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Environmental and Radiological Safety Studies

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

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

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Environmental and Radiological-Safety Studies:

Interaction of $^{238}\text{PuO}_2$ Heat Sources with
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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
April 1 - June 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 yr. A topical report is scheduled to be issued next quarter.

B. MHFT-27

During this 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 yr. The two experiments were continued without the fragments, under winter weather conditions of 3 to 28°C and 21 to 87% 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 content in the percolated rainwater after the last rain prior to removal of the fragments was 0.03 μCi . This increased to 0.5 and 1.4 μCi for the two rains after removal of the fragments. The average value

of the plutonium contents from the rains in the previous quarter was 1.2 μCi , also under arid winter conditions. The average collection rate for the dehumidifier condensate was 1.0 $\mu\text{Ci}/\text{week}$, not significantly different from the rate of 1.1 $\mu\text{Ci}/\text{week}$ observed last quarter.

In chamber 7, which contained 32 g of fine fragments, less than 6-mm in diameter, the plutonium content in the percolated rainwater from the last rain prior to removal of the fragments was 0.005 μCi . This increased to 0.014 and 0.029 μCi for the two rains after removal of the fragments. These values are significantly lower than those of 1.1, 0.15, and 0.76 μCi observed during the previous quarter, also under arid winter conditions. Again, the lowest plutonium content was in the sample collected before removal of the fragments. The collection rate in the dehumidifier condensate was 0.3 $\mu\text{Ci}/\text{week}$ compared to 0.1 last quarter. Over half the plutonium in the eight bottles of condensate collected during the current quarter was in the one collected during the period when the fragments were removed; the relative increase in the plutonium concentration may have been caused by disturbance of the soil at this time.

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-yr exposure to humid climate, and the experiments were continued for 1 yr 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 had 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 summer conditions. The average plutonium contents of the percolated waters from weekly rains were 0.4 nCi for sand, 0.5 nCi for silt loam, 2 nCi for loam, 0.5 nCi for humus, compared to 3 nCi for sand, 4 nCi for silt loam, 3 nCi for loam, and 1 nCi for humus during the previous quarter under the same conditions. The significant reductions of plutonium content in the percolated waters of the sand and silt loam are surprising, and there is no explanation at this time. The average dehumidifier condensates contained 2.5 $\mu\text{Ci}/\text{wk}$, not significantly different from the 2.6 $\mu\text{Ci}/\text{wk}$ 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 in five contains nutrients that support the growth of this microorganism. The percolated water from the rains this quarter contained an average of 0.1 nCi of plutonium, significantly less than the usual contents of several hundred nanocuries from other chambers with fuel pellets on the soil and less than the 6 nCi obtained last quarter. 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. It is also possible that the humus soil has a higher ion exchange capacity for plutonium than other soils.

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 in this chamber in August. 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 834 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

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	1470	370	53	20
Prior quarter	1420	316	71	14

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 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 two quarters 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 220 days.

Plutonium release data for five experiments using HPZ-designated sources over periods of 1 to 7 yr are summarized in Table II. These experiments are nearing completion, and four of the fuel sources have been removed. Measurements of the plutonium contents in these four aquariums will continue to establish the decrease rate. The plutonium concentrations in these four chambers have remained approximately the same for the last two quarters, indicating that the remaining plutonium 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	3014	fresh	10	90	-10
HPZ-59-4 ^a	25	2773	sea	10	0.6	0
HPZ-174	18	1675	sea	10	5	+20
HPZ-186-4 ^a	19	1672	sea (tidal)	10	20	-5
HPZ-186-2 ^{a, b}	15	690	sea	10	1.2	0
ET-1-5	15	430	sea	10	120	0
ET-2-2	15	430	fresh	10	170	+6
ET-2-4	15	430	fresh	37	88	+22
ET-2-6	15	430	sea	37	0.2	-50

^aSource removed from aquarium March 1981.

^bAquarium glass broken 5-24-82. Experiment was terminated.

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