LA-9487-MS

UC-338^A

Issued: December 1982

LA--9487-MS

DE83 005607

Long-Term Exposure of $^{238}\text{PuO}_2$ to a Terrestrial Environment

James H. Patterson
Benjimen Herrera
Gilbert B. Nelson
Frederick J. Steinkruger
George M. Matlack
Daniel Pavone

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.

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

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

LONG-TERM EXPOSURE OF $^{238}\text{PuO}_2$ TO A TERRESTRIAL ENVIRONMENT

by

James H. Patterson, Benjimen Herrera,
Gilbert B. Nelson, Frederick J. Steinkruger,
George M. Matlack, and Daniel Pavone

ABSTRACT

In many space missions that use radioisotope thermoelectric generators, $^{238}\text{PuO}_2$ is used as the source of heat. We exposed some of this heat-source material to simulated terrestrial environments for an 8-yr period. During this time we monitored the release of plutonium to water, air, and soil. Plutonium was found in the air, especially after the beginning of a rain, and in the water that percolated through the soil after a rain, but the major part of the plutonium was held in the soil.

I. INTRODUCTION

Radioisotope thermoelectric generators (RTGs) provide auxiliary electric power for instrument operation and data transmission on many space missions. An RTG produces electric energy from a thermopile whose hot junctions are heated by the decay of a radioactive isotope. Plutonium-238 is the isotope used almost exclusively in the space program because its 88-yr half-life provides a source of intense heat that is relatively constant for several years. The current RTG heat sources consist of hot-pressed massive pieces of plutonium oxide that is 80% enriched in ^{238}Pu . In the multihundred watt (MHW) configuration the basic fuel unit is a 37-mm-diam sphere of this pressed plutonium oxide (PPO). The 250-g sphere is encapsulated by a vented postimpact containment shell (PICS) of 0.6-mm-thick iridium. This postimpact sphere assembly (PISA) is enclosed by a 12-mm-thick graphite impact shell (GIS) of carbon-impregnated Thorne[®] yarn to make up a fueled-sphere assembly (FSA). A number of FSAs compose the heat source for an RTG.

In the MHW Safety Sequential Testing (SST) program FSAs were heated to simulate reentry from satellite orbit and then impact tested on granite at terminal velocity.¹ To simulate possible fates of the heat source material after impact, some impacted FSAs were tested in environmental experiments. An FSA and a PISA were exposed to simulated terrestrial environments in environmental test chambers.^{2,3} In this and other experiments, we used bare PPO to simulate the unlikely extreme case of failure of both the GIS and the PICS on impact.

II. EXPERIMENTS

A. Chamber Operation

The fragmented PPO from the impacted test FSA (numbered MHFT-12) was removed from the PICS and separated into three size fractions: particles with diameters $< 10\ \mu\text{m}$, "airborne"; particles with diameters from $10\ \mu\text{m}$ to 6 mm, "fines"; and pieces with diameters $> 6\ \text{mm}$, "chunks" (Fig. 1). The airborne fraction had

Fig. 1. The PPO chunks on soil.

been used to determine the airborne source term. We placed the fines (28 g) and the chunks (219 g) in two environmental test chambers with working volumes of 1.8 m³. The two PPO fractions were centrally placed on 0.9-m-square, 0.3-m-deep trays filled with loam soil (Table I). The air temperature and humidity were cycled diurnally over ranges of 20 to 40°C and 85 to 97% RH, respectively, in both chambers. This humid summer regime was alternated with a humid winter one of 0 to 17°C and 60 to 100% RH for 6-month periods. A spray from four heads that distributed the water evenly simulated a weekly 32-mm rain in each chamber.

B. Plutonium in Soil Cores

Near the end of each summer and winter season, for the first nine seasons, we took two soil cores for plutonium analysis. We obtained the cores, from positions equidistant from the plutonium source in opposite directions, by pushing sections of 12-mm copper tubing into the full soil depth. We filled the resulting holes with melted paraffin wax to prevent channeling of the rain-water through the soil. Each core was divided into from 5 to 14 sections that were individually analyzed to determine plutonium variation with depth. After the

sections were dissolved in nitric and hydrofluoric acids, the solutions were mixed with a commercially available xylene-based scintillation cocktail, for liquid scintillation counting.

C. Plutonium in Aqueous Effluents

The soil in each tray rested on a perforated sheet of stainless steel supported by a grid above the tapered tray bottom. The water that percolated through the soil after each rain was collected and analyzed by liquid scintillation counting to determine ground water contamination by plutonium. Plutonium in the condensate from the refrigeration-type dehumidifier of the chamber conditioning system was also determined. Analysis of the condensate indicated trends in plutonium concentration in the chamber air but did not quantitatively measure it.

D. Plutonium in Air

We determined airborne concentrations of particulate plutonium by passing measured volumes of chamber air

TABLE I. Loam Composition^a

Sand, %	49.5
Silt, %	37.6
Clay, %	12.9
Carbon, organic, %	0.6
Carbonate as CaCO ₃ , %	trace
H ₂ O at 15 bars, %	7.6
H ₂ O at saturation, %	40
pH	8.0
Calcium, meq/100 g	16.3
Magnesium, meq/100 g	1.9
Sodium, meq/100 g	0.5
Potassium, meq/100 g	1.0
Cation exchange cap	13.6
Electrical conductivity, mmho/cm	0.6
Clay mineral ratio	
Mica	4
Montmorillonite	3
Kaolinite	3

^aSoil analysis provided by the US Soil Conservation Service Laboratory, Riverside, California.

through filters with ~40-mm diam. These measurements were made before, during, and after rains, as the greatest concentration changes were expected then. We first determined the prerain concentration. We changed the filters several times during the 1-h rain to determine the effect of rain on the concentration. The measurements continued until prerain values were reached. This sequence of measurements was repeated four times in the chunk chamber. We varied the position of collection in the chamber, the type and pore size of the filters, and the dehumidifier operation (on/off). One such measurement sequence was run in the fines chamber. We measured the volume of air by recording the length of time that air from the chamber was pumped through the filter at a constant known rate. We measured the alpha activity of the filters directly in a gas proportional counter and then dissolved some of the filters for liquid scintillation counting.

E. Photographic Experiments

Temperature estimates were made from studies of the infrared emission of the chunks shown in 15- to 63-h time-exposure photographs taken without external lighting.

These photographs, which distinguished emissions in the near-ir, visible, and near-uv regions by filtering various spectral bands, showed PPO surface temperatures of 250 to 300°C in agreement with optical

pyrometer measurements. Around the chunks was an air glow, which was probably due to alpha particle ionization of the air and was mainly in the visible region.

III. RESULTS

The condensates collected in the chunk chamber for periods that included rains were higher in plutonium than those collected between rains (Fig. 2). This agrees with the measurements of airborne plutonium during four rain sequences in this chamber (Table II). Although we varied filter type and pore size, position of collection, and dehumidifier status, the plutonium concentrations in air showed remarkably similar trends during corresponding times in the rain cycles. In each case the prerain concentration was comparatively low. During the first 5 min of the rain, the concentration rose very rapidly, usually by four orders of magnitude. This was immediately followed by a rapid exponential decrease, with a half-time of less than 10 min. After the rain ended, the concentration returned to the prerain value within 3 h.

We attribute this high plutonium concentration in air, at the beginning of a rain, to spalling of the hot chunks from thermal shock when cold rain strikes them. Spalling has been observed in nature when rocks heated by the sun were hit by rain.⁴ During one rain a small particle was observed flying from one of the chunks in the initial period when the water striking the pieces immediately evaporated with a puff of steam. Within 5 min the

Fig. 2. Effect of rain on plutonium content of dehumidifier condensates.

TABLE II. Plutonium Concentrations in Air Over Chunks of PPO During Rains (pci/m³)

Variable	Experiment Number			
	1	2	3	4
Time				
Prerain (min)	3 (24)	960 (29)	8 (21)	11 (31)
First 5 min of rain	30 000	328 000	59 000	110 000
Second 5 min of rain	6 000	104 000	10 000	54 000
Second 10 min of rain	4 000	41 000	6 000	21 000
Third 10 min of rain	3 000	12 700	1 500	6 800
Third 15 min of rain	900	6 700	1 000	1 900
Final 15 min of rain	800	2 800	200	630
Postrain (min)	200 (50)	2 300 (30)	250 (15)	320 (30)
	3 (180)	290 (67)	90 (63)	41 (30)
	3 (860)	100 (60)	20 (60)	4 (60)
		490 (53)	7 (60)	0 (138)
		290 (60)	4 (60)	4 (120)
		41 (55)	4 (54)	9 (56)
		6.4 (910)	2 (900)	9 (909)
Filter type	glass fiber	glass fiber	Millipore® (0.45 µm)	Millipore® (0.22 µm)
Collection point	below tray	below tray	below tray	above tray
Dehumidifier	on	off	off	on

chunks cooled below the boiling point and became wet, so that the spalling stopped. Collection of particles by the dehumidifier and washout by rain helped remove plutonium from the air, but the main mechanism must have been gravitational settling, because the process continued after the rainfall when the dehumidifier was off. The high prerain concentration in experiment 2 was probably caused by our inadvertently splashing water on the pieces before the rain started. In most experiments the final concentration was less than that before the rain, possibly because of decreased resuspension of plutonium-bearing dust from the wet soil.

Spalling was not observed in the fines during rains, as the smaller pieces were not thermally hot enough. In the fines chamber, the rate of plutonium collection by the dehumidifier condensate seemed independent of the time of rains. There also was no rapid release of plutonium in air during a rain (Table III). The concentration was an order of magnitude lower after the rain than before it. Counts were so low that totals taken during the rain were combined, as they also were for the first 164 min after

TABLE III. Plutonium Concentration in Air Over Fines During a Rain

Filter No.	Collection		Pu Concentration (pCi/m ³)
	Time	Duration (min)	
1	Before rain	103	1.9
2	First 5 min of rain	5	2.1
2-7	During rain (av)	60	1.9
8-12	After rain (av)	164	0.12
13	Overnight	926	0.14

the rain. The rate of plutonium collection by the dehumidifier for the fines chamber was only one-thirteenth of that for the chamber containing the chunks.

The water that percolated through the soil after the first rain on the chunks contained 1.8 µCi of plutonium. This very rapid plutonium breakthrough indicated that rainwater released extremely small particles or a colloid

from the surface of the PPO. The low carbonate and organic carbon content of this soil (Table I) indicates that the strong complexing agents that keep plutonium in solution at the pH of the percolated water (~8) are not present. Also, ionic plutonium is held, at least for a time, by ion exchange in the clay fraction of the soil. The percolated water from the first rains of the fines chamber also had high plutonium contents indicating a release mechanism different from the spalling of the large hot pieces.

The presence of plutonium oxide particles in percolated rainwater was confirmed by autoradiography of a residue from a rainwater sample from the chunk chamber.* Particles up to 0.8 μm in diameter were detected. However, all the particles found accounted for only 1 ppm of the activity in the sample, indicating that nearly all the particles in the water had diameters smaller than 0.05 μm , the limit of detection for this analysis.

Fleischer and Raabe⁵ found that very fine particles, each containing less than 10^4 plutonium atoms, are released by water from the surface of plutonium dioxide. Fleischer⁶ showed that the amount of plutonium released in a given time is proportional to the decay rate of the plutonium for different plutonium isotopic mixtures and that the fine particles are formed on the oxide surfaces by the disruption of crystal structure by the recoil nuclei. Fleischer and Raabe⁷ also found that very little plutonium is released from $^{239}\text{PuO}_2$ in a vacuum. Using fission track analysis, they found an average of less than one plutonium atom per atom of escaping uranium daughter in the dry state, compared with ~1000 atoms released per escaping recoil nucleus in water. This contrasts with other research with ^{242}Cm , in which aggregate recoil was observed in a vacuum.⁸ Perhaps the higher alpha-particle energy of the curium, 6.1 MeV compared with 5.2 MeV for ^{239}Pu , provides enough energy to the recoil nucleus to overcome the forces holding the fine oxide particles to the parent piece. The lower plutonium concentration in the air after rains probably indicates that the between-rains plutonium concentration in air comes from resuspension rather than aggregate recoil.

Twenty soil cores were taken in the chunks chamber; eighteen, in the fines chamber. Core 10 of the chunks chamber was analyzed for plutonium dioxide particle size by LFE Environmental Analysis Laboratories.** The other 37 cores were sectioned and analyzed for

plutonium (Tables IV and V). The second and third lines of each table show the number of days of exposure to the plutonium dioxide source and the weather regime (summer or winter), respectively, at the time each core was collected. The position from which each core was taken, in relation to the plutonium dioxide source, appears on the fourth line. The number is the approximate distance in centimeters, and the letters indicate the direction from the source (front, back, left, right, or a combination of these). The plutonium data are in nanocuries, except where marked μ for microcuries or m for millicuries. For cores divided into fewer than 10 sections in the chunks chamber or 14 sections in the fines chamber, the internal section values were placed on rows with sections of roughly equivalent depths, with dashes on the other lines.

The total plutonium in these cores is quite high, considering that less than 0.3% of the soil in the trays was sampled. If we assume that these samples are representative, the soil in the chunks chamber would appear to contain 72 mCi plutonium, and the fines chamber soil, 4.5 Ci. This compares with 1.1 and 0.3 mCi, respectively, for these two chambers in the sum of the dehumidifier condensates and the percolated rainwaters. For the fines chamber this is more than 1% of the source as originally added. The higher plutonium release rate for the fines may be related to their greater surface area. The greater water-to-soil ratio for the chunks may result from the thinner layer of soil in the chunks tray (~18 cm) compared with 25 cm for the fines.

The range of plutonium contents in the cores is at least seven orders of magnitude, from 0 (less than 0.3 nCi) to 6 mCi. There is no clear evidence that plutonium increases with time; other factors, such as location and season, seem more important. In the fines chamber plutonium content seems to correlate with distance from the source, in that the average for the cores at 13 cm is greater than that for cores at 25 cm, which is greater, in turn, than the average for cores at 38 cm. The correlation is not as clear for the chunks chamber. However, there is an apparent correlation with season for chunks, as the average plutonium measurement for summer cores is more than twice that of the winter cores. In the fines cores seasons do not seem to affect these averages.

In measuring plutonium distribution within the cores, we found ~90% of the activity in the top sections. This agrees with the results of other researchers who determined plutonium profiles in soils in the field. Essington et al.⁹ studied plutonium distribution in soils of the safety shot areas at the Nevada Test Site and classified plutonium profiles as low, normal, and abnormal. The

*D. Rohr, Los Alamos National Laboratory, unpublished data, 1973.

**LFE Environmental Analysis Laboratories, 2030 Wright Avenue, Richmond, CA 94804.

TABLE IV. Plutonium in Cores from Chunks Chamber^a

Core number	1	2	3	4	5	6	7	8	9
Days exposed	168	168	271	271	391	391	575	575	663
Season	S	S	W	W	S	S	W	W	W
Position	23L	23R	25F	25B	25FL	25BR	25FR	25BL	20L
Top section	0	50	0.40 μ	28.1 μ	2.21 μ	9.1	0.63 μ	15.4	332
Inner sections	0	4.9	---	---	0.3	0.7	0	0.5	0
	0	42	0	0	115	0	0	0	0
	78	0.26 μ	---	2.6	611	0	0	1.4	0
	0	9.0	4.4	---	0	0	0	2.7	0
	0	89	0	0	0	0	0	1.5	0
	0	6.0	---	0	0	0	0	6.0	0
	0	1.6	0	7.1	0	0	0	61.8	0
	0	26	---	---	0	0	0	8.5	0
Inner total	78	0.44 μ	4.4	9.7	726	0.7	0	82.4	0
Bottom section	0	1.91 μ	0	1.2	0	0	0.4	2.2	0
Core total	78	2.40 μ	0.40 μ	28.1 μ	2.94 μ	9.8	0.63 μ	100.0	332

^aData are expressed in nanocuries (nCi) except where marked μ for microcuries.TABLE V. Plutonium in Cores from Fines Chamber^a

Core number	1	2	3	4	5	6	7	8	9
Days exposed	62	62	172	172	292	292	476	476	804
Season	S	S	W	W	S	S	W	W	S
Position	25L	25R	25F	25B	25FL	25BR	25FR	25BL	13F
Top section	0	86 μ	254	8.60 μ	16.1	4.95 μ	8.04 μ	0.7	23.1 μ
Inner sections	0	45	---	---	0.3	43.5	1.9	1.11 μ	---
	---	72	---	76	0	14.5	0.4	126	4.5
	0	8.3	---	---	0	3.1	0.7	44	1.2
	---	40	---	---	0	3.0	0	12.0	---
	0	---	20.2	1.08 μ	0	0.5	0	3.7	0.4
	0	106	---	---	0	1.8	0	2.6	0
	0	3.47 μ	---	15	0	1.9	0	1.9	0
	0	130	0	---	0	2.3	1.1	99.1	---
	---	---	---	0	0	0	0	1.2	0
	0	9.3	0	0	---	0	0.8	1.4	0
	---	---	0	---	0	---	0	2.0	0
	---	---	---	---	---	0	5.5	1.9	0
Inner total	0	3.88 μ	20.2	1.17 μ	0.3	70.6	10.4	1.41 μ	6.1
Bottom section	0	0.85m	0	38	0	1.10 μ	386 μ	697	0
Core total	0	0.94m	2.4	9.81 μ	16.4	6.12 μ	394 μ	2.10 μ	23.1 μ

^aData are expressed in nanocuries (nCi) except where marked μ for microcuries or m for millicuries.

11 903 S 13F	12 903 S 8B	13 1083 W 13FL	14 1083 W 13BR	15 1309 S 13FR	16 1309 S 13BL	17 1463 W 38L	18 1463 W 38R	19 1667 S 38F	20 1667 S 38B	Total
58.1	131 μ	767	8.09 μ	5.16 μ	2.55 μ	20.9 μ	0.07 μ	0	3	200 μ
---	---	---	---	---	31	---	---	18.0	31.7	
---	2.40 μ	11.9	0	777	---	---	0	---	4.2	
0	---	---	---	49.4	184	1.5	---	---	---	
0	19.7	0	0	15.3	---	---	0	0	1.0	
---	17.6	---	---	16.7	61.3	0.8	0	---	1.5	
1.2	---	0	0	38.9	---	---	---	0	---	
---	22.2	0	---	1.1	31.0	0	0.5	---	0.5	
---	---	---	---	---	---	---	---	---	0	
1.2	2.46 μ	11.9	0	898	307	2.3	0.5	18.0	38.9	5.08 μ
19.8	463	1.8	2.0	2.9	8.7	0	3.3	0.4	0	2.42 μ
79.1	134 μ	781	8.09 μ	6.06 μ	2.87 μ	20.9 μ	0.07 μ	18.4	42	208 μ

10 804 S 13B	11 984 W 13L	12 984 W 13R	13 1210 S 13FR	14 1210 S 13BL	15 1364 W 13FL	16 1364 W 13BR	17 1568 S 38L	18 1568 S 38R	Total
206 μ	39.7 μ	2.93m	3.73 μ	5.72m	143 μ	1.69m	475	1.01 μ u	10.86m
104	57.5	3.36 μ	11	3.07 μ	7.4	340	15.8	1.5	
---	12.6	1.08 μ	---	549	2.7	---	4.5	1.0	
25.3	---	16.4	5.6	231	2.7	284	1.2	---	
9.0	1.2	17.9	385	95.1	---	---	0.7	0	
---	0	5.0	---	56.1	0.3	14.3	0.5	0	
2.7	0	13.0	0.5	33.4	---	1.9	0.4	---	
0.7	0.7	0.5	1.2	26.9	0.1	---	0.7	0	
0.5	0	74.1	---	6.6	0	8.1	0.4	---	
0.4	4.8	---	313	21.7	---	---	0	---	
0.4	146	40.4	---	6.1	190	1.5	0	0	
0.5	46.3	2.93 μ	0	62.2	0	14.9	0	0	
0.5	912	37.0	76	83	---	---	0	---	
144	1181	7.57 μ	792	4.24 μ	203	665	24.2	2.5	21.39 μ
3.5	145	426	16	1.25 μ	6.4	27.3 μ	0	0	1.27m
206 μ	41.0 μ	2.94m	4.54 μ	5.73m	143 μ	1.72m	499	1.01 μ	12.15m

low profile had plutonium concentrations that were near or below the detection limit. In the normal distribution, at least 95% of the plutonium was in the top 5 cm of the soil and regularly decreased with depth. Soils with lower fractions of plutonium in the top 5 cm or with marked concentration of plutonium at greater depths were considered abnormal. Most of the profiles in Tables IV and V would be classified abnormal. Essington et al. found causes for abnormality that might apply to the soil in our experiments: removal or covering of the surface soil after deposition of the plutonium, physical or chemical changes with depth, and vertical cracking. The high plutonium content of many bottom sections represents a special case of abnormality caused by a physical change at the deepest soil level.

Core 10 from the chunk chamber was divided into eight equal sections at LFE Environmental Analysis Laboratories.* Sections 1, 2, 4, and 8 were spread on slides for autoradiography. Particles ranged to 0.25- μ m diam, with very few larger than 0.1 μ m. The particle size distributions of the first two sections were very much alike, except that section 2 had three particles larger than 0.15 μ m, whereas the first section had no particles that large. The particle size distributions in sections 4 and 8 were much alike, with diameters averaging less than half of those in sections 1 and 2. Theoretically, the largest particles should be on the surface, with a nearly uniform particle size distribution below the surface. Our data indicate a possible mixing of the soil to 2- or 3-cm depths at the core 10 position (20 cm to the right of the source).

Of particles that formed stars in the autoradiography of widely dispersed material from another chunks chamber core, 25 were examined microscopically for agglomeration.** Each plutonium dioxide particle was associated with one microscopically visible soil particle. Apparently, agglomeration with soil, rather than direct filtration of fine oxide, traps plutonium dioxide particles. This factor may determine which particles are retained by soil and which pass through it. The soil particles in the agglomerates had the same size distribution as the bulk of the soil.

*A. J. Soinski and M. W. Nathans, LFE Environmental Analysis Laboratories, unpublished data, 1975.

**M. W. Nathans, LFE Environmental Analysis Laboratories, unpublished data, 1974.

IV. POSTEXPOSURE EXAMINATION

We examined a moderate-sized chunk, exposed to the simulated humid environment for 8 yr in the chunk chamber. When we removed the chunk from the chamber, it was coated with loosely agglomerated soil. The soil was removed by scraping until only a thin, relatively adherent coating remained.

Photomicrographic examination of a sample of this chunk showed plutonium dioxide chunks were of uniform density with angular, elongated pores. Because of the irregular surface configuration of the sample, it is difficult to be certain whether surface corrosion or spalling of plutonium oxide particles has occurred. However, no specific evidence of such degradation was detected. Many of the near-surface pores were filled with nonmetallic deposits. Electron microprobe analysis of the deposits indicated the presence of potassium, calcium, silicon, aluminum, iron, and sodium, which are common constituents of soil minerals. Their presence in the pores suggests dissolution by the simulated rain and reprecipitation under the influence of the slightly elevated temperature of the plutonium oxide during the dry cycles.

Because the relatively adherent agglomerated soil coating did not survive the polishing process, no estimate of coating thickness could be made.

V. SUMMARY

We exposed fines and larger fragments from an impacted PPO sphere to simulated humid summer and winter conditions on loam soil, in separate environmental test chambers, for 8 yr. The principal mechanism for plutonium release from these sources is the removal by water of the submicroscopic oxide particles on the surface of the PPO. These particles were formed by disruption of the crystal structure by the recoiling daughter nuclei during the alpha decay process. We also observed thermal spalling when the larger and hotter chunks were first hit by rain.

Most of the released plutonium oxide remained in the soil, probably agglomerated with soil particles; however, appreciable amounts of plutonium were found in the rainwater that percolated through the soil. Airborne

plutonium in appreciable amounts was seen only within 2 h after the beginning of a rain on chunks. There was no evidence of plutonium in the environment in any form other than fine particles of oxide. A postexposure photomicrographic examination of a chunk showed that the PPO had elongated pores, some of which were filled with a deposit of common soil constituents.

REFERENCES

1. E. M. Cramer, "Performance of Multihundred-Watt Fueled-Sphere Assemblies in the Safety Sequential Test Program," Los Alamos Scientific Laboratory report LA-5916 (July 1975).
2. F. J. Steinkruger, J. H. Patterson, B. Herrera, G. B. Nelson, G. M. Matlack, G. R. Waterbury, and D. Pavone, "Interaction of a ^{238}Pu Fueled Sphere Assembly with a Simulated Terrestrial Environment," Los Alamos National Laboratory report LA-8597 (February 1981).
3. J. H. Patterson, B. Herrera, G. B. Nelson, G. M. Matlack, and G. R. Waterbury, "Interaction of an Iridium-Clad RTG Heat Source Unit with a Simulated Terrestrial Environment," Los Alamos Scientific Laboratory report LA-6255 (April 1976).
4. W. W. Weir, *Soil Science* (J. B. Lippincott Co., Chicago, 1949), p. 132.
5. R. L. Fleischer and O. G. Raabe, "Fragmentation of Respirable PuO_2 Particles in Water by Alpha Decay—a Mode of 'Dissolution'," *Health Physics* 32, 253-257 (April 1977).
6. R. L. Fleischer, "On the 'Dissolution' of Respirable PuO_2 Particles," *Health Physics* 29, 69-73 (July 1975).
7. R. L. Fleischer and O. G. Raabe, "On the Mechanism of 'Dissolution' in Liquids of PuO_2 by Alpha Decay," *Health Physics* 35, 545-548 (October 1978).
8. J. H. Patterson, E. J. Franzgrote, A. L. Turkevich, W. A. Anderson, T. E. Economou, H. E. Griffin, S. L. Grotch, and K. P. Sowinski, "The Alpha Scattering Experiment on Surveyor 7: Comparison with Surveyors 5 and 6," *J. Geophys. Res.* 74, 6120-6148 (November 1969).
9. E. H. Essington, E. B. Fowler, R. O. Gilbert, and L. L. Eberhardt, "Plutonium, Americium, and Uranium Concentrations in Nevada Test Site Soil Profiles," in *Transuranic Nuclides in the Environment* (International Atomic Energy Agency, Vienna, 1976), p. 157.