INTERIM REPORT

**Quarterly Progress Report
Covering Period April 2
Through June 29, 1979—
The Study of Plutonium Oxide
Leak Rates from Shipping
Containers**

July 1979

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OF PLUTONIUM OXIDE LEAK RATES FROM SHIPPING
CONTAINERS

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QUARTERLY PROGRESS REPORT

April 2 - June 29, 1979

STUDY OF PLUTONIUM OXIDE LEAK RATES FROM SHIPPING CONTAINERSINTRODUCTION

This study was initiated in October 1976, as outlined in the 189 research proposal submitted previously. Several tasks are to be undertaken in this study which, when combined, have the end objective of defining the leak rates of plutonium oxide powder from characterized leaks.

This is the eleventh, and final, quarterly report of this work. Previous reports were issued as BNWL-2260-1, -2, -3, -4, -5, -6, -7, -8, -9, and -10. Each task will be identified and the progress during the reporting period briefly described.

PROGRESS TO DATETASK A -- Review literature and theoretical work relating to transmission of particles through channels

Task objective has been fulfilled, and two reports issued: Supporting Information for the Estimation of Plutonium Oxide Leak Rates Through Very Small Apertures, by L. C. Schwendiman, BNWL-2198; and Transport of Particles Through Gas Leaks -- A Review, by L. C. Schwendiman and S. L. Sutter, BNWL-2218, January 1977.

TASK B -- Investigate the relationship of gas flow rates, leak geometries, pressures and temperatures

Milestone 1. Review literature on topic.

Milestone 2. Report on technical literature.

These milestones were reached and a document, Estimation of Gas Leak Rates Through Very Small Orifices and Channels, by H. J. Bomelburg, BNWL-2223, was issued.

Milestone 3. Select method and design apparatus for flow experiments.

Milestone 4. Fabricate and assemble apparatus.

Milestone 5. Test apparatus.

Milestone 6. Conduct first test.

Milestones 1 through 6 were completed in FY 1977.

Milestone 7. Complete test series (orifices). January 1978.

Milestone 8. Draft Report. March 1978.

Milestone 9. Issue report.

The document Measured Airflow Rates Through Micro-orifices and Flow Prediction Capability, NUREG/CR-0066 (PNL-2611), was issued in July 1978.

Milestone 10. Fabricate microcapillaries.

Milestone 11. Complete test series. April 1978.

Milestone 12. Draft report.

Measured and Predicted Gas Flow Rates Through Rough Capillaries, by P. C. Owzarski, S. L. Sutter, J. Mishima, L. C. Schwendiman and T. J. Bander, NUREG/CR-0745 (PNL-2623), Pacific Northwest Laboratory, Richland, WA, was issued in May, 1979. The document compares experimental flow data through 50- to 250-micrometer-diameter metal capillaries, 0.76 to 2.54 cm long, with data generated by a computer model, Code CAPIL.

TASK C -- Measure transmission of a well-characterized simulant (UO₂ powder) through leaks characterized in Task B

Milestone 1. Pressure vessel for simulating container available for experiments. June 1978.

Milestone 2. Convert airflow apparatus. Completed January 1978.

Milestone 3. First experiment completed.

All experiments were completed in April 1979 and included in the last quarterly report (PNL-2260-10).

In addition, the length-to-diameter (L/D) ratio of the orifices has been investigated further. An L/D ratio of 1 or less had been specified for orificies in Measured Air Flow Rates Through Microorifices and Flow Prediction Capability, NUREG/CR-0066 PNL-2611, by S. L. Sutter, T. J. Bander, J. Mishima and L. C. Schwendiman, Pacific Northwest Laboratory, 1978. However, a nondestructive method to accurately measure the length of the orifice was not

readily available. Use of the depth of focus of an optical microscope indicated that for one 20- μm orifice the length of the orifice could have been up to 193 μm . In the present investigation, one 20- μm orifice was impregnated with resin, sectioned and measured after the powder leak experiments were completed. This single orifice had a leak path length of 560 μm , which would indicate an L/D of 28. This same path length would have an L/D ratio of 2.8 in a 200- μm orifice. Time and funding restraints precluded a comprehensive investigation of the leak path length problem.

Milestone 4. Draft Report.

Completion of the report was delayed, pending statistical analysis of data and decisions pertaining to presentation of data. Questions related to these issues are being resolved and the document finalized.

TASK D -- Measure Fuel Grade PuO_2 Leaks Through a "Standard Leak" Incorporated into a Suitable Container

Milestone 1. Design of experimental equipment. Completed.

Milestone 2. Assembly of experimental system. Completed.

Milestone 3. Simulant experiments. Completed.

Milestone 4. Transfer to glove box. Completed.

Milestone 5. Conduct "hot" experiments. Completed.

Experiments Pu 119 through Pu 127a and Pu 130 and 130a were conducted under conditions that duplicated those of experiments Pu 111 through Pu 118a reported in the tenth quarterly report. Those past and present experiments were conducted with 50- μm orifices. However, shortly after the installation of the orifice used during runs Pu 111 through Pu 118a, the orifice appeared to become plugged as the helium leak drastically decreased, and the observed helium leak rates, as determined by the pressure decay method, were comparable to those characteristics of the 10- and 20- μm orifices used in previous experiments. To allow comparisons to be made between these tenth quarterly report experiments and the more recent 50- μm experiments (runs Pu 119 through Pu 127a and Pu 130 and Pu 130a), the results of runs Pu 111 through Pu 118a are presented in Figure 1 and Table I. As is apparent from inspection of these results, three quarters of the runs exhibited no net PuO_2 emission.

FIGURE 1. Effects of Experimental Conditions on PuO₂ Emissions Through a 50-µm Orifice

TABLE I
SUMMARY OF PuO₂ LEAK RATE EXPERIMENTS USING A 50- μ m ORIFICE

Run Number	Tube Position	Helium Pressure, psig	Vibration	Helium Leak Rate ^(b) scc/sec	Quantity of PuO ₂ Detected, μ g ^(a)	
					Total	Net Total ^(c)
Pu 111	Up	1000	Yes	8.7	0.0006	-0.0007
Pu 111a	Up	1000	Yes	37.2	0.0007	-0.0006
Pu 111b	Up	1000	Yes	2.6	0.0007	-0.0009
Pu 111c	Up	1000	Yes	3.5	0.0022	0.0009
Pu 112	Down	1000	Yes	2.6	0.0179	0.0166
Pu 112a	Down	1000	Yes	3.5	0.0009	-0.0004
Pu 112b	Down	1000	Yes	2.6	0.0005	-0.0008
Pu 113	Up	500	Yes	1.3	0.0014	0.0001
Pu 113a	Up	500	Yes	1.3	-0.0002	-0.0015
Pu 114	Down	500	Yes	1.3	0.0293	0.0280
Pu 114a	Down	500	Yes	1.3	0.0005	-0.0008
Pu 114b	Down	500	Yes	1.3	0.0011	-0.0002
Pu 115	Up	1250	Yes	4.8	0.0034	0.0021
Pu 115a	Up	1250	Yes	4.8	0.0007	-0.0006
Pu 116	Down	1250	Yes	5.4	0.0006	-0.0007
Pu 116a	Down	1250	Yes	5.2	0.0035	0.0022
Pu 117	Up	Ambient	Yes	-	0.0008	-0.0005
Pu 117a	Up	Ambient	Yes	-	0.0019	0.0006
Pu 118	Down	Ambient	Yes	-	0.0059	0.0046
Pu 118a	Down	Ambient	Yes	-	0.0022	0.0009

(a) Based on a specific activity of 0.096 Ci/g for the PuO₂ powder.

(b) Helium leak rate determined by the pressure decay method at the midpoint of the run.

(c) The net total amount above the average containment box background of 0.0013 μ g.

Spare 50- μm orifices were chemically cleaned and microscopically examined to insure that openings were unobstructed and of sizes comparable to the one used in the Pu 111 through Pu 118a experiments. The results of experiments Pu 119 through 127a are presented in Figure 2 and Table II. As is apparent from an examination of Table II, flow rates were not constant between runs, and a significant decrease was observed between runs Pu 120a and Pu 121. It appears that some plugging of the orifice may have occurred between those two runs with the orifice remaining plugged through run Pu 123. Five of the six runs exhibited very low PuO_2 emissions as might be expected for such low flows. The high emission of PuO_2 observed for run Pu 121 may be explained by assuming that the plugging may have occurred during the initial pressurization period when the flow cannot be determined but after the observed PuO_2 was emitted. It should be noted that recordings of the helium flow rates during the course of the runs show only minor flow rate changes. This observation indicates that if, in fact, plugging or unplugging of the orifice is present, these plugging or unplugging conditions probably occur during the initial pressurization or depressurization of the leak tube.

An additional 25 experiments were conducted with the 50- μm -diameter orifice. All of these runs were carried out with a helium pressure of 1000 psi. The results of these runs and the actual experimental conditions are presented in Table III and Figure 3. Experiments Pu 128 through 128d were "zero-time" runs, and Pu 129 through Pu 129d were 60-min runs; otherwise, the run conditions were identical. An inspection of these data reveals that there is no direct relationship between the run time and the total PuO_2 emission, an observation verified by a statistical analysis that indicates that there is no significant difference between the two sets of data.

Runs Pu 131 through Pu 131n (also presented in Table III and Figure 3) were conducted under one set of conditions to allow the data to be examined for a correlation between the helium flow rate and the PuO_2 emissions. No such direct correlation is apparent from a simple inspection of the data; however, the flow rate changes are small and the effects of such a change may not be noticeable. The data are currently being subjected to a more rigorous analysis. Another reason for running this series of experiments is that it

FIGURE 2. Effects of Experimental Conditions on PuO₂ Emissions Through a 50-µm Orifice

TABLE II
SUMMARY OF PuO₂ LEAK RATE EXPERIMENTS USING A 50- μ m ORIFICE

Run Number	Tube Position	Helium Pressure, psig	Vibration	Helium Leak Rate ^(b) scc/sec	Quantity of PuO ₂ Detected, μ g ^(a)	
					Total	Net Total ^(c)
Pu 119	Up	1000	Yes	49.6	12.80	12.80
Pu 119a	Up	1000	Yes	50.0	7.206	7.20
Pu 119b	Up	1000	Yes	50.0	2.4934	2.492
Pu 119c	Up	1000	Yes	50.4	0.9838	0.9825
Pu 119d	Up	1000	Yes	50.7	1.424	1.423
Pu 130	Down	1000	Yes	14.8	0.0065	0.0052
Pu 130a	Down	1000	Yes	103.1	0.0527	0.0514
Pu 120	Up	500	Yes	47.8	7.974	7.973
Pu 120a	Up	500	Yes	47.4	2.468	2.467
Pu 121	Down	500	Yes	7.4	3.427	3.426
Pu 121a	Down	500	Yes	7.4	0.0078	0.0065
Pu 122	Up	500	No	7.4	0.0019	0.0006
Pu 122a	Up	500	No	7.4	0.0011	-0.0002
Pu 123	Down	500	No	7.4	0.0008	0.0005
Pu 123a	Down	500	No	7.4	0.0060	0.0047
Pu 124	Up	1250	Yes	30.3	30.06	30.06
Pu 124a	Up	1250	Yes	32.9	2.924	2.923
Pu 125	Down	1250	Yes	33.7	0.0425	0.0412
Pu 125a	Down	1250	Yes	28.9	0.0336	0.0323
Pu 126	Up	1250	No	35.9	0.0413	0.0400
Pu 126a	Up	1250	No	36.3	0.0275	0.0262
Pu 127	Down	1250	No	31.8	0.0697	0.0684
Pu 127a	Down	1250	No	32.2	2.567	2.566

(a) Based on a specific activity of 0.096 Ci/g for the PuO₂.

(b) Helium leak rate determined using in-line flowmeter. Standard conditions: 1 atm, 25°C.

(c) The net total is the amount above the containment box background of 0.0013 μ g.

FIGURE 3. Effects of Experimental Conditions on PuO₂ Emissions Through a 50-µm Orifice

TABLE III
SUMMARY OF PuO₂ LEAK RATE EXPERIMENTS USING A 50- μ m ORIFICE

Run Number	Tube Position	Helium Pressure, psig	Vibration	Helium Leak Rate ^(b) scc/sec	Quantity of PuO ₂ Detected, μ g ^(a)	
					Total	Net Total ^(c)
Pu 128	Up	1000	Yes	-	12.45	12.45
Pu 128a	Up	1000	Yes	-	0.0287	0.0284
Pu 128b	Up	1000	Yes	-	0.0132	0.0129
Pu 128c	Up	1000	Yes	-	0.1622	0.1619
Pu 128d	Up	1000	Yes	-	0.0063	0.0060
Pu 129	Up	1000	Yes	9.6	0.2905	0.2840
Pu 129a	Up	1000	Yes	13.4	0.1463	0.1398
Pu 129b	Up	1000	Yes	13.0	0.0196	0.0131
Pu 129c	Up	1000	Yes	13.7	0.0997	0.0932
Pu 129d	Up	1000	Yes	14.8	0.0600	0.0535
Pu 131	Up	1000	No	14.5	0.0562	0.0549
Pu 131a	Up	1000	No	13.7	0.1111	0.1098
Pu 131b	Up	1000	No	21.2	0.0149	0.0136
Pu 131c	Up	1000	No	16.3	0.0105	0.0092
Pu 131d	Up	1000	No	14.1	0.0046	0.0034
Pu 131e	Up	1000	No	13.7	0.0046	0.0033
Pu 131f	Up	1000	No	13.0	0.0026	0.0013
Pu 131g	Up	1000	No	11.5	0.0050	0.0037
Pu 131h	Up	1000	No	13.4	0.0040	0.0027
Pu 131i	Up	1000	No	12.6	0.0342	0.0329
Pu 131j	Up	1000	No	13.4	0.0042	0.0029
Pu 131k	Up	1000	No	12.6	0.0066	0.0053
Pu 131l	Up	1000	No	21.2	0.0131	0.0118
Pu 131m	Up	1000	No	20.1	0.0112	0.0099
Pu 131n	Up	1000	No	19.7	0.0038	0.0025

(a) Based on a specific activity of 0.096 Ci/g for the PuO₂ powder.

(b) Helium leak rate determined using in-line flowmeter. Standard conditions: 1 atm, 25°C.

(c) The net total is the amount above the containment box background of 0.0013 μ g for a ten-minute run. Runs Pu 128 thru Pu 128a are "zero time" runs; Pu 129 thru Pu 129a are 60-min runs. All others are 10-min runs.

provides some information concerning the inherent variability of the experiment. The average emission for these runs is 0.0178 μg . The actual PuO_2 emissions ranged between 0.0013 μg and 0.1098 μg .

- Box Background Determination

The average box background used for the calculations of the net total columns of Tables I through III was obtained by averaging all box background measurements made over the course of the experiment. This average value was chosen since the box background measurements seem to be subject to the same lack of reproducibility as the sample runs. Since the background measurements showed no regular pattern of change, it seemed appropriate to use an average value. The overall average for 22 runs is 0.0013 μg with a standard error of 0.0002 μg .

The collection time for most background runs was 12 min, and the value obtained was subtracted directly from the 10-min sample run values to produce the net total numbers. (For a 10-min run, the collection device is actually operating for 12 min although the leak tube is pressurized for only 10 min). Multiples of these values were then used for background subtraction in the case of extended time runs.

However, a series of background runs (B69 through B77), presented in Table IV, indicates that it may not be valid to use such multiple values for the subtraction. As may be seen by inspection, the average for the 12-min runs was 0.0010 μg whereas for the 60-min runs, the average was only 0.0019. Additionally, in some cases the 12-min background values were higher than the 60-min values. For comparison, the overall box background average is 0.0013 μg .

TASK E -- Investigate PuO_2 Leaks Through Simulated Defected Containers

- Milestone 1. Fabricate leaky container. Completed.
- Milestone 2. Simulant tests. Completed.
- Milestone 3. PuO_2 test series. Completed.

A statistical analysis of the results of this completed test series is being performed.

TABLE IV
TIME DEPENDENCE OF BOX BACKGROUND RUNS

Run Number	Run Time (min)	Total PuO ₂ Detected (μg)
B69	12	0.0012
B70	60	0.0002
B71	12	0.00004
B72	60	0.0016
B73	12	0.0022
B74	60	0.0034
B75	12	0.0005
B76	60	0.0006
B77	12	0.0010
Average	12	0.0010
Average	60	0.0019

TASK F -- Develop analysis methodology

This task objective is to provide a calculational technique supported by the experimental programs of Tasks B through D in order to evaluate the potential release of airborne PuO₂ from faulted shipping containers during accidents.

The first part of the calculational technique is a computation method used for gas leaks from the interior sealed container of the Plutonium Air Transport Package (PAT). The TB-1 container of the PAT described in NUREG-0361^(a) serves as the basic model container for these calculations. A computer code, PAGAN (Particulate and Gas Accident Numerics) has been written, utilizing prescribed temperatures from the air, water vapor and gases from decomposed plastic, to provide the gas leakage history for a container such as the TB-1. Time-dependent mass balances are solved as the gas mixture leaks through a specified leak. The principal output from PAGAN is accumulated gas leakage from the TB-1 during the duration of accident conditions.

(a) Plutonium Air Transportable Package Model PAT-1. Safety Analysis Report, NUREG-0361, U.S. Nuclear Regulatory Commission, June 1978.

The second part of the calculational technique is designed to relate powder transmission data to gas leakage. Since the analysis of the data is not complete at this date, no formulas have been finalized to relate powder transmission to gas leakage. These relationships might be incorporated into code PAGAN as planned. However, since the final judgments that relate powder transmission to gas flow might have to rely on paths of reasoning beyond the scope of a computer, the involvement of PAGAN here at this time is uncertain.

The final report on Task F is scheduled for publication in September 1979 as NUREG/CR-0958 PNL-3067: Analysis of Particulate Transmission Through Small Openings Resulting From Container Stresses.

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