236
12-15-81
⑧

LA-9070-PR

Progress Report

①
I-389
MASTER

DR 141

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

Environmental and Radiological Safety Studies

*Interaction of $^{238}\text{PuO}_2$ Heat Sources with
Terrestrial and Aquatic Environments
July 1—September 25, 1981*

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

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

The four most recent reports in this series, unclassified, are LA-8664-PR, LA-8811-PR, LA-8932-PR, and LA-8996-PR.

This work was supported by the US Department of Energy, Space and Terrestrial Division, Office of Coordination and Special Projects.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

**Environmental and
Radiological Safety Studies**
**Interaction of $^{238}\text{PuO}_2$ Heat Sources with
Terrestrial and Aquatic Environments**

July 1—September 25, 1981

**G. M. Matlack
J. H. Patterson**

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof nor any of their employees makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

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

2B

QUARTERLY REPORT
ENVIRONMENTAL AND RADIOLOGICAL SAFETY STUDIES

Interaction of $^{238}\text{PuO}_2$ Heat Sources
with Terrestrial and Aquatic Environments

July 1—September 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.

No reference to results cited in this report should be published without the explicit permission of the person in charge of this work.

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 an impact-tested PPO sphere MHFT-27. Experiments were changed from arid-winter conditions of 3 to 28°C and 21 to 87% RH to arid-summer conditions of 26 to 53°C and 7 to 33% RH in late June 1981. Chamber 10 contains 221 g of PPO

pieces >6-mm diam on sandy soil. The rain-water that percolated through the soil from three month-separated rains contained from 0.07 to 0.27 μCi plutonium. This is less than the 0.13 to 0.76 μCi released during the previous quarter under arid-winter conditions, but greater than earlier arid-summer releases. For the last 3 yr. the release each summer has been higher than for the previous summer. The average collection rate in the dehumidifier condensate was 1.3 $\mu\text{Ci/wk}$, similar to previous rates.

In chamber 7, in which 32 g of PPO MHFT-27 pieces <6-mm diam were under the same test conditions as in chamber 10, the percolated water contained 0.09 to 0.9 μCi , higher than the previous winter, a peculiarity of this chamber. The average collection rate in the dehumidifier condensate was 0.29 $\mu\text{Ci/wk}$, slightly higher than the 0.19 $\mu\text{Ci/wk}$ rate of the last quarter.

C. MHFT-50

Fragments from an impact-test PPO sphere, MHFT-50, and the top inch of loam soil were previously 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 diam 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 since it was changed late last quarter from humid-winter conditions.

In chamber 8, the plutonium contents of the percolated waters from both the inner and outer compartments were measured after each of 13 summer rains at 1-wk intervals. There was no trend of increasing or decreasing plutonium contents of the weekly values during the 13 weeks. The average weekly value of 0.89 μCi for the inner compartment is not significantly different from the

overall average of 0.87 μCi for the 27 previous winter rains, but these contents are higher than any previous winter or summer average before removal of the fuel material and top layer of soil. The average weekly value for the outer compartment during this quarter was 0.16 μCi , somewhat lower than the 0.22 μCi average observed for the preceding season.

The average collection rate in the dehumidifier condensate in chamber 8 increased to 2.3 $\mu\text{Ci/wk}$ from 1.0 $\mu\text{Ci/wk}$ 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 μCi to 10 μCi per rain, in the average plutonium content of the percolated water from the inner compartment. During this quarter, the rate has gradually decreased to the former level, but the system is plugged again. The percolated waters from the outer compartment show an average plutonium content of 0.02 μCi , significantly lower than the 0.07 μCi average for the previous 27 winter rains. The dehumidifier condensate shows an average weekly content of 0.63 μCi , slightly less than the 0.82 $\mu\text{Ci/wk}$ for the previous quarter.

D. ETl-1

On February 27, 1981, we placed a 15-W cylinder of GROG fuel, designated ETl-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 were 0.5 nCi for sand, 0.6 nCi for silt loam, 4.6 nCi for loam, 0.4 nCi for humus, compared to 0.3 nCi for sand, 0.1 nCi for silt loam, 2.4 nCi for loam, and 0.6 nCi for humus during the previous quarter under summer conditions. The average dehumidifier condensates contained 2.4 μ Ci/wk, lower than the 4.0 μ Ci/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 Experiment

Last quarter, we placed a 15-W GROG fuel pellet, ET3-5, on humus soil that had been inoculated with *Streptomyces Pilosus* microorganism. 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 nCi 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 21 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°C 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 cold fresh water is higher than 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

Plutonium release data for six experiments using HPZ-designated sources over periods of 1 to 7 yr are summarized in Table II. This experiment is nearing completion, and four of the fuel sources have been removed. Measurements of the plutonium contents in these four aquariums will continue for three 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, and the aquarium is overgrowing with algae. The decreasing release rate in this aquarium is probably caused by sorption of plutonium by the organic material.

The last four experiments shown in Table II were started last quarter, using GROG heat sources. The objective is to determine the effects of sea water and

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	1910	310	70	30
Prior quarter	1960	330	74	24

TABLE II
PLUTONIUM RELEASE RATES FROM PPO IN AQUATIC ENVIRONMENTS

<u>Sample</u>	<u>Power (W)</u>	<u>Elapsed days</u>	<u>Water</u>	<u>°C</u>	<u>Release (nCi/m²-s)</u>	<u>Change from last quarter (%)</u>
HPZ-60-2 ^a	2.5	2725	Fresh	10	140	+12
HPZ-111-1 ^{a, b}	25	2417	Fresh	10	330	-14
HPZ-59-4 ^a	25	2525	Sea	10	1	-50
HPZ-174	18	1420	Sea	10	6	+20
HPZ-186-4 ^a	19	1416	Sea(Tidal)	10	28	-26
HPZ-186-2	15	432	Sea	10	1.5	0
ET-1-5	15	174	Sea	10	97	+21
ET-2-2	15	174	Fresh	10	146	- 3
ET-2-4	15	174	Fresh	37	59	+18
ET-2-6	15	174	Sea	37	4	-71

^aSource removed from aquarium last quarter.

^bTerminated this quarter because of hazardous leak.

freshwater immersion on fuel stability. The findings to date are consistent with data obtained for older PPO fuel materials. The release rates are highest for cold fresh water and lowest in warm sea water. 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 diam, each filled with silt loam soil. Each column contains 3 mg of ²³⁸PuO₂ particles. The approximate particle sizes are 50 μm for column 7, 12 μm for 8, and 7 μm for 9. All particles are on top of the soil column. Distilled water flows through the columns at 0.4 to 0.8 μl/s. The columns have now operated 59 months. The total plutonium contents of

the effluents are 40 ng for both columns 7 and 8 and 49 ng for column 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 μg/l of ²³⁸Pu in distilled water and flows at a rate of 2 μl/s. Last quarter, after 505 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. Late this quarter, a sudden breakthrough of plutonium occurred with significantly more in the effluent than in the feed. This was never observed with loam soil columns, even after 5 yr of operation.