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Environmental and Radiological Safety Studies

*Interaction of $^{238}\text{PuO}_2$ Heat Sources with
Terrestrial and Aquatic Environments
September 26—December 25, 1981*

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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
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ENVIRONMENTAL AND RADIOLOGICAL SAFETY STUDIES

Interaction of $^{238}\text{PuO}_2$ Heat Sources with
Terrestrial and Aquatic Environments
September 26—December 25, 1981

by

G. M. Matlack and J. H. Patterson

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 Space and Terrestrial Division of the Department of Energy Office of Coordination and Special Projects 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 spring after 7 yr. A topical report will be written next quarter.

B. MHFT-27

Experiments were continued in two test chambers with fragments of the impact-tested PPO sphere MHFT-27. Experiments were changed to arid-winter conditions of 3 to 28°C and 21 to 87% RH from arid-summer conditions of 26 to 53°C and 7 to 33% RH at the end of the quarter. Chamber 10 contains 221 g of PPO pieces >6 mm in diameter on sandy soil. The rainwater that percolated through the soil after each of three monthly

rains contained from 0.05 to 0.14 μCi plutonium. This is less than the 0.07 to 0.27 μCi released during the previous quarter under arid-winter conditions, but the 0.12- μCi average for the two summer quarters is greater than earlier summer releases in this chamber. For the last 3 yr, the release each summer has been higher than for the previous summer and has now approached the lowest winter average. The average collection rate in the dehumidifier condensate was 0.8 $\mu\text{Ci}/\text{wk}$, 40% less than in the previous quarter.

In chamber 7, in which 32 g of PPO MHFT-27 pieces <6 mm in diameter were under the same test conditions as in chamber 10, the percolated water contained 0.04 to 0.09 μCi , much less than the 0.5 μCi for the previous quarter. The average collection rate in the dehumidifier condensate was 0.06 $\mu\text{Ci}/\text{wk}$, down by a factor of 5 from the 0.29 $\mu\text{Ci}/\text{wk}$ rate of the last quarter.

C. MHFT-50

Fragments from an impact-tested PPO sphere, MHFT-50, and the top inch of loam soil had been removed from two test chambers after a 5-yr exposure to humid climate, and the experiments were continued 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. The environment in each chamber continued as humid-summer climate at 20 to 40°C and 87 to 96% RH until the cleanup experiment was terminated in December, 1 yr after removal of the fuel material.

In chamber 8, the plutonium contents of the percolated waters from both the inner and outer compartments were measured after each of the last 12 summer rains at 1-wk intervals. The average plutonium content was slightly higher than during the previous quarter. The weekly average for the entire summer, which includes this quarter and the previous one, was 1.1 μCi , slightly higher than the 0.9- μCi average for the preceding winter, which began just after the sources were removed. These values are both higher than any previous winter or summer average before removal of the fuel material and top layer of soil. The disturbance caused by the removal operation may have broken up existing plutonium-soil agglomerations, thus allowing the plutonium to be carried downward by the rainwater.

Airborne plutonium was one-quarter of that found during the previous 3 months.

The average collection rate in the dehumidifier condensate in chamber 8 increased to 2.3 $\mu\text{Ci}/\text{wk}$ from 1.0 $\mu\text{Ci}/\text{wk}$ for the prior quarter. We believe that the increases seen in both the condensate and percolated water may be long-term effects associated with the removal of the PPO and top layer of soil.

During operation of chamber 6 in January 1981, an attempted clearing of plugged drains by air pressurization resulted in disturbance of the soil with formation of channels. This resulted in a 200-fold increase, from 0.05 to 10 μCi per rain, in the average plutonium content of the percolated water from the inner compartment. During the last quarter, the rate gradually decreased to the former level, but the system became plugged again. The percolated waters from the outer compartment show an average plutonium content of 0.09 μCi , significantly higher than the 0.02- μCi average for the previous quarter. The dehumidifier condensate shows an average weekly content of 0.1 μCi , much lower than the 0.6 $\mu\text{Ci}/\text{wk}$ for the previous quarter. As mentioned earlier, this experiment was terminated in December.

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 weekly plutonium contents of the percolated waters were 2 nCi for sand, 8 nCi for silt loam, 9 nCi for loam, 3 nCi for humus, compared to 0.5 nCi for sand, 0.6 nCi for silt loam, 4.6 nCi for loam, and 0.4 nCi for humus during the previous quarter under summer conditions. The average dehumidifier condensates contained 2.5 $\mu\text{Ci}/\text{wk}$, about the same as the 2.4 $\mu\text{Ci}/\text{wk}$ observed during the previous 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 10 nCi of plutonium, significantly less than the usual contents of several hundred nanocuries from other chambers with fuel pellets placed on the soil. 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.

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 22 months 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 rate in warm fresh water is higher than was measured last quarter. The rates in the other three are essentially unchanged. 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 this quarter with a Radioactive 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 46 days.

Plutonium release data for six experiments using HPZ-designated sources over periods of 1 to 7 yr are summarized in Table II. These experiments are nearing

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	1930	245	60	8
Prior quarter	1916	310	70	30

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	2826	fresh	10	24	-83
HPZ-59-4 ^a	25	2615	sea	10	1	0
HPZ-174	18	1510	sea	10	5	-17
HPZ-186-4 ^a	19	1513	sea (tidal)	10	21	-25
HPZ-186-2	15	529	sea	10	1.3	-13
ET-1-5	15	270	sea	10	103	+6
ET-2-2	15	270	fresh	10	156	+7
ET-2-4	15	270	fresh	37	69	+17
ET-2-6	15	270	sea	37	0.7	-83

^aSource removed from aquarium last quarter.

completion, and four of the fuel sources have been removed. Measurements of the plutonium contents in these four aquariums will continue for 3 months to establish the decrease rate. To date, the release rates are decreasing in the aquariums where the sources were removed.

Sample HPZ-186-2 is the heat source used in the sea cucumber experiment. All three sea cucumbers have died. The decreasing release rate is probably caused by plutonium deposition on underwater algae.

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.

C. Soil-Column Experiments

Of six soil-column experiments, three (designated 7, 8, and 9) use columns that are 14 cm high and 2.5 cm in diameter, each filled with silt loam soil. Each column

contains 3 mg of $^{238}\text{PuO}_2$ particles. The approximate particle sizes are 50 μm for column 7, 12 μm for column 8, and 7 μm for column 9. All particles are on top of the soil column. Distilled water flows through the columns at 0.4 to 0.8 $\mu\text{l/s}$. The columns have now operated 61 months. The total plutonium contents of the effluents are 50, 63, and 69 ng, respectively, for columns 7, 8, and 9. These results substantiate that smaller particles release more plutonium than larger particles. The total percentage released is less than 0.002%. These experiments are to be terminated.

In the other three columns, the sorption of soluble plutonium by sandy soils is being measured. The feed solution to the columns contains 15 $\mu\text{g/l}$ of ^{238}Pu in distilled water and flows at a rate of 2 $\mu\text{l/s}$. Last quarter, after 575 days, about 99.5% of 1280 μg plutonium remained on the columns. This was less than the 99.9+% retained by the silt loam soils. Very close to the end of last quarter, a sudden breakthrough occurred where the plutonium increased by a factor of about 10. The breakthrough occurred with about 1.5 mg of plutonium on each column. This was never observed with loam soil columns that were fed soluble plutonium, even after 5 yr of operation.

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