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Environmental and Radiological Safety Studies

*Interaction of $^{238}\text{PuO}_2$ Heat Sources with
Terrestrial and Aquatic Environments
July 1—September 30, 1982*

Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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**Interaction of $^{238}\text{PuO}_2$ Heat Sources with
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July 1—September 30, 1982**

G. M. Matlack
J. H. Patterson
N. D. Stalnaker

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ENVIRONMENTAL AND RADIOLOGICAL SAFETY STUDIES

Interaction of $^{238}\text{PuO}_2$ Heat Sources with

Terrestrial and Aquatic Environments

July 1 - September 30, 1982

by

G. M. Matlack, J. H. Patterson, and N. D. Stalnaker

ABSTRACT

Although existing radioisotope thermoelectric generator designs have proved more than adequately safe, more information is continually sought about the heat sources to improve their safety. The work here includes studies of the effects on the heat sources of terrestrial and aquatic environments and also of the effects of the heat sources on various simulated environments. This progress report presents recent data from environmental chamber and aquatic experiments and gives the present status of the experiments.

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I. INTRODUCTION

The containers for $^{238}\text{PuO}_2$ heat sources in radioisotope thermoelectric generators are designed with large safety factors to ensure that they will withstand reentry from orbit and impact with the earth and safely contain the nuclear fuel until it is recovered. Existing designs have proved more than adequately safe, but the Office of Space Nuclear Projects of the Department of Energy continually seeks more information about the heat sources to improve their safety. The work discussed here includes studies of the effects on the heat source of terrestrial and aquatic environments to obtain data for design of even safer systems and also information on the dispersal and environmental effects of the heat source fuels. This report includes data from environmental chamber experiments that simulate terrestrial conditions, soil-column experiments to measure sorption of plutonium by soils, and several aquatic experiments.

II. TERRESTRIAL ENVIRONMENTS

A. MHFT-12

The two experiments with chunks and fine particles from the impact-tested PPO sphere MHFT-12 were terminated last year after 7 years. A topical report is in press.

B. MHFT-27

During the previous quarter the two groups of fragments from the impact-tested MHFT-27 were removed from the two environmental test chambers that had simulated arid winter and summer climates on sand for 7 years. The two experiments were continued without the fragments and are now under arid summer weather conditions of 26 to 53°C and 7 to 33% RH, to study plutonium transport after a simulated fuel source recovery.

In chamber 10, which contained 221 g of large fragments, greater than 6 mm in diameter, the plutonium contents in the percolated rainwater after the three rains this quarter were 2.3, 1.0,

and 0.8 μCi , an order of magnitude higher than the highest value observed under arid summer conditions when the fuel fragments were present. The average collection rate for the dehumidifier condensate was 0.3 $\mu\text{Ci/wk}$, significantly lower than the rate of 1.0 $\mu\text{Ci/wk}$ observed last quarter, probably because of cessation of spalling after removal of the fragments.

In chamber 7, which contained 32 g of fine fragments less than 6 mm in diameter, the plutonium contents in the percolated rainwater from three rains were 0.01, 0.03, and 0.04 μCi . The average of these values is significantly lower than that of any of the last four summers. The collection rate in the dehumidifier condensate was 0.3 $\mu\text{Ci/wk}$, the same as last quarter.

C. MHFT-50

Fragments from an impact-tested PPO sphere, MHFT-50, and the top inch of loam soil have been removed from two test chambers after a 5 year exposure to humid climate, and the experiments were continued for 1 year to simulate a superficially cleaned area. The soil in each chamber is in two concentric compartments; the PPO was formerly in the inner compartment. The initial quantities have been 186 g of pieces larger than 2 mm in diameter in chamber 8 and 68 g of finer material in chamber 6. These experiments were terminated last year. A topical report will be issued.

D. ET-1-1

On February 27, 1981, we placed a 15-W cylinder of GROG fuel, designated ET-1-1, on a pedestal in chamber 12. The pedestal is located above a tray containing compartmented quadrants of sand, silt loam, loam, and humus. Rainwater striking the pedestal area drains directly, without flowing through the soils, to the dehumidifier condensate bottle. Thus, mainly spalled plutonium is present in the rainwater that percolates through each soil. The chamber was operated this quarter under humid winter conditions. The average plutonium contents of the percolated waters from weekly rains were 1 nCi for sand, 4 nCi for silt loam, 4 nCi for loam, 2 nCi for humus, compared to 0.4 nCi for sand, 0.5 nCi for silt loam, 2 nCi for loam, and 0.1 nCi for humus during the previous quarter under humid summer conditions. The increase in the average plutonium content in

the percolated waters of the silt loam is due entirely to the very large value this quarter. The average dehumidifier condensates contained 2.4 $\mu\text{Ci/wk}$, not significantly different from the 2.5 $\mu\text{Ci/wk}$ observed last quarter and the previous winter quarter. Most of this plutonium is from surface particles loosened by radioactive recoil and washed from the fuel source by rain. It appears that recoil processes contribute more significantly to plutonium release than does thermal spalling.

E. Microbiological Experiments

In May 1981, we placed a 15-W GROG fuel pellet, ET-3-5, on humus soil that had been inoculated with *Streptomyces pilosus* microorganism in chamber 11. This common soil organism produces Desferal, an organic chemical that is reported to complex plutonium. The objective is to determine whether this organism changes the amount of plutonium found in percolated rainwaters. The chamber is kept at 26°C and high humidity, and one weekly rain each quarter contains nutrients that support the growth of this microorganism. The percolated water from the rains this quarter contained an average of 1.7 nCi of plutonium, significantly less than the usual contents of several hundred nanocuries from other chambers with fuel pellets on the soil and approximately the same as would be expected from spalling only. Either the plutonium is being retained by Desferal or the fuel particles carried by the percolating rainwater are solubilized by Desferal and held by ion exchange action in the soil. The increase from 0.1 nCi seen last quarter came mostly from one rain.

F. Florida Soils Experiment

Nine hundred kilograms of soil from Kennedy Space Center groves in Florida has been received for terrestrial environment experiments. A portion has been placed in a terrestrial chamber, maintained under a humid environmental condition where native Florida grasses and weeds are growing from seeds present in the soil. A bare GPHS fuel source is scheduled for insertion as soon as it is received from the Savannah River Plant. Columns of the Florida soil will be used to measure retention of plutonium under conditions similar to earlier soil-column experiments.

The purpose of these experiments is to evaluate the stability of fuels on soils typical of orange-grove soil in the vicinity of the Kennedy Space Center and to measure the plutonium release over several years' duration

III AQUATIC EXPERIMENTS

A Sealed-Glass-Chamber Experiments

A 15-W, bare, GROG fuel pellet has been in each of four sealed-glass chambers for 945 days for experiments designed to furnish material-balance data that are more complete than are obtainable from aquarium experiments. Two chambers contain simulated sea water at different temperatures of 10 and 35°C, the other two contain fresh water at the same temperatures. Table I gives the average of weekly measured plutonium release rates during this and the previous quarter. The release rates in warm waters continue to increase but are still significantly smaller than in cold waters. These data agree with previous data showing releases to be highest in cold fresh water

Table I Plutonium Release Rates from Bare Pellets in Sea Water and Fresh Water in Sealed-Glass Chambers

Measurement Period	Average Release Rate (nCi/m ² -s)			
	Fresh Water		Sea Water	
	10°C	35°C	10°C	35°C
This quarter	1520	280	60	24
Prior quarter	1470	370	53	20

and lowest in warm sea water. The causes for these differences are not understood at this time.

B Aquarium Experiments

A new experiment was started 9 months ago with a Radioisotope Heat Unit (RHU). The graphite-iridium-clad assembly was placed in an aquarium containing simulated sea water at 10°C. No significant plutonium levels have been detected in the water for 275 days.

Plutonium release data for four experiments using HPZ-designated sources over periods of 5 to 8 years are summarized in Table II. These experiments are nearing completion, and three of the fuel sources have been removed. Measurements of the plutonium contents in these three aquariums are to continue to establish changes after source removal. Figures 1, 2, and 3 show the plutonium concentration in these tanks for the last 2 years. The sources were removed in each case about day 180. The precision of each measurement is less than the circle diameter. The erratic changes indicate that the plutonium in the water is not in ionic form.

The last four experiments shown in Table II were started in March 1981, using GROG heat sources. The objective is to determine the effects of sea water and fresh water immersion on fuel stability. The release rates are highest in cold fresh water and lowest in warm sea water, consistent with earlier experiments. This is the first time the effect of warm fresh water has been studied in aquariums.

Table II Plutonium Release Rates from PPO in Aquatic Environments

Sample	Power (W)	Elapsed Days	Water	°C	Release (nCi/m ² -s)	Change from Last Quarter (%)
HPZ-60-2 ^a	2.5	3103	fresh	10	85	-6
HPZ-59-4 ^a	25	2885	sea	10	0.4	-33
HPZ-174	18	1787	sea	10	5	0
HPZ-186-4 ^a	19	1781	sea(tidal)	10	30	+50
ET-1-5	15	542	sea	10	100	-17
ET-2-2	15	542	fresh	10	195	+15
ET-2-4	15	542	fresh	37	87	-1
ET-2-6	15	542	sea	37	0.14	-30

^aSource removed from aquarium March 1981

Fig. 1. Plutonium concentration changes for HPZ-60-2. Fresh water at 10°C.

Fig. 2. Plutonium concentration changes for HPZ-186-4. Sea water at 10°C.

Fig. 3. Plutonium concentration changes for HPZ-59-4. Sea water at 10°C.