INTERIM REPORT

**Quarterly Progress Report
Covering Period October 1
Through December 29, 1978—
The Study of Plutonium Oxide
Leak Rates from Shipping
Containers**

February 1979

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OF PLUTONIUM OXIDE LEAK RATES FROM SHIPPING
CONTAINERS

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QUARTERLY PROGRESS REPORT

October 1 - December 29, 1978

STUDY OF PLUTONIUM OXIDE LEAK RATES FROM SHIPPING CONTAINERS

INTRODUCTION

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

PROGRESS TO DATE

TASK 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. Bommelburg, 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 Microorifices 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.

A new version of computer code CAPIL was written to predict capillary gas flow for both adiabatic (insulated) and maximum heat transfer conditions in the capillary. The original CAPIL code was limited to isothermal flow that closely represented the gas flow situation for long capillaries only. In contrast to the original code, CAPIL now includes entrance and exit pressure losses, which are important for short capillaries. This new version of CAPIL will be used to correlate with the experimental data in the document in preparation, "Measured Airflow Rates Through Tubing of Fine Bore and Flow Prediction Capability."

TASK C -- Measure transmission of a well-characterized simulant (UO_2 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.

Initial experiments to measure the transmission of depleted UO_2 when the leakpath is above (APLA) or below (UPL) the powder level were completed in the third quarter, and further experiments continued into the fourth quarter of FY 1978.

- Leakpaths Above The Static Powder Level With Powder Agitation (APLA)

Uranium measurements from 44 APLA experiments were received during this reporting period. These results are shown in Table A-1 (see Appendix).

The quantity of depleted uranium dioxide (herein signified as DUO) measured is reported as total μg DUO transmitted, μg DUO/min, and μg DUO/cc air.

-- Individual Orifice or Capillary Experiments

Experiments completed during this reporting period included 1) a series of experiments with no attempt to maintain a desired flow by use of a vacuum; 2) "zero time" runs investigating powder flow during pressure buildup and decay; and 3) pressure decay experiments. Each group will be described briefly.

1) In the initial experiments, the known airflow rate at the designated pressure for the orifice was maintained using a vacuum. This compensated for the buildup of backpressure that would lower the flow below the value characterized in the flow experiments. In contrast, the present experiments did not use a vacuum to maintain flow and exhibited flows 6% to 98% of the characterized orifice flow. The uranium measurements received indicate comparable or even greater powder transmissions than those transmissions when the flow was maintained. These values are shown in Table I, which compares DUO transmitted for both experimental series and includes the measured flow as percent of characterized flow.

TABLE I
DUO TRANSMITTED THROUGH ORIFICES,
UNAIDED AND AIDED AIR FLOW

<u>psig</u>	<u>Aided Flow DUO $\mu\text{g}/\text{min}$</u>	<u>Unaided Flow* DUO $\mu\text{g}/\text{min}$</u>	<u>% of Characterized Flow</u>
<u>200 μm Orifice</u>			
1000	616 $\pm$ 443	334 $\pm$ 250	84, 87
500	46 $\pm$ 50	25.4 $\pm$ 11	98, 94
30	2.7 $\pm$ 2	5.6	86
<u>110 μm Orifice</u>			
1000	22.4 $\pm$ 9.2	109 $\pm$ 10	63, 6

* Average of 2 runs.

The powder transmitted was significantly greater through the 110- μ m orifice with a loss of volumetric flow of as much as 94%.

Plots correlating powder transmissions with known gas leak rates⁽¹⁾ were updated to include results received in this quarter (see Figures 1 and 2). The same trend to a finite amount of uranium transmitted in each cc of air and increased μ g/min DUO at the higher flow rates continued.

2) "Zero time" runs investigated the flow of powder into the powder collection chamber during the time required to bring the vessel to the experimental pressure and to return the vessel to ambient pressure. The flow during these times cannot be separated experimentally. The time required to pressurize and depressurize the vessel is short and is a function of pressure rather than orifice size since the bulk of the air exits through a throttling valve at the top of the vessel. Therefore, the time required is similar for all orifices at one pressure. Typical time requirements for pressurization/depressurization of the APLA vessel are shown in Table II.

Results from a few experiments indicate that a large portion of the powder can be leaked during this pressurization/depressurization time. The total DUO transmitted regardless of time through two orifices of different sizes is compared to "zero time" transmittance in Table III. The results are plotted as a function of airflow rate for 1000 psig with the variance in Figure 3. An experiment matrix was run in an attempt to quantify the amount of powder transmitted during pressurization/depressurization of the vessel.

3) The pressure decay experiments measured powder transmission under a decaying pressure regime. The vessel was pressurized to 1000 psig and allowed to depressurize with the entire flow passing through the orifice. The results of these experiments are tabulated in Table IV.

The pressure decay results are slightly higher than the "zero time" leakage. The total times required were also greater, 27 to 132 min, as compared to about 3 min for the "zero time" runs.

FIGURE 1. APLA Powder Flow Rates Through Characterized Leaks as a Function of Airflow Rates, $\mu\text{g/cc}$

FIGURE 2. APLA Powder Flow Rates Through Characterized Leaks as a Function of Airflow Rates, $\mu\text{g}/\text{min}$

TABLE II
TIMES REQUIRED FOR PRESSURIZATION/DEPRESSURIZATION
OF APLA VESSEL

<u>Pressure psig</u>	<u>Time to Reach Pressure, sec</u>	<u>Total Time for Pressurization/ Depressurization, sec</u>
1000	27 ± 3	190 ± 7
500	19 ± 4	132 ± 8
100	11 ± 2	68 ± 7
30	9.3 ± 2	40 ± 6

TABLE III
DUO TRANSMITTED DURING "ZERO TIME" RUNS COMPARED
WITH THE AVERAGE TOTAL DUO TRANSMITTED

<u>Nominal Orifice Size</u>	<u>Pressure psig</u>	<u>"Zero Time" Total DUO Transmitted μg</u>	<u>Average Total DUO Transmitted μg</u>
200	1000	4740	5912 ± 2334
110	1000	218	225 ± 165
200	500	1680	1417 ± 300
200	30	180	170 ± 108

TABLE IV
DUO TRANSMITTED IN PRESSURE DECAY
AND "ZERO TIME" RUNS

<u>Nominal Orifice Size</u>	<u>Pressure Decay: DUO Transmitted μg</u>	<u>"Zero Time": DUO Transmitted μg</u>
200	6850	4740
110	449	218
	1380

FIGURE 3. Total DUO Transmitted in all 1000 psig APLA Experiments Compared with "Zero Time" DUO Transmission

-- Multiple Orifice and Capillary Experiments

Analytical results from capillary experiments indicated that the short 78- μm capillary had about the same DUO powder transmission as the nominal 63.5- μm -diameter orifices and significantly less than the 110- μm -diameter orifice. The long 50-micron-diameter capillary transmitted the same DUO as the 36- μm orifices. One experiment sampled the same aerosolized powder simultaneously through the 150- μm long and short capillaries and

the 200-micron orifice. The DUO transmitted through the long capillary and the orifice were comparable, and the short capillary had about a third of this amount of DUO transmitted. The capillaries subsequently broke, and further investigation of capillary powder flow will be attempted when replacements are received.

- Statistical Validation

A matrix of experiments (36) has been completed and the collected samples sent for chemical analysis. The 36 experiments are composed of six different orifices with three pressures and three replicates for each of the six orifices. The 18 "zero time" experiments are composed of three different orifices with three pressures and three replicates for each of the orifices. These experiments will give a statistical analysis of the APLA experiments, the goals of which are:

1. Estimate the experiment-wide reproducibility.
2. Estimate uniformity of aerosolization.
3. Estimate reproducibility of aerosolization.
4. Quantify the magnitude of pressurization and depressurization effects.

This matrix of experiments is an analysis of the variance with replicate treatments. All of these experiments were run for 30 min, since indications are that time has little effect on DUO transmissions. Time was held constant to simplify the analysis. At this time results have not indicated significant differences in DUO leak rates through the 20-, 36-, and 63.5- μ m orifices, so the 36-, 110-, and 200-micron orifices were selected to be used.

- Other Considerations

Some of the orifices changed size during the experiments. The sizes of the orifices as they were used in the matrix are shown in Table V and compared to the original orifice size.

Photomicrographs of the orifices were taken after every run so that changes with use can be monitored and considered when results are evaluated. The present airflow rates through all the orifices are now being measured, and replacement orifices with original diameters have been ordered.

TABLE V
ORIGINAL AND PRESENT ORIFICE SIZE

<u>Orifice Designation</u>	<u>Original Diameter μm</u>	<u>Present Diameter μm</u>
3-30	33	22
2-36	33	32
1-110	100	111
3-110	100	89
1-200	200	226
3-200	200	190

The design of the sampling system has been modified as shown in Figure 4. This design allows the sampler to operate at high pressure, shutting off immediately downstream of the sample collection chamber. The equipment has been designed and installed.

FIGURE 4. APLA Sampling System Modification

After 103 APLA experiments, 800 g DUO were added to the vessel. All of the APLA DUO powder was replaced after 188 experiments. The size distribution of the material removed was the same as the original DUO powder.

An order has been placed for replacement capillaries. The nominal 50, 100, and 150 capillaries, four of each length, were selected for further investigation.

- Leakpath Underneath the Static Powder Level (UPL)

Measurements of DUO transmitted through leakpaths underneath the powder level received in this quarter are tabulated in Table A-2 in the Appendix.

The objective of this study is to correlate powder transmission with gas-leak rates. In the UPL experiments the airflow rate was measured, and no attempt was made to maintain a specific air-leak rate. The measured leak rates during the experiments ranged from not detectable to values comparable to the characterized leak rates. In all cases, a quantifiable amount of DUO powder passed through the leak. Therefore, to correlate the powder transmission with gas-leak rates, the characterized gas-leak rate at each pressure for the orifice was selected. The $\mu\text{g/cc}$ and $\mu\text{g/min}$ are plotted as a function of the characterized leak rate in Figures 5 and 6. Since the agitation (with vibrascribe) did not have apparent effect, the results of the agitated and not-agitated experiments have been averaged.

The plot of the $\mu\text{g/cc}$ shows a maximum at the flow rate corresponding to the 100 psig pressure (except for the 200-micron orifice) for each of the nominal sizes. The $\mu\text{g/min}$ also shows a maximum at the 100-psig flow rate, with the same exception of 200 μm . This maximization of flow at 100 psig might be accounted for by arching and breaking up of these arches favored by the 100-psig pressure.

The 200-micron orifice seems to fit into a different flow regime, which might be accounted for by the orifice-diameter/particle-diameter ratio. In gravity free-flow of solids, it has been suggested that the internal diameter of a conduit should exceed 5 to 7 times the largest particle diameters where these particles are present in high proportions.⁽²⁾ The largest particle size in the DUO powder is about 20 microns, which would require a 100- to 140- μm

FIGURE 5. DUO Transmitted Through Leak Paths Underneath the Static Powder Level, μg/cc

FIGURE 6. DUO Transmitted Through Leak Paths Underneath the Static Powder Level, $\mu\text{g}/\text{min}$

orifice diameter to meet this criterion. Thus, the 200-micron orifice would be sufficiently large to allow freer particle flow, if the orifice criterion held true here.

There has been some published information on the prediction of flow rates for powders in gravity free-flow through restrictions.⁽²⁾ For 2-in. orifices, the flow of bead or pellet catalyst in pounds per second fits the equation:

$$W_A = 0.156 D_o^{2.84}$$

where

W_A = solids flow, pounds per second

D_o = orifice diameter, inches

Flow from a leak below the powder level at low pressures might relate to gravity flow of powder, and thus this equation could be used to predict powder flow. To evaluate this possibility, some powder flow predictions were made using this formulation. A 200- μ m-diameter orifice, where the orifice-diameter/particle-diameter (OD/PD) ratio is such that it could permit free flow, and the 125- μ m orifice where some restriction might be anticipated, were selected for flow predictions. The results of these calculations are shown in Table VI, and compared to low-pressure flow measured through the orifices.

The results in Table VI show that the calculated flow was several orders of magnitude larger than the measured flow. This equation was formulated from measurements using a bead catalyst. It is evident that the equation cannot be applied to DUO. The fine particle sizes of the DUO give the powder a tendency to stick and agglomerate, whereas the larger bead catalyst would tend to flow freely.

TABLE VI
DUO TRANSMITTED USING GRAVITY FREE FLOW EQUATION

<u>Orifice Diameter</u>	<u>Calculated DUO Flow μg/min</u>	<u>15 psig DUO Flow μg/min</u>	<u>5 psig DUO Flow μg/min</u>
200	4499	9.9	0.1
125	1180	0.5	

An experimental matrix has been completed which investigates pressure and depth effects on powder flow from a leak underneath the powder level. A new powder reservoir of the same internal diameter as the one used in earlier work, enables powder to cover the orifice to a maximum depth of 8.6 in. (See Figure 7.) Three pressures, 100, 500, and 1000 psig, were investigated. Three initial depths of DUO, 8.6, 2.9, and 0.6 in., cover the orifice. The powder is weighed; the packing is a pour volume and loose. Because of packing discrepancies, the weight at maximum packing shows a variability, but is approximately 250 g. The 2.9 in. depth is 100 g; the 0.6 in. depth is 25 g. The powder exhibits compaction during the run, both at 100 psig and 1000 psig. For all pressures, a volume with a depth of 8.6 in. before the run can be 4 in. after a run. Thus, the final depth is about half the original depth. This compacted powder is caked, difficult to remove, and must be scraped out of the chamber. The volumetric airflow rate is not changed as the packing occurs during a run; however, for the deepest powder, a depressurization time of about 40 min is required.

The DUO powder collected from these runs has been sent for chemical analysis.

FIGURE 7. New UPL Powder Reservoir

The UPL and APLA leak rates have been compared in Figure 8 as a function of pressure for all of the orifices. The leak rates are comparable, except the APLA leak rates do not show the trend to maximize leakage at 100 psig exhibited in UPL experiments.⁽³⁾ The phenomena that relate to this leakage, i.e., the degree of arching and bed characteristics, might not be operating in the APLA leak rates.

FIGURE 8. Comparison of Powder Transmitted Through Leaks Above (APLA) and Below (UPL) the Static Powder Level

TASK D -- Measure fuel grade PuO₂ 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 glovebox. Completed.
- Milestone 5. Conduct "hot" experiments.

- Conduct "Hot" Experiments (Phase One)

As discussed in the June, 1978 proposal for extended work (Proposal No. 585-J-4069 R), the data at the bounding limits of each parameter must be adequately measured in order to determine the effects of the parametric interactions. An internal data point must also be provided to determine whether the relationships are linear or curvilinear. In light of such considerations, a series of 38 tests were defined to complete the four-parameter matrix (size/type, position, pressure, vibration).

The experiments conducted during this quarter satisfied the requirements for the four-parameter matrix completion. Additionally, extension experiments were begun using an 8- μ m orifice.

The data for the 5- μ m orifice experiments were presented in the eighth quarterly progress report. During the present reporting period, experiments were conducted using the 10- and 20- μ m orifices.

-- 10- μ m Orifice

The 10- μ m orifice experiments were planned to provide the internal data point necessary for the matrix completion. The helium leak rate, as measured by the pressure decay method at the mid-point of each run, varied between 0.4 and 0.8 cc/sec at a pressure of 440 psig He, whereas the observed PuO₂ emission varied between 0 and 10.03 nanograms. The actual experimental conditions under which these runs were conducted and the results of the 10- μ m experiments are shown in Table A-3 (see Appendix) and Figure 9.

FIGURE 9. Effect of Experimental Conditions on PuO_2 Emissions Through a 10- μm Orifice

-- 20- μm Orifice

Data at the bounding limits of the four-parameter matrix were provided by the 5- and 20- μm experiments. (As already noted, the data for the 5- μm orifice experiments were presented in the eighth quarterly progress report.) The 20- μm experiments were conducted during the present reporting period. Experiments were conducted at ambient pressure and at 920 psig He. The leak rates at 920 psig He varied between 3.91 and 14.1 cc/sec with an average of 5.41 cc/sec. The observed PuO_2 emissions varied between 0 and 5.36 nanograms at ambient pressure and between 0 and 22.08 nanograms at 920 psig He. The experimental conditions, together with the results are shown in Table A-4. Figure 10 shows the results graphically.

FIGURE 10. Effect of Experimental Conditions on PuO_2 Emissions Through a 20- μm Orifice

-- Summary of Matrix Experiment

Table A-5 provides a summary of the data for the 5-, 10- and 20- μm orifice experiments in the format of a four-parameter matrix (size/type, position, pressure, vibration). The entries not marked with an (a) represent the initial data, referred to as Replicate 1 for the purposes of discussion of the statistical analysis. The matrix completion experiments are referred to as Replicate 2.

Certain characteristics of the data in Table A-5 are obvious. The most noticeable is the extreme variability within some, but not all, cells.

There is also a great deal of variability among cell averages, most of which does not follow a consistent pattern. Another obvious characteristic is the disparity between replicates. Instead of confirming or amplifying conclusions drawn from Replicate 1 data, the Replicate 2 data seem to weaken or contradict those conclusions. These characteristics are also borne out by the array of cell means and standard deviations presented in Table A-6 (for Replicate 1 data) and Table A-7 (for Replicate 2 data), respectively.

Although the main features of the data base are apparent from visual examination, for the sake of thoroughness the data were analyzed by standard statistical methods. Cell means were plotted against cell standard deviations (using Replicate 1 data only) on log-log graph paper, and a definite linear trend was observed. On the strength of this evidence, which is limited by the relatively small sample size, the data were converted to logarithms prior to additional analysis.

In order to determine with reasonable assurance whether the observed difference between Replicate 1 and Replicate 2 data was real or imaginary, an objective statistical test was used. The mechanics of the test were a two-way analysis of variance with repeated measurements in unequal subgroups.^(a) The two factors tested in this analysis were replicate and block. The first factor represented the changes in laboratory techniques or conditions that took place between the time the Replicate 1 data were being generated and the time the Replicate 2 data were being generated. The second factor represented the changing test conditions under which the two replicates had to be compared. The analysis showed that the difference between replicates, and the interactions of replicate and block, were statistically significant. These results imply that in other kinds of statistical analysis it would be unwise to pool data from both replicates.

The difference between replicates, as indicated, weakens considerably the prospects for showing functional relationships between total PuO_2 leak

(a) Subprogram ANOVA in the SPSS package of computer programs was used in performing the calculations.

and the experimental parameters. The findings here, based on fragmentary kinds of statistical analysis afforded by an incomplete and unbalanced design matrix, can be summarized by the following list of tentative conclusions:

1. The PuO_2 particulate leak rate generally increased with orifice size, internal gas pressure, and vibration.
2. The average emission was greatest, in most cases, when the leak tube was in the down position.
3. There is no significant difference between the PuO_2 emission through a 5- μm orifice and through a 50- μm Internal Diameter (ID) x 4.4-cm long capillary.
4. Cell standard deviations are directly proportional to cell means.
5. Tests of statistical significance should be preceded by a logarithmic transformation of the experimental data.
6. The difference between cell means is statistically significant in only a few of the many possible cell comparisons.
7. Two-way, three-way, and four-way interaction effects were to complicate the functional relationship between internal gas pressure and PuO_2 particulate leak rate.
8. The present data base is insufficient to determine whether the relationships of interest are linear or curvilinear.
9. The new data (Replicate 2) do not agree with the old data (Replicate 1). Statistical analysis shows a significant difference between the two replicates.

-- Correlation Data

The experiment for showing the effect on correlation by varying four parameters (position, pressure, vibration, and orifice size) produced the data in Table A-8. The entries that are not marked with an (a) represent original tests, referred to as Replicate 1. The entries marked with an (a) represent follow-up tests, referred to as Replicate 2.

Analysis of the data in Table A-8 paralleled the analysis of total PuO_2 leak data in Table A-5. Certain characteristics of the raw data were obvious from inspection and were verified by numerical analysis.

Cell means and standard deviations are in Table A-9 (for Replicate 1) and Table A-10 (for Replicate 2), respectively. Cell means were plotted against cell standard deviations (using Replication 1 data only) on log-log graph paper, and a definite linear trend was observed. Just as a logarithmic transformation was indicated for total PuO_2 leak, a logarithmic transformation was indicated here.

The difference between Replicate 1 data and Replicate 2 data was tested, as before, by an analysis of variance. The results were completely analogous to those obtained for total PuO_2 leak. As a consequence, the prospects for showing functional relationships between correlation and the experimental parameters are weakened considerably. The findings, based on fragmentary kinds of statistical analysis afforded by an incomplete and unbalanced design matrix, can be summarized by the following list of tentative conclusions:

1. The correlation is significantly lower for ambient pressure than for either 440 psig or 920 psig.
2. Increasing pressure from 440 psig to 920 psig has no significant effect on the correlation.
3. There is no consistent, observable effect by varying position, vibration, or orifice size.
4. Tests of statistical significance should be preceded by a logarithmic transformation of the experimental data.
5. The new data (Replicate 2) are not consistent with the old data (Replicate 1).

-- 8- μm Orifice

Experiments were planned using an 8- μm orifice in order to establish another point on the curve for particulate leakage as a function of orifice diameter and to provide additional data on the pressure dependency of the

PuO_2 emissions. Prior to the initiation of these runs, the vacuum gauges were re-calibrated, corrected at mid-range (750 psig), and new calibration curves produced. A pressure of 1000 psig now reads 995 psig, whereas prior to the mid-range correction the pressure read 920 psig. Runs were conducted at pressures of 995 and 1230 psig He with vibration and with the tube position up, down, and sideways. In addition to the standard 10-min runs, one- and two-hour runs were also carried out. Run conditions and results of the runs are shown in Table A-11. The results of the runs are shown graphically in Figures 11 and 12.

FIGURE 11. Effect of Experimental Conditions on PuO_2 Emissions Through an 8- μm Orifice at 995 psig He

FIGURE 12. Effect of Experimental Conditions on PuO₂ Emission Through an 8-µm Orifice at 1230 psig He

TASK E -- Investigate PuO₂ leaks through simulated defected containers

Milestone 1. Fabricate leaky container. Completed.

Milestone 2. Simulant tests. Completed.

Milestone 3. PuO₂ test series. Completed.

A statistical analysis of the results of this completed test series is being performed. More experiments will be identified when this analysis is accomplished.

REFERENCES

1. L.C. Schwendiman, et al. 1978. Quarterly Progress Report, July 1 through September 29, 1978 -- Study of Plutonium Dioxide Leak Rates from Shipping Containers. PNL-2260-8, Pacific Northwest Laboratory, Richland, WA 99352.
2. Frederick A. Zenz. 1960. Fluidization and Fluid Particle Systems. Reinhold Publishing Corporation, New York, p. 136.
3. L.C. Schwendiman, et al. 1978. Quarterly Progress Report, April 1 through June 30, 1978 -- Study of Plutonium Dioxide Leak Rates from Shipping Containers. PNL-2260-7, Pacific Northwest Laboratory, Richland, WA 99352.

APPENDIX

TABLE A-1
DEPLETED URANIUM DIOXIDE TRANSMITTED THROUGH
WELL-CHARACTERIZED LEAKS

APLA	ORIFICE OR CAPILLARY DESIGNATION	MEASURED DIAMETER μm	CHAMBER PRESSURE psig	TIME, min	AIRFLOW cc/min	TRANSMITTED DUO, $\mu\text{g}^{(1)}$	DUO, $\mu\text{g}/\text{min}$	DUO, $\mu\text{g}/\text{cc}$
56								
56-1	1-110	100	500	10	3060	23.3 $\pm$ 7	2.3	8 $\times 10^{-4}$
56-2	2-110	125	500	30	4500	119 $\pm$ 36	4.0	9 $\times 10^{-4}$
56-3	3-110	100	500	60	3100	55.3 $\pm$ 17	0.9	3 $\times 10^{-4}$
57	1-50	48	500	30	195	25 $\pm$ 7.5	0.8	4 $\times 10^{-3}$
58	250S	276	500	30	13300	675 $\pm$ 200	22.5	2 $\times 10^{-3}$
59								
59-1	1-110	100	500	30	3060	4.3 $\pm$ 1.8	0.1	5 $\times 10^{-5}$
59-2	2-110	125	500	30	4500	112 $\pm$ 34	3.7	8 $\times 10^{-4}$
59-3	3-110	100	500	30	3100	59 $\pm$ 18	2.0	6 $\times 10^{-4}$
60	200S	228	30	60	4800	13 $\pm$ 3.9	0.2	5 $\times 10^{-5}$
61	1-50	48	30	60	4	8.75 $\pm$ 2.6	0.2	4 $\times 10^{-2}$
62	100S	97	1000	10	4800	22.3 $\pm$ 6.7	2.2	5 $\times 10^{-4}$
63	200S	228	1000	10	25300	3.95 $\pm$ 1.8	0.4	2 $\times 10^{-5}$
64								
64-1	1-110	100	30	30	245	52.2 $\pm$ 16	1.7	7 $\times 10^{-3}$
64-2	2-110	125	30	60	365	52.4 $\pm$ 16	0.9	2 $\times 10^{-3}$
64-3	3-110	100	30	120	245	18.4 $\pm$ 5.5	0.2	6 $\times 10^{-4}$
65	2-75S	78	1000	10	2100	33.7 $\pm$ 10	3.4	2 $\times 10^{-3}$
66								
66-1	1-110	100	30	60	245	3.59 $\pm$ 1.8	0.1	2 $\times 10^{-4}$
66-2	2-110	125	30	60	365	36.0 $\pm$ 11	0.6	2 $\times 10^{-3}$
66-3	3-110	100	30	60	245	44.2 $\pm$ 13	0.7	3 $\times 10^{-3}$
67	1-50	48	1000	10	400	143 $\pm$ 43	14.3	4 $\times 10^{-2}$

⁽¹⁾ THE $\pm$ IS THE UNCERTAINTY IN THE URANIUM ANALYSIS AT THE 2 σ CONFIDENCE LEVEL

TABLE A-1. (contd)

APLA	ORIFICE OR CAPILLARY DESIGNATION	MEASURED DIAMETER μm	CHAMBER PRESSURE psig	TIME, min	AIRFLOW cc/min	TRANSMITTED DUO, μg	DUO, $\mu\text{g/min}$	DUO, $\mu\text{g/cc}$
70	100B	99	500	30	1560	3.8 ± 1.8	0.1	8×10^{-5}
71	250S	276	30	60	1650	217 ± 65	3.6	2×10^{-3}
73	100B	99	30	30	130	4.96 ± 1.8	0.2	1×10^{-3}
75	1-110	100	100	30	640	19.3 ± 5.8	0.6	1×10^{-3}
77	2-63.5	61	100	30	225	11.7 ± 3.5	0.4	2×10^{-3}
78	250S	276	100	30	4500	188 ± 56	6	1×10^{-3}
79								
79-1	1-200	200	30	60	1000	323 ± 97	5	5×10^{-3}
79-2	2-200 (310)		30	60	960	9.11 ± 2.7	0.2	2×10^{-4}
79-3	3-200	200	30	60	1000	108 ± 32	1.8	2×10^{-3}
80	1-20	22	100	30	25	6.7 ± 2	0.2	9×10^{-3}
81	1-36	43	100	30	77	4.74 ± 1.8	0.2	2×10^{-3}
82								
82-1	1-200	200	30	30	1000	62.1 ± 13	2.1	2×10^{-3}
82-2	2-200	200	30	60	$960^{(2)}$			
82-3	3-200	200	30	90	1000	38.4 ± 9.4	0.4	4×10^{-4}
85	1-200	200	100	30	2300	80.8 ± 15	2.7	1×10^{-3}
88	150S	182	100	30	1640	320 ± 43	10.7	7×10^{-3}
90								
90-2	250S	276	500	30	20000	11500 ± 1400	383	2×10^{-2}
97								
97-2	250S	276	1000	10	38800	15800 ± 2100	1580	4×10^{-2}
98								
98-1	150B	176	500	30	5600	959 ± 140	32	6×10^{-3}
98-2	150S	182	500	30	7400	267 ± 37	8.9	1×10^{-3}
98-3	3-200	200	500	30	11000	942 ± 140	31	3×10^{-3}

(2) PLUGGED

TABLE A-1. (contd)

APLA	ORIFICE OR CAPILLARY	MEASURED DIAMETER μm	CHAMBER PRESSURE psig	TIME, min	AIRFLOW cc/min	TRANSMITTED DUO, μg	DUO, $\mu\text{g}/\text{min}$	DUO, $\mu\text{g}/\text{cc}$
100	1-110	100	1000 PRESSURE DECAY			449 $\pm$ 58		
102	2-110	125	1000 PRESSURE DECAY			1380 $\pm$ 190		
104								
1	1-200	200	1000	10	18800	5690 $\pm$ 960	569	3 $\times 10^{-2}$
2	3-200	200	1000	10	22300	1880 $\pm$ 250	188	8 $\times 10^{-3}$
105	1-200	200	1000 PRESSURE DECAY			2670 $\pm$ 340		
112	3-110	100	1000	10	3650*	98.8 $\pm$ 17	9.9	3 $\times 10^{-3}$
113	3-200	200	1000	10	18500*	5110 $\pm$ 880	511	3 $\times 10^{-2}$
118	3-200	200	500	30	10800*	535 $\pm$ 68	17.8	2 $\times 10^{-3}$
129	3-200	200	30	30	860*	167 $\pm$ 25	5.6	6 $\times 10^{-3}$
133	3-110	100	1000	10	335*	113 $\pm$ 19	11.3	3 $\times 10^{-2}$
134	3-200	200	1000	10	19200*	1570 $\pm$ 210	157	8 $\times 10^{-3}$
140	3-200	200	500	30	10300*	991 $\pm$ 120	33	3 $\times 10^{-3}$
146	1-200	200	1000 PRESSURE DECAY			6850 $\pm$ 820		
164	1-200	200	30	0		180 $\pm$ 27		
166	3-20	23	500	0		452 $\pm$ 58		
171	3-110	100	1000	0		218 $\pm$ 31		
172	3-200	200	1000	0		4740 $\pm$ 580		
173	1-200	200	500	0		1680 $\pm$ 220		

* MEASURED ONLY

TABLE A-2
DEPLETED URANIUM DIOXIDE TRANSMISSION RATES
FOR LEAK PATHS UNDER THE POWDER LEVEL

UPL	ORIFICE OR CAPILLARY DESIGNATION	MEASURED DIAMETER μm	CHAMBER PRESSURE psig	AGITATION	TIME, min	AIRFLOW cc/min	TRANSMITTED DUO, $\mu\text{g}^{(1)}$	DUO, $\mu\text{g}/\text{min}$	DUO, $\mu\text{g}/\text{cc}$
80	2-200	200	100	NO	30	760	1240 $\pm$ 370	41	5 $\times 10^{-2}$
81	2-200	200	100	YES	30	540	709 $\pm$ 210	24	4 $\times 10^{-2}$
82	2-36	33	100	NO	30	95	24.6 $\pm$ 7.4	0.82	9 $\times 10^{-3}$
83	2-36	33	100	YES	30	9	19.8 $\pm$ 5.9	0.66	7 $\times 10^{-3}$
84	2-110	125	100	NO	30	490	236 $\pm$ 71	8	2 $\times 10^{-2}$
86	2-20	20	100	NO	30	ND	15.5 $\pm$ 4.7	0.5	
89	150S	182	100	NO	30	750	479 $\pm$ 62	16	2 $\times 10^{-2}$
90	250S	276	100	NO	30	1050	1730 $\pm$ 230	58	6 $\times 10^{-2}$
91	1-200	200	50	NO	60	495	153 $\pm$ 24	3	5 $\times 10^{-3}$
92	250S	276	100	NO	30	775	5370 $\pm$ 650	179	2 $\times 10^{-1}$
93	150S	182	100	YES	30	890	443 $\pm$ 58	15	2 $\times 10^{-2}$
96	1-200	200	50	YES	60	480	3320 $\pm$ 410	55	1 $\times 10^{-1}$
97	2-110	125	50	YES	60	180	70.1 $\pm$ 14	1	6 $\times 10^{-3}$
98	250S	276	50	NO	60	360	2440 $\pm$ 310	41	1 $\times 10^{-1}$
100	250S	276	50	YES	60	365	3150 $\pm$ 390	53	9 $\times 10^{-1}$
101	150S	182	50	NO	60	250	113 $\pm$ 19	2	4 $\times 10^{-1}$
104	1-200	200	15	NO	60	99	734 $\pm$ 91	12	1 $\times 10^{-1}$
105	250S	276	15	YES	60	155	481 $\pm$ 62	8	5 $\times 10^{-2}$
106	2-110	125	1000	NO	10	3800	1760 $\pm$ 230	176	5 $\times 10^{-2}$
107	250S	276	1000	NO	10	1000	3.81 $\times 10^4 \pm 4.7 \times 10^3$	3810	3.81
108	2-110	125	1000	NO	10	2930	544 $\pm$ 69	54	2 $\times 10^{-2}$

(1) THE $\pm$ IS THE UNCERTAINTY IN THE URANIUM ANALYSIS AT THE 2 σ CONFIDENCE LEVEL

A-4

TABLE A-3
SUMMARY OF PuO₂ LEAK RATE EXPERIMENTS USING A 10-μm ORIFICE

Run Number	Tube Position	Helium Pressure, psig	Vibration	Helium Leak Rate, (b) cc/sec	Quantity of PuO ₂ Detected, μg ^(a)			
					Inlet Nozzle	Final Filter	Total	Net ^(c) Total
B 58 ^(d)	Sideways	---	---	---	0.00055	0.00031	0.00086	---
B 59 ^(d)	Sideways	---	---	---	0.00169	0.00078	0.00247	---
Pu 87	Down	440	Yes	0.4	0.00052	0.000654	0.00707	0.00541
Pu 87a	Down	440	Yes	0.4	0.00057	0.00132	0.00188	0.00022
Pu 87b	Down	440	Yes	0.4	0.00075	0.00117	0.00192	0.00026
Pu 87c	Down	440	Yes	0.4	0.00123	0.00543	0.00657	0.00491
Pu 88	Down	440	No	0.4	0.00521	0.00141	0.00662	0.00496
Pu 88a	Down	440	No	0.4	0.00383	0.00135	0.00518	0.00350
Pu 88b	Down	440	No	0.4	0.00018	0.00009	0.00027	0.00000
Pu 88c	Up	440	No	0.4	0.00092	0.00030	0.00122	0.00000
Pu 89	Up	440	No	0.8	0.00070	0.01100	0.01169	0.01003
Pu 89a	Up	440	No	0.8	0.00150	0.00090	0.00240	0.00074
Pu 89b	Up	440	No	0.8	0.00058	0.00205	0.00263	0.00097
Pu 98c	Up	440	No	0.8	0.00043	0.00042	0.00085	0.00000

(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 mid-point of the run

(c) The net total is the amount above the average containment box background of 0.0166 μg

(d) Containment box background

TABLE A-4
SUMMARY OF PuO₂ LEAK RATE EXPERIMENTS USING AN 20- μ m ORIFICE

Run Number	Tube Position	Helium Pressure, psig	Vibration	Helium Leak Rate, (b) cc/sec	Quantity of PuO ₂ Detected, μ g (a)			
					Inlet Nozzle	Final Filter	Total	Net Total (c)
B 60b ^(d)	Sideways	---	---	---	0.00102	0.00014	0.00116	---
B 61 ^(d)	Sideways	---	---	---	0.00198	0.00031	0.00229	---
Pu 90	Up	Ambient	Yes	---	0.00123	0.00585	0.00708	0.00536
Pu 90a	Up	Ambient	Yes	---	0.00170	0.00051	0.00221	0.00049
Pu 91	Down	Ambient	Yes	---	0.00135	0.00112	0.00246	0.00074
Pu 91a	Down	Ambient	Yes	---	0.00087	0.00070	0.00156	0.00000
Pu 92	Up	Ambient	No	---	0.00217	0.00073	0.00290	0.00118
Pu 92a	Up	Ambient	No	---	0.00018	0.00050	0.00068	0.00000
Pu 93	Down	Ambient	No	---	0.00071	0.00364	0.00435	0.00263
Pu 93a	Down	Ambient	No	---	0.00095	0.00046	0.00141	0.00000
Pu 94	Up	920	Yes	6.52	0.00220	0.01677	0.01898	0.01726
Pu 94a	Up	920	Yes	3.91	0.00028	0.00034	0.00063	0.00000
Pu 94b	Up	920	Yes	3.91	0.00043	0.00098	0.00141	0.00000
Pu 94c	Up	920	Yes	3.91	0.00098	0.00097	0.00195	0.00023
Pu 95	Down	920	Yes	3.91	0.00090	0.00055	0.00144	0.00000
Pu 95a	Down	920	Yes	3.91	0.00066	0.02313	0.02380	0.02208
Pu 95b	Down	920	Yes	3.91	0.00167	0.01217	0.01384	0.01212
Pu 95c	Down	920	Yes	3.91	0.00139	0.00028	0.00167	0.00000
Pu 96	Up	920	No	9.5	0.00188	0.00034	0.00221	0.00049
Pu 96a	Up	920	No	3.5	0.00018	0.00039	0.00057	0.00000
Pu 97	Down	920	No	14.1	0.00047	0.00159	0.00206	0.00034
Pu 97a	Down	920	No	3.91	0.00206	0.00161	0.00367	0.00195

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

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

(c) The net total is the amount above the average containment box background of 0.00173

(d) Containment box background

TABLE A-5
MATRIX EXPERIMENTS--TOTAL PuO₂ LEAK (NEAREST ng)

Position:	Up			Sideways			Down		
Pressure:	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig
<u>With Vibration</u>									
5- μ m Orifice	0 0	2 5	2 0	14 9	2 2	12 1	8 ^(a) 8 ^(a) 16 ^(a) 99 ^(a) 5 ^(a)	4 7	2 ^(a) 3 ^(a)
10- μ m Orifice	6 4	6 9	423 8 2	2 0	6 8	29 134		11 36 5 ^(a) 0 ^(a) 0 ^(a) 5 ^(a)	
20- μ m Orifice	6 14 ^(a) 5 ^(a) 0 ^(a)	435 293 9 488	142 17,588 5,996 ^(a) 17 ^(a) 0 ^(a) 0 ^(a) 0 ^(a)	25 6 0	208 101 11	1 89	14 0 ^(a) 1 ^(a) 0 ^(a)	3 1,615	2 38 ^(a) 0 ^(a) 22 ^(a) 12 ^(a) 0 ^(a)
<u>Without Vibration</u>									
5- μ m Orifice	3 ^(a) 5 ^(a)		0 ^(a) 0 ^(a)				2 5		9 3 ^(a) 12 ^(a) 1 ^(a) 1 ^(a) 1 ^(a)
10- μ m Orifice		10 ^(a) 1 ^(a) 1 ^(a) 1 ^(a)					14 52	5 ^(a) 4 ^(a) 0 ^(a) 0 ^(a)	227 25
20- μ m Orifice	0 8 ^(a) 1 ^(a) 0 ^(a)	319 84	113 173 ^(a) 0 ^(a) 0 ^(a)	4 0	8 3	0 58	25 25 6 ^(a) 3 ^(a) 0 ^(a)	79 445	54 29 ^(a) 0 ^(a) 2 ^(a)

(a) Replicate experiments conducted to complete matrix for statistical analysis

TABLE A-6

ARITHMETIC MEAN VALUE OF PuO_2 POWDER LEAKED AND THE STANDARD DEVIATION FOR EACH EXPERIMENTAL CONDITION (MEAN STANDARD DEVIATION)^(a) (REPLICATE 1)

Position:	Up			Sideways			Down		
Pressure:	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig
<u>With Vibration</u>									
5- μm Orifice	0/0	3/2	1/1	12/4	2/0	7/8	-	5/2	-
10- μm Orifice	5/1	7/2	144/241	1/1	7/1	82/74	-	24/18	-
20- μm Orifice	10/6	306/215	7909/8879	10/13	107/99	45/62	7/10	809/1140	20/25
<u>Without Vibration</u>									
5- μm Orifice	-	-	-	-	-	-	3/2	-	6/4
10- μm Orifice	-	-	-	-	-	-	33/27	-	151/178
20- μm Orifice	4/6	202/166	143/42	2/3	5/4	29/41	19/11	262/259	42/18
50- μm Orifice	-	-	-	-	-	-	22/29	-	3/5

(a) Mean in nearest ng; standard deviation in ng

TABLE A-7

ARITHMETIC MEAN VALUE OF PuO_2 POWDER LEAKED AND THE STANDARD DEVIATION FOR EACH EXPERIMENTAL CONDITION (MEAN/STANDARD DEVIATION)^(a) MATRIX

COMPLETION EXPERIMENTS (REPLICATE 2)

Position:	Up			Down		
Pressure:	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig
<u>With Vibration</u>						
5- μm Orifice	-	-	-	32/45	-	2/1
10- μm Orifice	-	-	-	-	3/3	-
20- μm Orifice	3/3	-	4/9	0/1	-	9/11
<u>Without Vibration</u>						
5- μm Orifice	4/1	-	0/0	-	-	4/6
10- μm Orifice	-	3/5	-	-	2/3	-
20- μm Orifice	1/1	-	0/0	1/2	-	1/1

(a) Mean in nearest ng; standard deviation in ng

TABLE A-8
MATRIX EXPERIMENTS--CORRELATION

Position:	Up			Sideways			Down		
Pressure:	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig
<u>With Vibration</u>									
5- μ m Orifice	M ^(b) M	2E09 5E08	2E09 M	7E03 1E04	1E09 2E09	5E08 6E09	1E04 ^(a) 6E03 ^(a) 9E02 ^(a) 2E04 ^(a)	7E08 4E08	3E09 ^(a) 2E09 ^(a)
10- μ m Orifice	6E04 10E04	2E09 1E09	5E07 3E09 1E10	2E05 2E06	2E09 1E09	7E08 2E08		9E08 3E08 ^(a) 1E09 ^(a) 3E10 ^(a) 2E10 ^(a) 1E09 ^(a)	
20- μ m Orifice	2E05 1E05 ^(a) 3E05 ^(a) 3E06 ^(a)	7E07 1E08 3E09 6E07	4E08 3E06 10E06 ^(a) 4E09 ^(a) M ^(a) M ^(a) 3E11 ^(a)	6E04 2E05 M	2E08 3E08 3E08	5E10 7E08	1E05 M ^(a) 2E06 ^(a) M ^(a)	1E10 2E07	3E10 2E09 ^(a) 3E09 ^(a) 6E09 ^(a) M ^(a) M ^(a)
<u>Without Vibration</u>									
5- μ m Orifice	3E04 ^(a) 2E04 ^(a)		M ^(a) M ^(a)				6E04 2E04		7E08 2E09 ^(a) 4E08 ^(a) 7E09 ^(a) 4E09 ^(a) M ^(a)
10- μ m Orifice		6E08 ^(a) 8E09 ^(a) 6E09 ^(a) M ^(a)					2E04 7E03	1E09 ^(a) 2E09 ^(a) M ^(a) M ^(a)	8E07 9E08
20- μ m Orifice	M 2E05 ^(a) 1E06 ^(a) M ^(a)	10E07 4E08	5E08 3E08 ^(a) 1E11 ^(a) M ^(a)	3E05 M	4E09 10E09	M 1E09	6E04 6E04 2E05 ^(a) 6E05 ^(a) M ^(a)	4E08 7E07	1E09 2E09 ^(a) 2E11 ^(a) 4E10 ^(a)

(a) Replicate experiments conducted to complete matrix for statistical analysis

(b) Missing correlation data due to minute amount of PuO₂ detected

TABLE A-9

ARITHMETIC MEAN VALUE OF He: PuO₂ ATOM: MOLECULE RATIO AND THE STANDARD FOR
EACH EXPERIMENTAL CONDITION (MEAN/STANDARD DEVIATION) (REPLICATE 1)

Position:	Up			Sideways			Down		
Pressure:	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig
<u>With Vibration</u>									
5- μ m Orifice	M ^(a)	1.2E9/9.0E8	2.5E9	8.9E3	1.6E9/3.5E8	3.2E9/3.8E9	-	5.4E8/2.1E8	-
10- μ m Orifice	7.8E4/2.8E4	1.4E9/2.8E8	4.6E9/5.7E9	8.5E5/9.2E5	1.4E9/2.8E8	4.4E8/4.0E8	-	5.6E8/4.3E8	-
20- μ m Orifice	1.7E5/8.5E4	8.6E8/1.6E9	1.4E8/2.3E8	1.4E5/1.2E5	2.4E8/7.9E7	2.4E10/3.3E10	1.0E5	6.0E9/8.5E9	1.6E10/2.1E10
<u>Without Vibration</u>									
5- μ m Orifice	-	-	-	-	-	-	3.7E4/2.8E4	-	1.1E9/6.7E8
10- μ m Orifice	-	-	-	-	-	-	1.6E4/1.3E4	-	4.6E8/5.5E8
20- μ m Orifice	1.9E5	2.3E8/1.9E8	4.3E8/1.3E8	3.3E5	6.7E9/4.2E9	1.0E9	1.2E5/1.0E5	2.3E8/2.3E8	1.6E9/6.4E8

(a) Missing correlation data due to minute amount of PuO₂ detected

TABLE A-10

ARITHMETIC MEAN VALUE OF He: PuO_2 ATOM: MOLECULE CORRELATION AND THE
STANDARD DEVIATION FOR EACH EXPERIMENTAL CONDITION (MEAN/STANDARD DEVIATION)

MATRIX COMPLETION EXPERIMENTS (REPLICATE 2)

Position:	Up			Down		
Pressure:	Ambient	440 psig	920 psig	Ambient	440 psig	920 psig
<u>With Vibration</u>						
5- μm Orifice	-	-	-	8.8E3/7.1E3	-	2.5E9/1.1E9
10- μm Orifice	-	-	-	-	1.4E10/1.5E10	-
20- μm Orifice	1.6E6/1.9E6	-	1.5E11/2.1E11	2.0E6	-	4.4E9/1.8E9
<u>Without Vibration</u>						
5- μm Orifice	2.5E4/7.6E3	-	m(a)	-	-	3.8E9/3.1E9
10- μm Orifice	-	5.2E9/4.1E9	-	-	1.5E9/3.8E8	-
20- μm Orifice	1.2E6	-	1.4E11	5.6E5	-	1.2E11/1.2E11

(a) Missing correlation data due to minute amount of PuO_2 detected

TABLE A-11
SUMMARY OF PuO₂ LEAK RATE EXPERIMENTS USING AN 8- μ m ORIFICE

Run Number	Tube Position	Helium Pressure, psig	Vibration	Helium Leak Rate, (b) cc/sec	Quantity of PuO ₂ Detected, μ g (a)			
					Inlet Nozzle	Final Filter	Total	Net Total (c)
B 62 ^(d)	Sideways	---	---	---	0.00092	0.00016	0.00108	---
B 63 ^(d)	Sideways	---	---	---	0.00032	0.00067	0.00099	---
Pu 98	Up	995	Yes	5.2	0.00240	0.10889	0.11129	0.11026
Pu 98a	Up	995	Yes	3.0	0.00777	0.00040	0.00817	0.00714
Pu 98b	Up	995	Yes	23.7	0.00152	0.00047	0.00199	0.00096
Pu 98c	Up	995	Yes	1.1	0.00152	0.00037	0.00189	0.00086
Pu 99	Down	995	Yes	0.87	0.00254	0.06587	0.06841	0.06738
Pu 99a	Down	995	Yes	1.1	0.00057	0.00086	0.00144	0.00041
Pu 99b	Down	995	Yes	1.1	0.00050	0.00068	0.00117	0.00014
Pu 99c	Down	995	Yes	2.2	0.00306	0.00260	0.00566	0.00463
Pu 100	Sideways	995	Yes	1.1	0.00110	0.00050	0.00160	0.00057
Pu 100a	Sideways	995	Yes	1.1	0.00113	0.00094	0.00208	0.00105
Pu 100b	Sideways	995	Yes	2.2	0.00207	0.00031	0.00238	0.00135
Pu 100c	Sideways	995	Yes	2.2	0.00078	0.00095	0.00173	0.00070
*Pu 101	Down	995	No	1.3	0.00089	0.06448	0.06538	0.06435
*Pu 101a	Down	995	No	4.8	0.00077	0.00739	0.00816	0.00713
+Pu 102	Down	995	No	1.1	0.00082	0.00162	0.00244	0.00141
+Pu 102a	Down	995	No	7.8	0.00161	0.00141	0.00302	0.00199
Pu 103	Up	1230	Yes	24.8	0.00270	0.00059	0.00330	0.00227
Pu 103a	Up	1230	Yes	1.3	0.00034	0.00060	0.00094	0.00000
Pu 104	Down	1230	Yes	2.2	0.00001	0.00081	0.00082	0.00000
Pu 104a	Down	1230	Yes	6.9	0.00138	0.00037	0.00175	0.00072
*Pu 105	Down	1230	No	1.3	0.00016	0.00064	0.00080	0.00000
*Pu 105a	Down	1230	No	1.7	0.00091	0.00257	0.00348	0.00245
+Pu 106	Down	1230	No	2.2	0.00033	0.00138	0.00171	0.00068
+Pu 106a	Down	1230	No	10.9	0.00045	0.00085	0.00129	0.00026

(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 mid-point of the run

(c) The net total is the amount above the average containment box background of 0.00103 μ g

(d) Containment box background

* 1-hr run

+ 2-hr run

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