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J. H. Patterson

B. Herrera

G. B. Nelson

G. M. Matlack

G. R. Waterbury

**los alamos
scientific laboratory**

of the University of California

LOS ALAMOS, NEW MEXICO 87545

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INTERACTION OF AN IRIIDIUM-CLAD RTG HEAT SOURCE UNIT WITH A SIMULATED TERRESTRIAL ENVIRONMENT

by

J. H. Patterson, B. Herrera, G. B. Nelson,
G. M. Matlack, and G. R. Waterbury

ABSTRACT

An iridium-clad, 100-W $^{238}\text{PuO}_2$ sphere, a prototype for the multihundred-watt radioisotope thermoelectric generator, was exposed for 1 yr to a simulated temperate humid climate in an environmental test chamber containing sandy soil. The hot sphere sank into the soil after the first rain, then gradually acquired a hard crust around it as a result of the rainwater reacting with the hot soil during successive rains. Time and temperature profiles of the sphere were recorded during the weekly rains, and the air and rainwater that percolated through the soil were monitored for plutonium. No plutonium was released from the sphere. Aside from the crust formation, very little reaction occurred between the hot iridium shell and the soil.

I. INTRODUCTION

Radioisotope thermoelectric generators (RTGs) provide auxiliary electric power on many space missions. An RTG produces electric energy by a thermopile, whose hot junctions are heated by the decay of a radioactive isotope. Heat sources now used in RTGs for space applications contain pure plutonium oxide (PPO), which is prepared by the application of heat and pressure from purified plutonium dioxide powder that is 80% enriched in ^{238}Pu . The heat sources are then suitably packaged to prevent environmental contamination in case of an accident. As part of the packaging testing program, we subjected several fueled-sphere assemblies (FSAs) to a sequential safety test (SST) series that simulates the experience of FSAs during launch, reentry, and impact.¹ The final test, after impact, was to expose one of the postimpact sphere assemblies (PISAs), a PPO sphere in its postimpact containment shell (PICS), to environmental conditions in sandy soil for 1 yr.

An FSA is a basic fuel unit in the multihundred-watt (MHW) system that consists of a 37-mm-diam

sphere of PPO that weighs ~250 g. The sphere is encapsulated in a vented PICS of ~0.6-mm-thick iridium, which is, in turn, enclosed in a 12-mm-thick graphite impact shell (GIS) of carbon-impregnated Thorneal yarn.

II. ENVIRONMENTAL EXPOSURE

We used an environmental test chamber with a 1.8-m³ working volume to study the release of ^{238}Pu to the environment from an intact PISA after accidental land impact of a spacecraft with an RTG. The chamber was equipped with temperature and humidity controls to simulate a natural diurnal cycle. Rain was simulated by a spray from four heads that distributed the water evenly.

The impacted PISA (MHFT-27) was partially buried in 0.3-m-deep sandy soil (Table I) in a 0.9- by 0.9-m tray in the environmental chamber. This PISA had one superficial crack that apparently did not penetrate the shell.¹ It was kept under humid conditions in the chamber for 373 days, of which 202

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TABLE I
SOIL COMPOSITION^a

Sand (%)	88.6
Silt (%)	4.6
Clay (%)	6.8
Carbon, organic (%)	0.01
Carbonate as CaCO ₃ (%)	4
H ₂ O at 15 bars (%)	2.6
H ₂ O at saturation (%)	22
pH	9.1
Calcium (meq/100 g)	17.8
Magnesium (meq/100 g)	1.7
Sodium (meq/100 g)	0.9
Potassium (meq/100 g)	0.5
Cation exch cap (meq/100 g)	6.0
Elect cond (mmho/cm)	0.6
Clay mineral ratio:	
Montmorillonite	5
Kaolinite	3

^a Analysis provided by the U.S. Soil Conservation Service Laboratory, Riverside, CA.

were under summer conditions (20 to 40°C, 87 to 96% RH) and 171 were under winter conditions (0 to 17°C, 71 to 100% RH). The PISA received ~32 mm of rain weekly during the experiment. The water that percolated through the soil during and after each rain was collected for plutonium analysis. We measured the air concentration of plutonium by analyzing the water condensed from the air by the refrigeration-type dehumidifier in the chamber conditioning system. To obtain a depth profile of the plutonium, two pairs of 13-mm-diam soil cores, collected about every 6 months, were sectioned by horizontal cuts. Thermocouples welded to the top and bottom of the PISA continuously monitored the surface temperature, which was recorded using a Honeywell circular chart recorder.

During the first rain, the PISA, which had been placed with 60% of its height above the surface of the soil, sank to a depth that left only 10% of its height exposed. We intentionally left the soil under the PISA very loose with large voids to avoid breaking the thermocouple welds during emplacement. Lubrication of the sand grains by the rain allowed the heavy PISA to sink into the loosely filled space. The surrounding soil may have been temporarily fluidized by the boiling of the rainwater by the hot PISA.

When most of the sphere was above the surface, the initial temperature of the PISA, as measured by the bottom thermocouple, was 620°C. At the beginning of the first rain, the temperature dropped to 100°C in 5 min, where it remained for 42 h. It then began to rise, finally leveling off at 520°C 1 wk after the rain. Evaporation of water in the surrounding soil seemed to be the mechanism that kept the PISA at 100°C for so long a time. After heating the water from 25 to 100°C, the 100 W of thermal power from the PISA would probably evaporate about 6 liters of water in 42 h. The temperature profile changed gradually to shorter lengths of time at 100°C. Seven months later the PISA stayed at 100°C only during the rain. When the rain stopped, the temperature rose rapidly to 300°C, then more slowly to the equilibrium temperature (Fig. 1). This indicated that a crust gradually formed and isolated the PISA from the water in the surrounding soil.

After the PISA sank into the soil during the first rain, the equilibrium temperature for the bottom thermocouple was 520°C, compared with 620°C before the rain. The temperature of the bottom thermocouple was also consistently lower than that of the top one. The temperature differential range was 0 to 70°C, and most often was less than 30°C. The greatest difference occurred when the temperature started to rise from 100°C, because the temperature of the top thermocouple started to rise before the bottom one. At higher temperatures, the top and bottom readings were often identical. These temperature trends indicate that there was good thermal conductivity between the top and bottom of the PISA, probably through the iridium shell, and

Fig. 1.
Temperature response of PISA (MHFT-27) to a rain sequence.

that the heat was more efficiently removed by conduction to the soil than by convection and radiation in air. Radiation losses however would be greater in the open under clear skies.

Although the surface of the PISA had been previously contaminated by handling in a drybox that had been used with ^{238}Pu , we found very little plutonium in the water and soil samples from this experiment. Only 8 of 52 percolated rainwater samples and 6 of 74 dehumidifier condensate samples contained enough plutonium to be distinguishable from background. The total plutonium in these 14 samples was $0.23\ \mu\text{g}$. The soil cores contained even less plutonium. One core had $0.2\ \text{ng}$ of plutonium in each of its top two sections. We found no significant amount of plutonium in any of the other sections or cores. The plutonium found in this system undoubtedly came from the previous contamination of the PISA surface.

III. TERMINATION OF THE EXPERIMENT

After 1 yr of exposure to this humid environment, we removed the PISA from the environmental chamber and found it almost completely encased in a 0.5- to 3-cm-thick soil crust that could not be broken manually without tools. This crust, apparently formed by the heat from the PISA baking the soil dry between rains, caused the change with time of the response of the surface temperature of the PISA to rain.

After its removal from the chamber, the PISA was cut open and the iridium shell and its plutonium dioxide contents were examined.² A bubble test revealed that both of the fritted vents for helium, located at opposite poles of the PICS, were closed. Metallographic and electron microprobe examinations indicated that the vents were closed by a combination of vapor-deposited plutonium dioxide and a glassy phase consisting mostly of calcium, silicon, aluminum, and oxygen. Sintering of the frits also may have contributed to the plugging. These causes, common to closed vents in other PICS, probably happened before the PISA was exposed to the test environment. A plug, found at the mouth of

the vent near the bottom of the PISA in the soil, contained the above elements plus phosphorus, which is not usually found in vents. The phosphorus may have been a product of interaction between the PISA and the soil. We found no evidence of water penetrating into the PICS or of attack on the iridium from inside or outside the PICS. A heavy coating of vapor-deposited plutonium dioxide seemed to have been dislodged from its normal tight adherence to the inside of the PICS by the rapid thermal changes caused by the rains in the environmental chamber.

IV. CONCLUSIONS

A PISA on, or just below, a sandy soil surface maintains a temperature below 650°C . At the beginning of a rain, the surface temperature drops to 100°C in 5 min. After 1 yr of temperate humid conditions, including a weekly 32-mm rain, there is evidence of only a mild interaction between the PISA and its surroundings. There was no evidence of water penetrating into the PICS or of plutonium escaping from its containment. These results may be compared with those from another experiment in which a PISA was buried 23 cm deep in dry soil for 67 days.¹ In that experiment, the PICS reached 1150°C and on recovery was found to have a thick coating of fused soil. In many places where the iridium was not protected by the fused soil, it was perforated by an erosive reaction with air and water vapor. Also, $0.02\ \text{g}$ of plutonium dioxide was found in the fused soil.

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