**Supporting Information for the
Estimation of Plutonium Oxide Leak
Rates Through Very Small Apertures**

January 1977

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SUPPORTING INFORMATION FOR THE ESTIMATION OF PLUTONIUM
OXIDE LEAK RATES THROUGH VERY SMALL APERTURES

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SUPPORTING INFORMATION FOR THE ESTIMATION OF PLUTONIUM OXIDE LEAK RATES THROUGH VERY SMALL APERTURES

INTRODUCTION

High integrity containers now under development and testing will permit assurance that when plutonium oxide is shipped by air, even the severe stresses imposed in an airplane accident will result in acceptably small consequence to the public. An analysis has been made of expected stresses and criteria developed for testing containers.^[1]

The acceptably small release of plutonium oxide will be determined by that amount which will preclude a significant radiation to individuals or the public. Various guides and standards are available to assure public safety.^[2,3,4,5]

To permit an assessment of the safety of the public from a container which leaks requires the answer to two important technical questions:

- If the container leaks a test gas at a given rate and volume under selected conditions of pressure and temperature, what will the gas leak rate be at other conditions?
- Having thus determined the gas rate and integrated gas leak, how does one determine the probable particle leak rate through apertures of the geometry resulting from distortion of mating parts normally sealing the container?

These questions have been under technical consideration, and it is the purpose of this report to provide supporting information, qualitative and quantitative, to help answer the above two questions.

At the outset it must be accepted by all interested in this topic that there must be assumptions made which place constraints on the general applicability of a technical assessment. The definition of the leak geometry is one of these. The distortion of mating surfaces during the ultimate stress can give rise to various leak geometries. A single "hole" as small as a pin prick or a series of even smaller holes with combined equivalent area may be produced. Path length of the leak may be short or long, depending on the failure mode. Other uncertainties include changes in leak geometry during the period following impact in which the container temperature may rise to 1500°F - 1700°F over a period up to an hour or so with pressures being developed to 60 - 70 atmospheres.

The detail properties of the solid material shipped are also of significance. The specifications for acceptable plutonium oxide powders allows for a wide range of particle sizes. The settling rate, packing, and bulk density are functions of particle size distribution and particle morphology.

These and the minimal experimental data to confirm theoretical calculations place limitations at the present on the calculational methods for leak assessment. Experimental studies have been proposed and a few experiments performed to get initial directly usable data on this problem. Because of the constraints of dealing with a many-faceted problem, conservative assumptions have been made to assure that leak rates will be overestimated. The following discussion presents the current course which has been taken to calculate leak rates.

LIMITING ASSUMPTIONS

1. The leak during the instant of impact and subsequent impacts prior to the container coming to rest is not included in the analysis.

Rationale -- Although the elastic distortion may momentarily (milliseconds) result in an opening of greater magnitude than the leak measured in the leak test, there is no internal pressure to drive the oxide through the leak. Spinning of the container subsequent to impact could throw some of the powder through large leaks of the proper orientation. These possibilities have not been addressed since leaks permitting this to happen are believed to exceed acceptable limits. The size of a leak permitted from pressure considerations would probably be more restrictive than leaks permitting powder to flow by gravity or spinning forces. Should it be mandatory to address such situations, the only way to handle this situation is through experimental study. The range of rotational speeds would have to be determined from impact tests.

2. During the temperature rise and holding period, the geometry (size and shape) of the leak does not change due to pressure or temperature.

Rationale -- This is a necessary assumption unless the thermal distortion and resulting leak can be calculated from the container properties and the "initial" assumed geometry, gasket behavior, etc. This information is not currently available.

3. The size distribution of particles is that corresponding to 1000°C calcined plutonium oxalate with a 15-minute dispersion prior to measurement.

Rationale -- Of the many size distributions available, the distribution measured with a limited degree of dispersion by liquid and wetting agents should be used. Liquid dispersion procedures lead to unrepresentative size distributions for this problem at hand. The particle size data chosen is believed to represent the dry powders to be shipped. The size distribution is as follows.

20	μm and smaller	100%	by weight
18	μm and smaller	99%	by weight
10	μm and smaller	81%	by weight
8	μm and smaller	56%	by weight

6	μm and smaller	39%	by weight
4	μm and smaller	20%	by weight
2	μm and smaller	4%	by weight
1	μm and smaller	1%	by weight
0.5	μm and smaller	0.15%	by weight
0.1	μm and smaller	0.1%	by weight

Figure 1 is a plot of size vs. mass.

GAS FLOW THROUGH SMALL APERTURES

For the purpose of leak testing, an equation derived from Poiseuille flow (laminar, capillary) and molecular flow has been recommended for use in the draft ANSI standard N14.5 as Equation B.3 in the Appendix. The limits of pressure and flow geometries over which the equation is applicable have not been defined. The equation is:

$$L = 3810 \frac{D^3}{a} \left[323 \frac{D}{\mu} (P_u^2 - P_d^2) + \sqrt{\frac{T}{M}} (P_u - P_d) \right], \quad (1)$$

in which

D = equivalent leak diameter, cm
 a = leak path length in cm
 μ = viscosity, centipoise (use 0.018 for air)
 P_u = upstream pressure = 1 atm.
 P_d = downstream pressure ≈ 0
 T = temperature, °K
 M = M. wt. air - 29

Initial calculations using the development in A.H. Shapiro's, The Dynamics and Thermodynamics of Compressible Fluid Flow, Vol. I,^[6] show that for leaks as large as 100 μm and 1 cm long equation (1) will overestimate the leak rate considerably for the condition of upstream pressure of 1 atmosphere and downstream pressure of near 0, the conditions of the test. The calculation requires time consuming iterations and operations more efficiently performed with a computer and a program for this calculation, which have not been feasible at this point in time. The reason for the lower predicted flow for those conditions than predicted by Eq. 1 is that compressibility in the leak path is not accounted for in the equation. Without a comprehensive analysis of all the path lengths, diameters, flow regimes and temperatures, it is recommended for the time being to accept the conservatism of the above equation in estimating flow rate for the region of

FIGURE 1
NOMINAL PuO_2 PARTICLE SIZE DISTRIBUTION

pressures and hole diameters giving sub-sonic velocities. When leaks are such that sonic velocities appear to be exceeded, the leak rate should be based on sonic velocity. In Figure 2 is plotted a series of leak rates as a function of diameter and length of hole for the leak test conditions for estimating an equivalent leak diameter. Also shown are points showing the prediction of excessive flows with Equation (1) for higher pressures. These flows exceed the sonic velocity.

The leak path length, a , to be used must be determined subjectively by examining the container design to determine the likely mode of failure. The assumption of a single leak vs. a multiple leak would lead to the higher potential for powder leakage, thus a conservative assumption.

AVAILABILITY OF PARTICLES IN THE AIR SPACE

The bulk powder will behave initially more nearly as a continuous fluid than as individual suspended particles. There is no theoretical treatment which will permit anything but crude estimates which can be made very conservative. The most realistic information available comes from four experiments carried out with a fine UO_2 powder which is a reasonable simulant for PuO_2 . After a container with 4.5 kg of the powder was rotated several times, the air was rapidly sampled, particles airborne were collected on filters which were analyzed for uranium. Results are shown in Figure 3.

The concentration in the gas space (6 cm from powder level to wall) can be represented by:

$$C = C_0 \exp(-0.0987t) + 5.5 \times 10^{-9} \exp(-0.00056t). \quad (2)$$

The value of C_0 is conservatively estimated to be $1 \times 10^{-4} \text{ g/cm}^3$, since the highest concentration measured was about $1 \times 10^{-5} \text{ g/cm}^3$.

Other efforts to approximate the settling rate and disappearance are uncertain because it is impossible to realistically describe the degree to which the powder will act as single clumped

FIGURE 2

AIR FLOW RATE Vs. EQUIVALENT HOLE SIZE
USING VARIOUS FLOW RELATIONSHIPS

FIGURE 3

URANIUM OXIDE AIRBORNE OVER THE BULK POWDER FOLLOWING DISRUPTION

aggregate from the instant the container touches the ground until the powder is essentially settled from the air space.

The measurements on which the foregoing relationship was derived are preliminary, yet enough data were taken to show the relatively low airborne concentration soon after the container comes to rest. The results are believed to be more descriptive of the actual particle behavior than were the earlier derived air concentrations based on simplistic coagulation-settling calculations.^[7] The experimental measurement and resulting equation, nevertheless, are reasonably consistent with an estimate of the airborne concentration made on the basis of coagulation theory. Simple coagulation theory leads to an estimate that for a high initial concentration, 10^{10} or 10^{11} particles per cc, in 50 seconds the concentration will drop to about 4×10^7 particles per cc. (There would initially be about 2×10^{11} particles per cc if 2 kg of plutonium oxide were assumed to be instantaneously airborne in a 3000 cc container.) Fifty seconds is chosen for this comparison since it corresponds approximately to the time (46 seconds) that particles 2 μ m and larger would have settled 6 cm. From equation 2 the calculated mass concentration is 1×10^{-6} g/cc at 46 seconds. Converting this to particles/cc, assuming the 99% of the particles now are 2 μ m and smaller and particle masses follow a log normal distribution with a geometric standard deviation of 2, the 1×10^{-6} g is equivalent to $1 - 3 \times 10^7$ particles per cc. For the assumptions necessary and the uncertainties in the empirical relation, this agreement is quite acceptable and lends credence to this analysis. A rigorous calculation is not possible with the information available.

The size distribution of the airborne powder will shift slowly after 50 seconds to smaller sizes and the concentration will slowly decline. Airborne concentrations become so low after 100 seconds in the static situation that unless leaks are large, only very small quantities of powder will be available to pass through the leak.

The longer the period between touchdown and pressure build-up from the ensuing fire, the less significant will be the powder

leak rate. Even particles as small as 2 μm (sp. gr. 10) particles will settle 6 cm in less than one minute (approximately 50 sec).

ESTIMATION OF PARTICLE REDUCTION IN PASSAGE THROUGH THE LEAK

From The Air Space

The data needed to estimate the retention of particles in small channels are the following:

- a. The particle size.
- b. The dimensions of the passage -- the diameter cross section, and length.
- c. The flow rate of the gas.
- d. The orientation of the leak with respect to the vertical.
- e. The entrance effects (capture probability).

Because all these data are not available and most can only be arrived at by speculation and educated guesses, the ability to calculate retention is severely limited. Some experimental data are available but none which are directed expressly toward the problem at hand. Some observations are made with respect to each of the important variables.

a. Particle Size

The particle size distribution of the bulk plutonium oxide will not be the size distribution available for leakage. In the first minute following the instant the container comes to rest, the larger particles will have settled out, leaving a distribution of particles almost all of which are less than 2 μm . This distribution will continue to change slowly to smaller sizes until the air space is virtually free of powder. The size distributions are approximated by assuming a log normal distribution with a geometric standard deviation of 2. In Table I are shown estimated size distributions at various times after the container comes to rest:

TABLE I

FRACTION BY WEIGHT IN INDICATED SIZE AND SMALLER
RANGE

<u>Indicated Diameter and smaller μm</u>	<u>t = 50 sec</u>	<u>t = 100 sec</u>	<u>t = 500 sec</u>	<u>t = 1000 sec</u>
2	0.99	100	100	100
1	0.90	0.97	100	100
0.5	0.71	0.80	0.99	100
0.3	0.31	0.54	0.94	0.98
0.2	0.14	0.30	0.83	0.91
0.1	0.018	0.06	0.50	0.65
0.08	0.006	0.03	0.40	0.50
0.03	0.0001	0.0014	0.03	0.08
0.02	--	--	0.007	0.02
0.01	--	--	0.0003	0.002

b. Dimensions of the Leak Passage

As discussed earlier in this paper, the leak cross section and length can be approximated by assuming a circular orifice and length determined by reviewing the container design and likely failure mode. The calculation of fractional removal in the leak channel is compromised when the actual leak dimensions do not conform to the assumptions used in deriving the equations. For example, gravitational settling in a horizontal tube can be calculated for either a circular or rectangular channel. The equation for a circular cross section is used since leak rates are inferred from the test results, assuming circular cross section of the passage. The more likely plugging of leaks of low height-to-breadth ratios has also been pointed out earlier.

Another consideration is the roughness of the walls of the leak passage. If smooth passage walls are assumed, laminar flow will occur for leaks of the sizes anticipated. This is a reasonable assumption from which follows a conclusion that turbulent deposition would be minor.

Changes in direction of the passageway will increase the fraction removed in the passage. Equations are available for determining removal in bends which could be applied, but the leak path could only be surmised from the container

design and likely failure mode.

c. Flow Rate

The velocity in the leak channel is necessary for estimating particle retention. The velocity will be calculated by dividing the leak rate (cc/sec) by the assumed area of the leak based on circular cross section. Errors arising from this assumption are expected to be of lesser importance than errors from other uncertainties.

d. Orientation with Respect to the Vertical

Gravitational settling will occur to a degree during passage of particles through a horizontal channel. A calculation should be made for a horizontal channel, particularly during the early stages. Should the build-up of pressure be so gradual as to permit particles in the air space to settle before significant leak occurs, there will be little need for knowing the geometry of the passage and air velocity accurately.

e. Entrance Effects

It is assumed that all particles in the air space will move with the air flowlines to the leak entrance. This is a reasonable assumption for very small particles (less than 1 μ m), but the leak flow lines will be crossed by some particles and these will not enter the channel. For particle sizes anticipated after a few minutes of settling, entrance effects will be minimal.

Application of equations relating particle transmission in narrow passageways to the independent variables can give only approximations because of the many necessary assumptions. Experimental data are limited, but will be used in conjunction with analytical results.

Relationships have been developed for estimating the reduction in particle concentration when particles are carried by air flow through a passage. The unique character of leaks under considera-

tion (probably smaller than 100 μm) with ill-defined length, tortuosity, and surface roughness introduces uncertainties in the application of these relationships. Following are the principal mechanisms for particle removal and the equations for estimating attenuation in the leak path. An important assumption in the use of these equations is that particles once deposited are not re-suspended from the surface. For many situations this is a valid assumption. When particles build up into more than a single layer on the surface, or when high velocities are imposed following depositions, wash-off of particles may occur. Little information for the channel geometries and flow ratios anticipated is available.

Gravitational Settling

The equation for gravity settling in a circular tube with laminar flow is as follows:^[8]

$$F = \frac{2}{\pi} (\alpha\beta + \sin^{-1} \beta - 2 \alpha^3 \beta) \quad (3)$$

in which F = the fraction of particles not deposited

$$\alpha = \left(\frac{3}{8} \frac{LZ}{V_a R} \right)^{1/3}$$

$$\beta = \sqrt{1 - \alpha^2}$$

L = length of path

Z = settling velocity

V_a = average flow velocity

R = radius of tube.

For a range of assumptions the deposition due to gravity settling will be quite small. Nearly 100 percent of 2 μm particles will traverse a 2 cm long passage 20 μm in diameter, at a flow of 0.1 cc per minute; about 95 percent will pass through a slower leak, 0.01 cc per second.

With very small channels only a relatively small deposit in the channel will impede the passage of particles until complete plugging occurs. If the leak geometry is known, the quantity of powder to fill the leak path can be calculated and an assumption

made that at that point no further penetration would occur. The weight of plutonium oxide to fill 1 cm long channels of various diameters is as follows: 10 μm , 7.8×10^{-6} g; 40 μm , 1.25×10^{-4} g; 100 μm , 7.8×10^{-4} g. For these cases if only 1% were deposited in the leak path, the leak should plug when 100 times the above quantities had passed through. This would be quite conservative since as the leak began to plug it would permit less and less powder to pass through.

Turbulent Impaction

Particles in turbulent flow are lost to the walls of the channel by eddy transport and final penetration by inertial processes to the wall. Such deposition is described by the equation: [9]

$$\frac{C}{C_0} = e^{-\frac{4KL}{V_a D}} \quad (4)$$

in which C_0 = the entering concentration

C = the concentration at a distance L from the entrance

L = distance from the entrance to exit, cm

V_a = average velocity in tube, cm/sec

D = diameter of the tube, cm

K = deposition velocity, cm/sec.

For other than circular tubes, the equation is: [10]

$$\frac{C}{C_0} = e^{-\frac{P K L}{A V_a}} \quad (5)$$

in which P = perimeter of the leakage path

A = cross sectional area of leakage path.

The deposition velocity, K , must be known or estimated to evaluate the deposition in the leak path. Many experiments have been carried out to determine the correlation of K with important parameters, which are particle size, Reynolds Number, and surface roughness. [11,12] None of the experimental data include the range of hole diameters as small as considered in this study. Laminar flow will occur in most cases considered in this study and the

Reynolds Number should be above 2100 for turbulent deposition to be significant. In these cases the settling velocity would be used for K, since this will give a lower limit to the deposition. Experimentally determined values are not available for very small leaks.

One other impaction mechanism coming into play, but even less amenable to quantitative analysis, is the inertial removal by the deflection of the gas in the leak path. Abrupt changes in direction will cause plate-out of the particles.

Brownian Diffusion

Equations have been derived for calculating the particle loss in small channels by Brownian^[13] diffusion. Browning and Ackley used the following equations to predict the decontamination factor of very small circular and rectangular passageways.^[14]

For circular:

$$\log_{10} \text{ D.F. } = 0.0867 + 203.2 \frac{\mu D L^2}{\Delta P d^4} . \quad (6)$$

For rectangular:

$$\log_{10} \text{ D.F. } = 0.0386 + 39.35 \frac{\mu D L^2}{\Delta P b^4} . \quad (7)$$

D.F. = decontamination factor or the ratio of the number of particles entering the channel to the number penetrating.

μ = viscosity of air, poise

D = diffusion coefficient of particle in air, cm^2/sec .

L = length of leak, cm

ΔP = pressure drop through leak, dynes/cm^2

d = diameter of cylindrical leak

b = smallest dimension of rectangular leak.

In Figures 4 and 5 are shown the results of calculations for various particles and leak sizes for a leak passage length of 1 inch (2.5 cm).

These results show that for 5 μm diameter circular leaks, particles as large as 2 μm will be attenuated by a factor of

Fig. 4 Decontamination Factors Resulting from Diffusional Deposition of Particles in Cylindrical Leaks in Reactor Containment Vessels. [14]

Fig. 5 Decontamination Factors Resulting from Diffusional Deposition of Particles in Rectangular Leaks in Reactor Containment Vessels. [14]

1/10, and the attenuation of 0.5 μm particles could be as large as 10^{-5} . For a 20 μm leak, particles 0.1 μm would be deposited to the extent of 50 percent in passing through, but particles 0.05 μm diameter would be efficiently deposited in the 20 μm channel, with a D.F. of about 30 ($\sim 3\%$ would pass through).

Electrostatics

Unipolar electrostatic charges would enhance diffusional deposition due to repulsion of one particle by another, thus effectively increasing particle mobility. Fuchs^[15] calls attention to this as a possible mechanism and gives an equation for estimating the reduction in concentration. The reduction factor is given by the following expression:

$$\frac{C_0}{C} = 1 + \frac{4\pi q^2 B C_0 L}{V_a} \quad (8)$$

in which

- B = D/KT particle mobility
- D = diffusion coefficient
- k = Boltzman constant
- q = particle charge
- V_a = average velocity
- L = length of passage.

The significance of this effect for the leak dimensions under consideration has not been determined. Garrity^[10] presents sample cases for a reactor containment leak through capillaries ranging from 56 to 100 μm and particles ranging from 0.17 μm to 1 μm . The deposition length ranged from 10 feet to 32 feet. Reduction factors ranging from 27 to 263 were calculated. Diffusion alone gave corresponding reduction factors ranging from 28 to 5600. Assumptions regarding net charge per particle are not presented.

Thermophoretic Deposition

Deposition from a hot surface region to a cold surface will effectively remove small particles from a stream. The leak channels are so small it is not likely that the required thermal gradients will be present.

Leakage From The Bulk Powder

When the leak area is covered with powder, some limiting conditions can be set forth with assurance that the particles passing through the leak will be highly restricted. When these limiting conditions are not met, the estimation of the particle leak rate (when the leak entrance is covered with powder) becomes less certain. At one extreme the cross sectional area can be so small that a single large particle will block the passage. At the other extreme the powder will flow by gravity or with applied pressure through the opening. In the foregoing examination of the mechanisms for deposition the initial condition requires that particles are airborne as they enter the leak passage. When the powder covers the leak entrance, the fluidization forces will be low and the aerodynamic forces available to move particles to the leak will be restricted by the friction of particle against particle. The situation is much like a filter cake building up on the filter medium, the filter medium in the leak case being the single hole and the wall of the container. The compact layer of particles will serve as a filter bed entrapping the particles approaching the leak. Attention must be directed to the immediate vicinity of the leak entrance and the properties of the particles.

Following are observations relating to the limiting and other leaks.

a. Limiting Leaks

It is obvious that a passageway whose minimum dimension is smaller than a given particle diameter cannot be traversed by a particle of larger dimension. This mechanical blocking by the first particle of this size reaching the hole will set one limit to the leakage of powders. In the particle size distributions of plutonium oxide examined in this study there were always some particles as large as 20 μ m. For the nominal size distribution assumed for this analysis about 1% of the weight is associated with particles 18 - 20 μ m. This is equivalent to about 3×10^5 particles in a gram of nominal powder, assuming particles have a density of 10. Since there will be about 300 per mg of nominal powder which can block the passage, it is highly probable that only 1/300 of a milligram would enter the leak channel. Actually, a much smaller

quantity would pass through the channel before complete plugging would occur, since only 3×10^{-5} gram would be needed to fill a 20 μm diameter by one cm long capillary. Deposition and bridging would almost surely preclude the transmission of more than a few micrograms of powder through a circular aperture of this size.

It is unlikely that the leak channel will be circular in cross section. More likely the cross section will be like a crack, long and narrow. A leak 5 μm wide and 63 μm long would have the same area of a 20 μm diameter opening. There are about 5×10^8 5 μm or large particles per gram of nominal powder. For a leak of this geometry only 13 5 μm or even fewer larger particles would be required to effectively block the passage. At the point of complete blockage only $(13)(2 \times 10^{-9}) = 2.6 \times 10^{-8}$ g would have entered the leak channel.

A leak channel 18 μm by 63 μm would be blocked by four 20 μm particles at the moment when 4/300 of a mg of powder would have entered the channel. The latter leak would correspond to approximately 0.004 cc atm/sec through a 1 cm path.

b. Leaks not Limited by Mechanical Interference

When the leak is of such geometry that mechanical interference does not preclude particles from passing through the channel from the bulk powder, the estimation of particle transmission rate must be made with conservatism because of the lack of experimental data. The most conservative approach would be that all particles move through the passage in "piston" flow so that the bulk density of about 2 g per cm^3 would determine the leak rate. A 0.1 cm^3 per second gas leak would deliver then, 0.2 g per second. (A circular leak of about 60 μm and 1 cm long would permit 0.1 cm^3/sec to flow at a pressure difference of 1 atmosphere, $P_u = 1$, $P_d = 0$). The minimum hole dimension permitting essentially unrestricted flow of particles under the pressure regimes anticipated during the fire is not known, and is not amenable to calcula-

tion. The following observations are made to emphasize the extreme conservatism in the foregoing assumption. Unfortunately, the observations do not offer the calculational models for arriving at actual particle leak rates.

- Particles at the entrance to the leak will be aggregated and clumped together. The particle-particle adhesion will tend to bridge across holes of diameter larger than the largest particle in the bed. There will be considerable frictional resistance for particles to reduce slippage between particles as they are pressed into the leak entrance. Photomicrographs of plutonium oxide particles show marked surface roughness that would enhance bridging over small openings. [16]
- The weight of particles to seal or plug a leak of fairly large size, say 100 μm in diameter by 1 cm long, is very small (less than one mg). Only a very small quantity would pass before the leak would be sealed during the initial build-up of pressure. The important question of whether subsequent pressure build-up will blow particles from such plugged leaks will depend greatly on the structure of the leak passage and the physical nature of the particles. The total load on the particles from 1000 psi internal pressure would be about 6 g over the area of a 100 μm leak entrance. Frictional resistance exceeding this small force would prevent particles from discharging at the high pressures. There will be a passageway of such dimensions that particles will be continuously blown through by the gases under pressure. Information on this limiting dimension in relationship to particle properties has not been found in the literature. In the following section data are presented to show that leaks of larger dimensions than the particle sizes present will transmit a very small fraction of particles which reach the leak entrance.
- Mechanical interference has been considered above as a leak-limiting condition. Nearly as limiting is the situation in which the leak passageway changes direction very

abruptly. This passage geometry will be realized when a relatively soft metal gasket is used as the closure seal with the leak-tightness achieved by knife edges machined into one mating surface. The leak passage through such a leak provides an abrupt 90° change in direction. Inertial effects will force particles to be trapped just downstream of the transition. The mechanical support afforded by surfaces comprising the passageway will be significant in plugging such passageways.

EXPERIMENTAL AND RELATED ANALYSES

Relatively little experimental data are available which can be applied directly to the problem at hand. Following are brief summaries of experimental observations which can be used in a subjective way to show that particles entering very small apertures are effectively retained.

Fontana and Browning^[17] in an early paper analyzed the significance of agglomeration in a reactor containment vessel in reducing the release of fission product through high efficiency filters. High efficiency filters are similar in many respects to multiple leak paths of the dimensions of concern in the current problem. Assuming that iodine as a pure nuclide was released into containment, their conclusion was: "This analysis shows that agglomeration occurs at a rate fast enough to prevent significant penetration of the filtration system even if all particulate of a size smaller than 0.3μ were assumed to penetrate the filters. In one example the calculated amount of iodine released would be less than 10^{-5} of that available."

Witherspoon and Postma^[18] reported experiments on the leakage of fission products in the Containment Systems Experiment. Leaks were fabricated as small orifices, or valves were used which were made to leak. Gas leak rates were quite high, 1 liter per minute. Table II shows a summary of their data.

TABLE II^[18]

SUMMARY OF PENETRATION OF ARTIFICIAL LEAKS
BY IODINE AND CESIUM AEROSOLS

Nature Of Leak	Radionuclide On Particle	Leak Rate ℓ/min	Overall Decontamination Factor for Air- borne Activity
Valve leak with inlet flush with wall	Iodine	Run A-5 1.0	8.1
		Run A-16 1.0	10.8
"	Cesium	Run A-5 1.0	> 112
		Run A-16 1.0	> 1500
Valve leak with inlet probe extending into chamber	Iodine	Run A-11 1.0	3.6
		0.1	4.1
	Cesium	Run A-11 1.0	> 130
		0.1	> 537
	Iodine	Run A-16 0.5	3.1
		1.6	22
	(overall 3.73)		
	Cesium	Run A-16 0.5	96
		1.6	51
	(overall 81)		
Small pinhole leak in orifice on end of tube extending into chamber	Iodine	Run A-16 1.6 start	3.8
		0.7 end	
	Cesium	Run A-16 1.6 start	4.9
		0.7 end	
(during sampling period ranged from 0.25 to 77.5)			
Leakage from 8" butterfly valve	Iodine	Run A-16 1.0	34
	Cesium	Run A-16 1.0	210

The authors note that all "natural" leaks in the containment vessel plugged with steam, hence the artificial leaks of size to remain unplugged and deliver the indicated sample flow rate were prepared. The chamber had high humidity during the runs.

Nelson, et al^[19] measured the fraction of sodium oxide aerosol which would pass through a crack in a concrete plug. The data are reproduced from the report in Table III. In Figure 6 is shown the decontamination factor as a function of crack dimensions. These crack dimensions are much larger than the leak dimensions under consideration for a leakage of 10^{-3} cc/sec.

The length of the leak could be similar to that anticipated for the PuO_2 shipping container leak. As noted in Figure 3, for a crack width of about 0.002 in ($\sim 51 \mu\text{m}$), a decontamination factor of over 10^5 was recorded.

Nelson and co-workers have performed more recent experiments to measure the leakage of sodium oxide aerosols through prepared cracks or capillary tubing.^[20,21,22] Sodium oxide aerosols of average aerodynamic diameter of 3 to 5 μm at concentrations ranging from a fraction of a microgram per cc to 18 $\mu\text{g/cc}$ were sampled through capillaries ranging from 0.052 cm (520 μm) to 0.157 cm (1570 μm). Capillary lengths were from 4.9 to 7.6 cm. When the aerosol was dry, 1300 ppm water, (R.H. of about 5%), the capillaries did not plug for long test periods. When the humidity was adjusted to 8000 ppm, plugging of the capillaries occurred in times ranging from 0.1 min to 41 min. These tests show the importance of the aerosol properties, but are not directly applicable to the very small leaks under consideration. They do support a generalization that small leaks can be readily plugged with aerosol particles of much smaller dimension than the diameter of the leaks. The authors don't give explicit decontamination factors for the air passing through the leak up to the time of plugging, but they do indicate that the plug acts as a filter.

Membrane filters with average pore size from milli-microns to several microns are used in many applications where high effici-

TABLE III
EXPERIMENTAL TEST DATA [13]

Parameters	Porous Alundum Crucible	Concrete Specimen					
		1	2	3	4	3'	5
Diameter (in.)	1.5	2	2	2	2	2	2
Thickness (in.)	1/16	1.5	1.5	1.5	1.5	1.5	1.5
Initial Flow at 5 psig (l/min)	25	1.2	11.2	24	24	24	1.3×10^{-3}
Flow at 5 psig After Test (l/min)	-	1.2	10	1.4	12.5	1.0	$<10^{-4*}$
Type of Leak†	Porous	Crack	Crack and Orifice	Crack	Crack and Orifice	Crack	Porous
ΔP Across Specimen During Test (psig)	<0.5	5	0.5 to 1	2.5	2.5 to 4	2.5 to 3	3
Crack Width (mil)	None	~ 1.8	~ 3.8 to 4.5	~ 5	~ 5 to 6.3	~ 5	None
Reduction§ Factor for Sodium Oxide	$4 \times 10^4*$	$>10^5*$	5.5×10^3	2.5×10^3	150	10^3	$>5 \times 10^3*$
Reduction§ Factor - Agglomerated I_2^{131}	$>10^3*$	-	-	-	-	10^3	-

* Limit of detection

† Type of flow characterized by Equation 1 (see below).

§ Ratio of sodium or agglomerated I_2^{131} entering concrete as compared to that on membrane filter.

Equation (1): $V = \frac{gc \cdot b^3 \cdot L(Pu-Pb)}{128 \mu x}$, in which V = leak rate, b = crack width, x = crack depth, L = length of crack, gc = dimensional constant, μ = viscosity.

Figure 6. Aerosol Reduction Factor vs Crack Width^[13]

ency removal of particles is required. These filters are very thin, of the order of 50-100 μm , yet they are effective filters for small particles. Lindeken^[23] has shown the effectiveness of 5 μm pore size filters for collecting submicron natural radioactive aerosols. Face velocities may be chosen to favor diffusional deposition of very small particles. The relatively long leak pathway would be even more favorable to diffusional deposition. Increased filter efficiency with loading is generally observed. One example is shown in reference [24]. Whatman 41 filter paper which is a relatively porous filter showed an increase in efficiency for 0.365 μm particles from 75% to 97% in a 30 minute period of use. The concentration of the aerosol sampled was 0.5 mg/m^3 .

CONCLUSIONS

Information is presented from which an estimate can be made of the release of plutonium oxide from shipping containers. The leak diameter is estimated from gas leak tests of the container and an estimate is made of gas leak rate as a function of pressure over the time of interest in the accident. These calculations are limited in accuracy because of assumptions regarding leak geometry and the basic formulations of hydrodynamic flow for the assumed conditions. Sonic flow is assumed to be the limiting gas flow rate. Particles leaking from the air space above the powder will be limited by the low availability of particles due to rapid settling, the very limited driving force (pressure buildup) during the first minute, and the deposition in the leak channel. Equations are given to estimate deposition losses.

Leaks of particles occurring below the level of the bulk powder will be limited by mechanical interference when leaks are of dimension smaller than particle sizes present. Some limiting cases can be calculated. When the leak dimension is large compared to the particle sizes present, maximum particle releases can be estimated, but will be very conservative.

Further theoretical and experimental studies are needed to better define the hydrodynamics of gas flow in leaks of the size being considered, and to establish particle transport rates through known geometry leak paths.

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