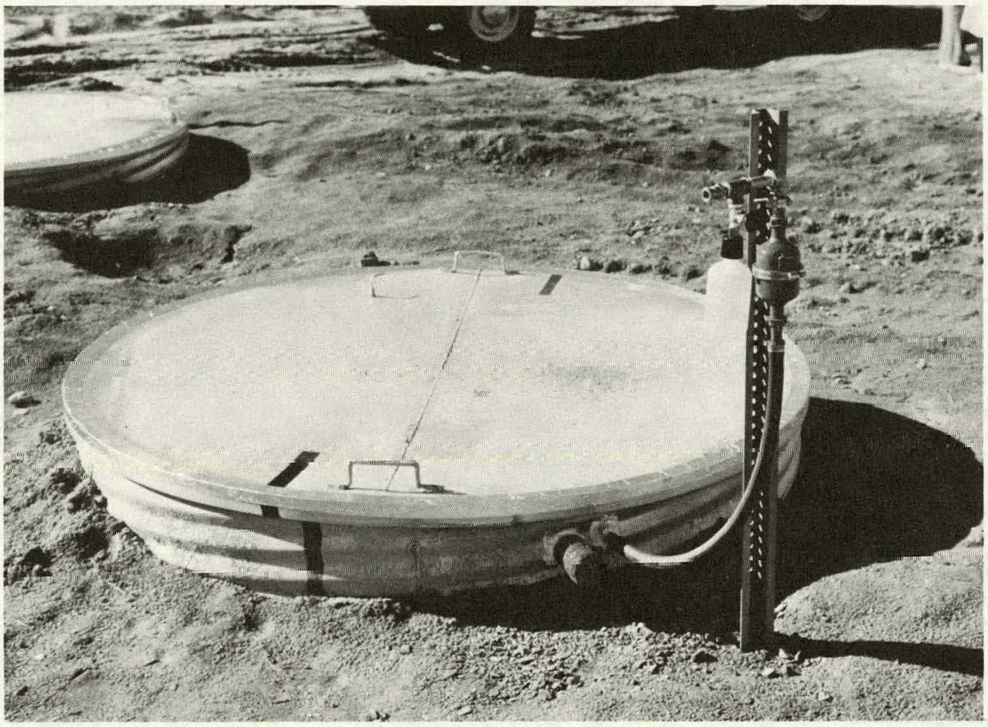

Figure 2. Lysimeter with top cover in place for air sampling.

top cover in place. Air is collected and passed first through a Linde* 4A molecular sieve trap to retain tritium oxide (HTO) and then through palladium coated Linde 4A molecular sieve to collect elemental tritium (HT) that passed through the first trap.

The four test lysimeters are denoted NE, SE, SW, and NW according to their compass position and contain respectively specimens 121-1, 121-2, 121-4, and 121-3. The specimens were each set in a nylon sling, wrapped in a polyethylene bag, and placed into a 30 gallon drum for shipment to SRP. Vermiculite was poured into the space between the drum and the polyethylene bag containing the sample. An air sampling valve and pressure gauge were mounted on the drum head, however, no pressurization occurred. Figure 3 shows one of the PITC specimens being lowered into its lysimeter. After it was placed in the lysimeter, the nylon sling was removed, and the specimen was then covered with soil. Additional soil is added if settling occurs in the lysimeter.

The cumulative aqueous tritium release from the PITC specimens (without containers) into their respective test lysimeters for the initial twenty-three weeks since burial is tabulated in Table 2. During this period the average cumulative tritium content was 618 µCi; no tritium release into the air was found. An average tritium release of 543 µCi is obtained by subtracting the background of 75 µCi found in the control lysimeter. Percolate water was not removed from the lysimeters for the first fourteen weeks. This time was required for saturation of the lysimeter soil. The total water removed from each lysimeter varied from 620 to 661 liters. This range is small so that the spread in the tritium release data is not attributed to differences in the amount of percolate water taken from the lysimeters. From this data, an average tritium fraction release rate of 3.37×10^{-7} day^{-1} is obtained.

Assuming a constant tritium fraction release rate of 3.4×10^{-7} day^{-1}, the cumulative fraction release has been plotted both with and without decay ($t_{1/2}$ = 12.26 yr) as a function of lysimeter leach time in Figure 4. A period of 8,060 years is required for the complete release of tritium from the PITC composite. The maximum fraction of the initial tritium content present in the environment is 8.04×10^{-4} after 17.7 years in burial when decay is considered.

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* Union Carbide Corp., N.Y., N.Y.

Figure 3. PITC specimen being lowered into lysimeter.

TABLE 2

Cumulative PITC Tritium Release in SRL Test Lysimeters (23 weeks since burial)

Lysimeter Designation	Specimen Number	Cumulative Aqueous Tritium Content, μCi
NE	121-1	663
SE	121-2	1,453
SW	121-4	235
NW	121-3	119
control	--	75

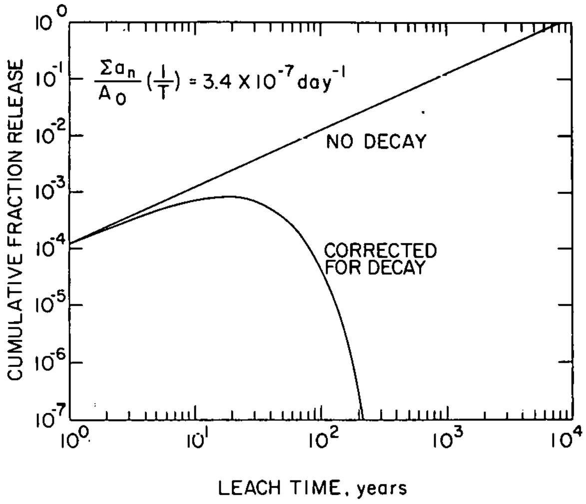

Figure 4. Projected tritium release versus time for SRL lysimeter test specimens (without containers) in burial based on the average tritium release rate for the first twenty-three weeks.

II. Static Leach Testing of the SRL Lysimeter Duplicate Specimen

A PITC specimen (without container) of the same dimensions and formulation as those in lysimeter testing at SRP is in static leaching to determine the rate of tritium release in distilled water. The data from this specimen, which also contains ten curies of tritium, will be correlated with that obtained from the lysimeter test specimens. This specimen had a density of 95.4 lb/ft^3 prior to impregnation and 107.7 lb/ft^3 afterwards with a polymer loading of 12.9%. The lower polymer loading in this specimen as compared to the lysimeter test specimens results from the higher initial density of the composite and a minimization of the polymer on the composites upper surface. The specimen was immersed in 40 liters of distilled water such that all surfaces were exposed to the leachant. The leachant was not changed; one cm^3 was taken daily for thirty-two days and weekly thereafter for analysis by liquid scintillation counting. The results of the first seventy-two days of leaching are shown in Figure 5. The tritium release is expressed in terms of (fraction tritium release) X (V/S) where V is the specimen volume and S is its geometric surface area. For this specimen (V/S) equals 4.545 cm. Multiplying the fraction tritium release by the factor (V/S) makes the tritium release independent of surface area effects.

The tritium release as a function of leach time is approximately linear with a value of 6.42×10^{-3} cm after seventy-two days. This corresponds to a fraction tritium release of 1.41×10^{-3}. Figure 6 shows the (cumulative tritium fraction release rate) x (V/S) as a function of leach time. The initial release rate decreased to become a slightly increasing linear function after fourteen days of leaching. The cumulative fraction tritium release of this specimen as a function of leach time is shown in Figure 7 assuming a constant release rate of 8.9×10^{-5} cm/day. By this estimation, the tritium release from this specimen will be complete after 140 years. The maximum fraction of the initially contained tritium present in the environment is 4.66×10^{-2} after 17.7 years. Static leaching in distilled water is a more severe test of the rate of tritium release than is expected in burial. An estimate of this increased severity can be derived by comparing the ratio of the

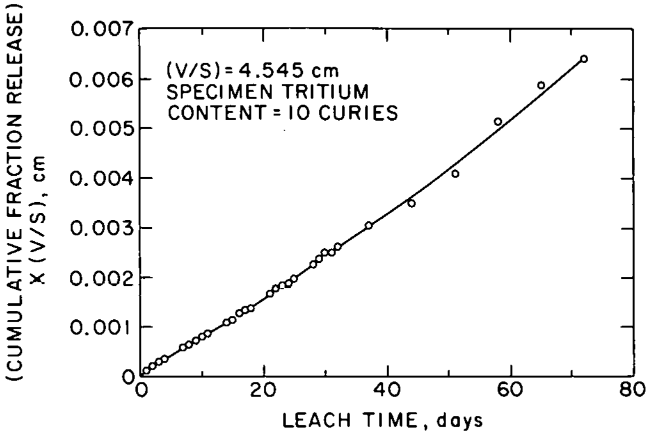

Figure 5. (Cumulative fraction tritium release) x (V/S) versus time for static leaching of the SRL lysimeter testing duplicate specimen (without container) in distilled water.

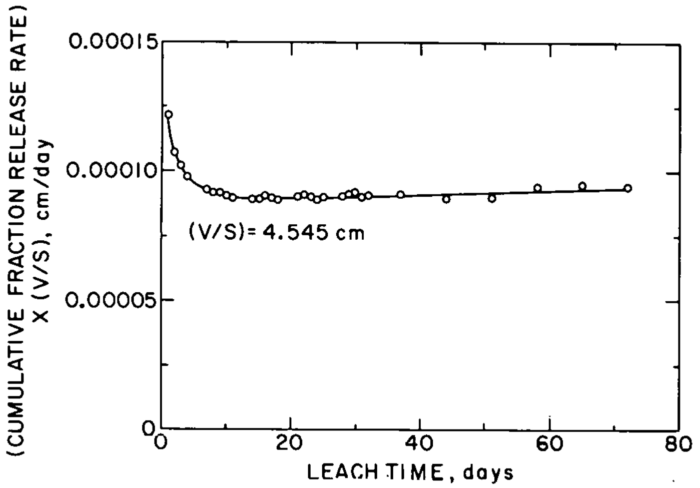

Figure 6. (Cumulative fraction tritium release rate) x (V/S) versus time for static leaching of the SRL lysimeter testing duplicate specimen (without container) in distilled water.

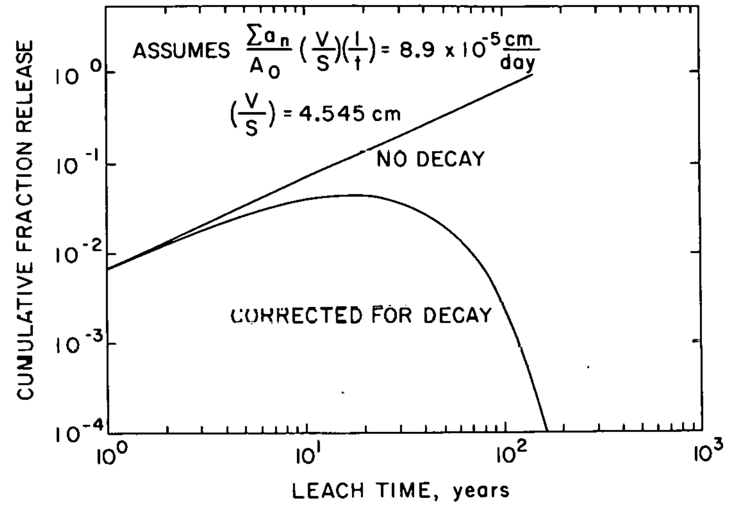

Figure 7. Projected tritium release versus time for static leaching of the SRL lysimeter testing duplicate specimen (without container) in distilled water.

fraction tritium release rate in static leaching with that for lysimeter testing of the ten curie specimens:

$$\frac{\dfrac{1.41\text{x}10^{-3}}{72 \text{ days}}}{\dfrac{5.43\text{x}10^{-5}}{161 \text{ days}}} = 58$$

The estimation of the relative severity of these leach tests will be improved as the leach testing continues. Thus far, it appears that tritium release values obtained in static leaching are approximately two orders of magnitude higher than those occurring in lysimeter testing at SRL.

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