CONTAINMENT EVALUATION OF PU-METAL TRANSPORT USING MULTIPLE BARRIERS

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ABSTRACT

A methodology was developed previously by SRNL to show that Al-SNF with cladding breaches can be directly transported in standard casks and maintained within the allowable release rates. This novel approach may be extended to other nuclear material systems. Utilizing an adaptation to the methodology, a containment analysis has been performed for the scenario of non-routine transfer of a damaged 9975 package containing plutonium metal from K-area monitored storage to F-area on the Savannah River Site. A multiple barrier system with each barrier having a defined leakage rate of less than $1x10^{-3}$ cm^3/sec of air at Standard Temperature and Pressure was analyzed to determine the number of barriers needed to transport the package under normal transportation conditions to meet transportation requirements for containment. The barrier system was analyzed parametrically to achieve a composite system that met the federal requirements for the maximum permissible release rate. The multiple barrier system acts to retard the release of radioactivity. That is, a build-up in the radioactivity release rate occurs with time. For example, a system with three barriers (e.g., sealed plastic barrier) with a total free volume of 4,500 cm^3 could be transported for a total time of up to approximately 10 days with a release rate within the permissible rate. Additional number of barriers, or volume of the barriers, or both, would extend to this period of time. For example, a system with seven barriers with a total free volume of 4,500 cm^3 could be transported for up to 100 days. Plastic bags are one type of barrier used in movement of radioactive materials and capable of achieving a leak rate of $1x10^{-3}$ cm^3/sec of air at STP. Low-density polyethylene bags can withstand high temperature (up to 180°C); a barrier thickness of 10 mils should be suitable for the barrier system.

INTRODUCTION

Transportation packages for nuclear materials are subject to DOT and NRC (10CFR71) requirements. Equivalency to these requirements can be used enable acceptable systems for on-site transport. One such equivalency is for containment during transport. The methodology to establish containment for transportation using a multiple-barrier system is defined in this report. A potential application to a hypothetical on-site shipment of a plutonium package is used as a case study. The 9975 transportation package is being used in the K-area monitored storage of plutonium metal. This package is considered leak-tight with respect to transportation of radioactive material (*leak rate* < 10^{-7} *cm*^{3}*/sec* air at STP). Scenarios have been proposed whereby the containment barriers of this package have been breached. In this case, the suspect package would need to be transported from KAMS to F-area for disposition. This paper provides a detailed evaluation of configurations of multiple barriers encasing the breached 9975 package to demonstrate containment using a multiple-barrier system. Figure 1 displays a drawing of the 9975 transportation package taken from the 9975 Safety Analysis Report.[1]

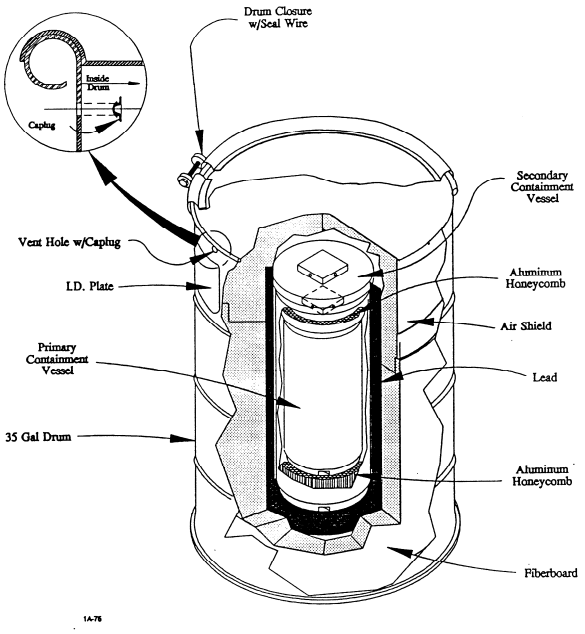

Figure 1 Sketch of the 9975 transportation package.[1]

CONTAINMENT ANALYSIS OF THE 9975 FOR TRANSPORTING PLUTONIUM METAL

The approach to demonstration of containment is to meet the 10CFR71 requirements for allowable release. The American National Standard for radioactive materials –leakage tests on packages for shipment provides the method to demonstrate allowable leakage rates of a containment system. This methodology is adapted to a multiple-barrier system. The determination of the activity source term, A_2, for the plutonium metal in a 3013 canister in a 9975 package is described.

Containment Analysis Methodology

The following describes the containment analysis methodology used for the containment evaluation of plutonium metal in the 9975 package.

Calculation of Permissible Leakage Rates

The containment criterion for Type B packages requires that a package have a radioactive release rate less than $A_2 x 10^{-6}$ in one hour under normal conditions of transport. The parameter A_2 has units of curies (*Ci*) and is isotope dependent. A_2 is calculated from the isotopic curie concentration in the fuel that is determined through the use of maximum allowable isotopic concentrations determined from information in Table 1.10 of the SAR.

The maximum permissible release rate for normal conditions of transport can be expressed as follows:

$$R_N = L_N C_N \le A_{2,N} * 2.78 \times 10^{-10} \ \ s^{-1} \qquad \text{(Eq. 1)}$$

where:

R_N is the release rate for normal conditions of transport [*Ci/s*],
L_N is the volumetric gas leakage rate [cm^3/s] under normal conditions of transport,
C_N is the curies per unit volume of the radioactive material, "activity density", that passes through the leak path for conditions of transport [Ci/cm^3], and
$A_{2,N}$ is the mixture A_2 of the radionuclides available for release under conditions of transport [*Ci*].

Activity Density Determination

There is only one source of radioactive material that may become airborne during transportation. This source is fines such that:

$$C_{total} = C_{fines} \qquad \text{(Eq. 2)}$$

where:

C_{total} is the total releasable activity density inside the containment vessel [Ci/cm^3] and
C_{fines} is the releasable activity density inside the containment vessel due to the release of fines [Ci/cm^3].

The releasable activity density inside the containment vessel due to the release of fines is described by Eq. 3.

$$C_{fines} = \frac{M_{oxide} \times f_f}{V_C} \ Ci/cm^3 \qquad \text{(Eq. 3)}$$

where:

M_{oxide} is the total number of curies of all radionuclides in the plutonium metal form [*Ci*],
f_f is the oxide spallation fraction [$f_f = 0.15$], and
V_C is the free volume of the PCV [cm^3].

Activity Values for Radionuclides

The A_2 value for the fines is derived from the values provided in Appendix A, Table A-1 of 10CFR71. The A_2 value for mixtures of isotopes is calculated from:

$$A_2 = \left(\sum \left(R_i / A_{2_i} \right) \right)^{-1} \ Ci \qquad \text{(Eq. 4)}$$

where:

R_i is the fraction activity of nuclide *i* in the mixture and
A_{2i} is the appropriate A_2 value for nuclide *i*.

Determination of the Maximum Permissible Leakage Rate

The maximum permissible leak rate is calculated by using the solutions to Eq. 2 and Eq. 4 and solving for L_i in Eq. 1 at normal conditions of transport.

Maximum Permissible Leakage Rate at Standard Conditions

The volumetric gas leak rate is modeled as a combination of continuum and molecular flow through a single leak path. The leak path is modeled as a smooth, right-circular cylinder with sharp edges. Based on these assumptions, the equation for gas leaking from the cask takes the following form.

$$L = L_c + L_m \qquad \text{(Eq. 5)}$$

where:

L is the volumetric gas flow rate at P_u [cm^3/sec],
L_c is the volumetric flow rate due to continuum flow [cm^3/sec], and
L_m is the volumetric flow rate due to molecular flow [cm^3/sec].

The volumetric flow rate, L_c, for continuum flow is given by

$$L_c = \left[\frac{2.49\times10^6 * D^4}{a\mu}\right] * \left(P_u - P_d\right) = F_c\left(P_u - P_d\right)\ {cm^3}/{s} \qquad \text{(Eq. 6)}$$

where:

F_c is the continuum flow coefficient [cm^3/s],
D is the capillary diameter [cm],
a is the capillary length [cm] (typically found in the SAR of a given cask),
μ is the fluid viscosity [cP] (typically found in CRC Handbook),
P_u is the upstream pressure [atm] (typically found in the SAR of a given cask), and
P_d is the downstream pressure [atm].

The volumetric flow rate, L_m, for molecular flow is given by

$$L_m = \left[\frac{3.81\times10^3 * D^3}{aP_a}\times\sqrt{T/M}\right] * \left(P_u - P_d\right) = F_m\left(P_u - P_d\right)\ {cm^3}/{s} \qquad \text{(Eq. 7)}$$

where:

L_m is the volumetric flow rate due to molecular flow [cm^3/sec],
F_m is the molecular flow coefficient [$cm^3/atm{\bullet}s$],
D is the capillary diameter [cm],
T is the gas temperature [K] (typically found in the SAR of a given cask),
M is the gas molecular weight [$g/mole$] (typically found in CRC Handbook),
a is the capillary length [cm] (typically found in the SAR of a given cask),
P_u is the upstream pressure [atm], and
P_d is the downstream pressure [atm].

To correlate the maximum permissible leak rate to the leak rate at standard temperature and pressure, Eq. 5 is solved for the capillary diameter (see Eq. 6 and Eq. 7) at the expected environmental conditions. The resulting diameter is then used in Eq. 5 with the temperature equal to *298-K*, the upstream and downstream pressures equal to *1.0 atm* and *0.01 atm*, respectively, and the gas molecular weight and viscosity equal to that of dry air at standard temperature and pressure. The resulting leak rate is maximum allowable testing leak rate of the shipping cask.

Evaluation of Plutonium Metal Source Term and Allowable Leakage Rate from a 9975 Package

The following details the calculations for the containment evaluation of the 9975 package containing plutonium metal. Input for the analysis is provided in the 9975 SAR, unless stated otherwise.

Activity Values

A_2 values are derived from the values provided in Appendix A, Table A-1 of 10CFR71. The A_2 value for mixtures of isotopes is calculated from Eq. 4. In the specific case of plutonium metal contents permitted in the 9975 package, a bounding composition for containment must be determined. The composition of the plutonium metal allowable in the 9975 package is provided in Table 1.10 of the current SAR as is reproduced in part in Table 1.

In determining the bounding composition for containment, two compositions were selected. The first composition was derived to provide the lowest value of A_2, i.e., lowest allowable release rate, while yielding the maximum activity. Maximizing the activity, while ignoring the A_2 value yielded the second composition. These compositions are provided in Table 2 as Case 1 and Case 2, respectively. The activity calculations assume a total mass of *4.4-kg*.

Using the compositions provided in Table 2, the A_2 values for the plutonium metal are calculated in Table 3 and Table 4, respectively.

Table 1 9975 Package – ^{239}Pu Metal

Isotopes	Max Weight percent
^{241}Am	5.00
^{243}Am	0.00010
^{244}Cm	0.00010
^{237}Np	2.00
^{236}Pu	0.000001
^{238}Pu	0.40
^{239}Pu	95.00
^{240}Pu	30.00
^{241}Pu	2.00
^{242}Pu	5.00
U	1.00

Table 2 Plutonium Metal Compositions Considered

Isotopes	Spec. Act. (Ci/g)	A_2 (Ci)	Case 1		Case 2	
			Wt %	Activity (Ci)	Wt %	Activity (Ci)
^{241}Am	3.4	0.00541	5.00	748.0	–	–
^{243}Am	0.2	0.00541	0.00010	0.00088	–	–
^{236}Pu	534.0	0.0189			0.000001	0.0235
^{238}Pu	17.0	0.00541	0.40	299.2	0.4	299.2
^{239}Pu	0.062	0.00541	64.6	176.2	67.6	184.4
^{240}Pu	0.23	0.00541	30.0	303.6	30	303.6
^{241}Pu	100.0	0.270	–	–	2	8800
Total Activity (Ci)				**1527**		**9587**

Table 3 Mixture A_2 Determination – *Case 1*

Isotope	A_2-Value (Ci) $A_{2,i}$	Activity (Ci) A_i	Fraction (Fr) $R_i=A_i/\Sigma A_i$	Fr/A_2 (1/Ci) $R_i/A_{2,i}$
^{241}Am	5.41E-03	7.48E+02	4.90E-01	9.05E+01
^{243}Am	5.41E-03	8.80E-04	5.76E-07	1.07E-04
^{238}Pu	5.41E-03	2.99E+02	1.96E-01	3.62E+01
^{239}Pu	5.41E-03	1.76E+02	1.15E-01	2.13E+01
^{240}Pu	5.41E-03	3.04E+02	1.99E-01	3.67E+01
Sum Totals	**ΣA_i=**	**1.53E+03**	**$\Sigma R_i/A_{2i}$=**	**1.85E+02**
			A_2=	**5.41E-03**

Table 4 Mixture A_2 Determination – *Case 2*

Isotope	A_2-Value (Ci) $A_{2,i}$	Activity (Ci) A_i	Fraction (Fr) $R_i=A_i/\Sigma A_i$	Fr/A_2 (1/Ci) $R_i/A_{2,i}$
^{236}Pu	1.89E-02	2.35E-02	2.45E-06	1.30E-04
^{238}Pu	5.41E-03	2.99E+02	3.12E-02	5.77E+00
^{239}Pu	5.41E-03	1.84E+02	1.92E-02	3.56E+00
^{240}Pu	5.41E-03	3.04E+02	3.17E-02	5.85E+00
^{241}Pu	2.70E-01	8.80E+03	9.18E-01	3.40E+00
Sum Totals	**ΣA_i=**	**9.59E+03**	**$\Sigma R_i/A_{2i}$=**	**1.86E+01**
			A_2=	**5.38E-02**

Activity Density Determination

The releasable activity density inside the containment vessel due to the release of fines is described by equation Eq. 3. Appendix 4.1 of the SAR provides a calculation for the amount of oxide that can form on the plutonium metal yielding the maximum activity. This methodology is followed to determine the composition and mass of oxide for this containment evaluation. It is assumed that the number of moles of O_2 that are available to react with the metal is equal to that calculated in the referenced appendix or *0.03122 moles* O_2. The following two sections provide calculations for the content of the oxides formed on the plutonium metal assuming the compositions detailed in Table 2. It is recognized that unless the barrier system for containment is installed immediately, additional oxidation of the plutonium metal may occur. Therefore, this limit to O_2 to produce MO_2 may not be conservative.

Since the mole ration of O_2 to MO_2 (metal oxide) is one, there are *0.03122 moles* of MO_2 formed by reaction with the oxygen in air and in the water vapor present when the metal is packaged. The mole ration of M to MO_2 is also one, therefore, *0.03122 moles* of M are in the oxide form due to this reaction. The mass of metal (M) formed by the metal oxidation requires the molecular weight of the metals being oxidized. The molecular weight of the metal is determined by first determining the number of moles of each metal constituent of the oxide. These values are used to determine the mole fraction of each isotope, which is then multiplied by the isotopes' molecular weight. The results are summed to provide the molecular weight of the metals. This molecular weight is then multiplied by the number of moles of metal that is reacted (i.e., the number of moles of O_2 that reacts) to yield the mass of metal in the oxide. These calculations are provided in Table 5 and Table 6, for *Case 1* and *Case 2*, respectively.

The amount of radioactive material that may be aspirated is then given by Eq. 3 as follows:

Case 1

$$C_{fines} = \frac{2.59 \times 0.15}{3389} = 1.15 \times 10^{-4} \ \ Ci/cm^3$$

Case 2

$$C_{fines} = \frac{16.3 \times 0.15}{3389} = 7.21 \times 10^{-4} \ \ Ci/cm^3$$

In the above calculations, the PCV free volume is assumed equal to that used in the calculations in Appendix 4.1 of the SAR (*3389* cm^3).

Table 5 Metal Oxidation Calculations – *Case 1*

Isotope	Molecular Weight	# Moles	Mole Fraction	Fractional Molecular Weight	Grams in Oxide	Curies in Oxide
^{241}Am	241.057	9.13E-01	4.97E-02	1.20E+01	3.74E-01	1.27E+00
^{243}Am	243.061	1.81E-05	9.85E-07	2.39E-04	7.48E-06	1.50E-06
^{236}Pu	236.046	7.39E-02	4.02E-03	9.58E-01	2.99E-02	5.08E-01
^{238}Pu	238.05	1.19E+01	6.47E-01	1.55E+02	4.83E+00	2.99E-01
^{239}Pu	239.052	5.50E+00	2.99E-01	7.18E+01	2.24E+00	5.16E-01
Totals		**1.84E+01**		**239.45**	**7.48**	**2.59**

$$\text{Total grams of M} = 0.03122\ moles\left(\frac{239.45\ g}{mole}\right) = 7.48\ g$$

Table 6 Metal Oxidation Calculations – *Case 2*

Isotope	Molecular Weight	# Moles	Mole Fraction	Fractional Molecular Weight	Grams in Oxide	Curies in Oxide
^{236}Pu	236.046	1.86E-07	1.01E-08	2.39E-06	7.47E-08	3.99E-05
^{238}Pu	238.05	7.39E-02	4.02E-03	9.58E-01	2.99E-02	5.08E-01
^{239}Pu	239.052	1.24E+01	6.77E-01	1.62E+02	5.05E+00	3.13E-01
^{240}Pu	240.054	5.50E+00	2.99E-01	7.18E+01	2.24E+00	5.16E-01
^{241}Pu	241.057	3.65E-01	1.99E-02	4.79E+00	1.49E-01	1.49E+01
Totals		1.84E+01		**239.39**	**7.47**	**16.3**

$$\text{Total grams of M} = 0.03122\ moles\left(\frac{239.67\ g}{mole}\right) = 7.47\ g$$

Maximum Permissible Release Rate and Maximum Permissible Leakage Rate

The maximum permissible release rate for normal conditions of transport are determined below using Eq. 1.

Case 1

$$R_N = L_N C_N \le A_{2,N} * 2.78\times10^{-10}\ \ s^{-1}$$

$$L_N \le \frac{5.41\times10^{-3} * 2.78\times10^{-10}}{1.15\times10^{-4}} = 1.31\times10^{-8}\ \ cm^3/s$$

Case 2

$$R_N = L_N C_N \le A_{2,N} * 2.78\times10^{-10}\ \ s^{-1}$$

$$L_N \le \frac{5.38\times10^{-2} * 2.78\times10^{-10}}{7.21\times10^{-4}} = 2.07\times10^{-8}\ \ cm^3/s$$

Permissible Leak Rate at Standard Conditions

Determination of the permissible leak rate at standard conditions utilizes Eq. 5 - Eq. 7.

$$L = L_c + L_m$$

$$L_c = \left[\frac{2.49\times10^{6} * D^4}{a\mu}\right] * \left(P_u - P_d\right) = F_c\left(P_u - P_d\right)\ cm^3/s$$

$$L_m = \left[\frac{3.81\times10^{3} * D^3}{aP_a} \times \sqrt{T/M}\right] * \left(P_u - P_d\right) = F_m\left(P_u - P_d\right)\ cm^3/s$$

$$L = \left[\frac{2.49\times10^{6} * D^4}{a\mu} + \frac{3.81\times10^{3} * D^3}{aP_a} \times \sqrt{T/M}\right] * \left(P_u - P_d\right)$$

$$L = \left[\frac{2.49\times10^{6} * D^4}{0.381\times0.0232} + \frac{3.81\times10^{3} * D^3}{0.381\times1.3} \times \sqrt{429.1/29}\right] * \left(1.6 - 1\right)$$

$$L = \left[2.82\times10^{8} * D^4 + 2.96\times10^{4} * D^3\right] * \left(0.6\right)$$

$$L = 1.69\times10^{8} * D^4 + 1.78\times10^{4} * D^3$$

Case 1

Substituting $L=1.31x10^{-8}\ cm^3/s$ and solving for D yields a capillary diameter of $7.44x10^{-5}\ cm$.

Case 2

Substituting $L=2.07x10^{-8}\ cm^3/s$ and solving for D yields a capillary diameter of $8.51x10^{-5}\ cm$.

To correlate the maximum permissible leak rates calculated in Section 0 to the leak rate at standard temperature and pressure, the calculated capillary diameter is substituted into Eq. 5-Eq. 7.

$$L_R = \left[\frac{2.49\times10^6 * D^4}{a\mu} + \frac{3.81\times10^3 * D^3}{aP_a} \times \sqrt{T/M}\right] * \left(P_u - P_d\right)$$

$$L_R = \left[\frac{2.49\times10^6 * D^4}{0.381\times0.0185} + \frac{3.81\times10^3 * D^3}{0.381\times0.505} \times \sqrt{298/29}\right] * \left(1-0.01\right)$$

$$L_R = \left[3.53\times10^8 * D^4 + 6.35\times10^4 * D^3\right] * \left(0.99\right)$$

$$L_R = 3.50\times10^8 * D^4 + 6.28\times10^4 * D^3$$

Case 1

Substituting $D=7.44x10^{-5}\ cm$, a reference standard leak rate of $L_R=3.66x10^{-8}\ cm^3/s$ is calculated.

Case 2

Substituting $D=8.51x10^{-5}\ cm$, a reference standard leak rate of $L_R=5.71x10^{-8}\ cm^3/s$ is calculated.

Containment Analysis Results and Discussion

A traditional containment system must be leak-tight (i.e., leak rate $\leq 1x10^{-7}\ cm^3/s$) to transport the plutonium metal. However, the next section will demonstrate the application of a multiple barrier containment system for transport of a hypothetically breached 9975 transportation package for a shipment of short duration. Multiple barrier systems have been recently evaluated. This present work extends that of Towell, et al.[2], and shows that a barrier system of several barriers would provide sufficient containment of the suspect 9975 package.

ANALYSIS OF A MULTIPLE BARRIER SYSTEM FOR SHIPPING A HYPOTHETICALLY BREACHED 9975 TRANSPORTATION PACKAGE CONTAINING PLUTONIUM METAL

Background

Recent work[3] has been completed concerning the performance of multiple leaky barriers, i.e., barriers with individual leak rates higher than the maximum allowable leak rate for the contents being shipped, in containing radioactive materials in a shipping package. The analysis employs finite element modeling of multiple watertight barriers to demonstrate that the nested barriers will provide containment of the radioactive contents for up to several weeks without exceeding the regulatory limit of $1x10^{-6}$ A_2/hr under normal conditions of transport for type B packages. This section will present the application of the finite element model with some minor modifications to the 9975 package in the event of a breached package. The barriers are sealed materials added to the outside of the 9975 package. That is, credit is not taken for components of the damaged 9975 package.

Barrier Requirements

This analysis is limited to normal conditions of transport. The multiple leaky barriers are required to maintain a leak rate less than $1x10^{-3}$ $std \cdot cm^3/s$ as determined by ANSI N14.5. Engineering controls must be implemented that will provide relief from structural and thermal requirements. However, this analysis is not applicable in the situation where atmospheric pressure variations generate pressures on the barriers that exceed the barriers' capability to provide $1x10^{-3}$ $std \cdot cm^3/s$ containment. Furthermore, detailed consideration of minor pressure changes due to ambient temperature variations is strongly suggested for long shipping times (greater than ~ 100 days) due to model sensitivity to ambient pressure.

Finite Element Model

Towell, et al. developed a model for describing the behavior of multiple leaky barriers with respect to the transportation requirements of 10CFR71. The model consists of seven simultaneous linear differential equations for each barrier, four equations for the mass balance of the carrier gas and three equations for the mass balance of radioactive aerosol particles. This model has been modified in the present analysis to allow for pressure and radioactive material reduction in the inner barrier by flow into the next barrier. Also, the model has been expanded to include up to ten nested barriers.

Consistent with the Towell modeling work, the current analysis assumes that the carrier gas behaves as a perfect gas and that the leak hole diameter is constant and is related to the value measured in the leak test of the barrier. Following are the finite difference equations solved for the 9975 package. This matrix is set up for ten barriers. In the interest of conserving space, the finite element equations for the second through the ninth barrier are omitted from the matrix. However, they may be readily derived from Eq. 8 - Eq. 14.

$$Lj_{z+1} = \left[\frac{F_m}{\left(\frac{Pj_z + Pamb}{2}\right)} + F_c\right] \times \left(Pj_z - Pamb\right) \quad \text{(Eq. 8)}$$

$$delNRTj_{z+1} = Li_z \cdot delt \cdot Pj_z - Lj_z \cdot delt \cdot Pj_z \quad \text{(Eq. 9)}$$

$$NRTj_{z+1} = NRTj_z + delNRTj_z \quad \text{(Eq. 10)}$$

$$Pj_{z+1} = \frac{NRTj_z}{V_b} \quad \text{(Eq. 11)}$$

$$ADj_{z+1} = \min\left(\frac{sumRMj_z}{V_b}, ADa_z\right) \quad \text{(Eq. 12)}$$

$$delRMj_{z+1} = Li_z \cdot ADi_z \cdot delt - Lj_z \cdot ADj_z \cdot delt \quad \text{(Eq. 13)}$$

$$sumRMj_{z+1} = sumRMj_z + delRMj_z \quad \text{(Eq. 14)}$$

Note: If the new calculated pressure for a given barrier is less than the new calculated pressure for the next outer barrier, the new pressure for that given barrier is assigned a value equal to the average of the pressure of the given barrier in the previous time step and the next outer barrier in the current time step (i.e., $Pj_{z+1}=(Pj_z+P_{amb})/2$).

$$La_{z+1} = \left[\frac{F_m}{\left(\frac{Pa_z + Pb_z}{2}\right)} + F_c\right] \times \left(Pa_z - Pb_z\right) \quad \text{(Eq. 15)}$$

$$delNRTa_{z+1} = La_z \cdot delt \cdot Pa_z \quad \text{(Eq. 16)}$$

$$NRTa_{z+1} = NRTa_z + delNRTa_z \quad \text{(Eq. 17)}$$

$$Pa_{z+1} = \frac{NRTa_z}{V_a} \quad \text{(Eq. 18)}$$

$$ADa_{z+1} = \frac{sumRMa_z}{V_a} \quad \text{(Eq. 19)}$$

$$delRMa_{z+1} = La_z \cdot ADa_z \cdot delt \quad \text{(Eq. 20)}$$

$$sumRMa_{z+1} = sumRMa_z - delRMa_z \quad \text{(Eq. 21)}$$

$$sumRMamb_{z+1} = sumRMamb_i + Lj_z \cdot ADj_z \cdot delt \quad \text{(Eq. 22)}$$

Note: If the new calculated pressure for a given barrier is less than the new calculated pressure for the next outer barrier, the new pressure for that given barrier is assigned a value equal to the average of the pressure of the given barrier in the previous time step and the next outer barrier in the current time step (i.e., $Pa_{z+1}=(Pa_z+Pb_{z+1})/2$).

In addition to the calculation for the matrix above, the following set of calculations are carried out to determine the release rate of the multiple barrier system in $A_2/hour$.

$$RLa_z = \left(ADa_z \cdot La_z \cdot 3600 \cdot A_2 mass\right) \qquad A_2/hour$$
$$\vdots \qquad \text{(Eqs. 23)}$$
$$RLf_z = \left(ADf_z \cdot Lf_z \cdot 3600 \cdot A_2 mass\right) \qquad A_2/hour$$

The release rate of the multiple barrier system is characterized by the release rate of the outer barrier of the containment system (i.e., RLf_z). This value must be less than $1x10^{-6}\ A_2/hour$ in order to comply with the containment requirements of 10CFR71.

The following paragraphs describe the variables used in this model and the initial conditions for solving the finite element matrix.

The containers are identified by letters, "*a*" for the innermost, assigned sequentially through the outermost, "*j*" in the current case, and the ambient is identified by the symbol "amb". The solution begins with the calculation of F_c and F_m. These variables are determined by first using Eq. 5 - Eq. 7 at standard conditions to determine the aperture diameter as follows. The diameter of the leak path is determined by assuming a leak rate of each barrier equal to $1x10^{-3}\ cm^3/s$ and solving for D.

$$L = L_c + L_m$$

$$L_c = \left[\frac{2.49\times10^6 * D^4}{a\mu}\right] * \left(P_u - P_d\right) = F_c\left(P_u - P_d\right)\ cm^3/s$$

$$L_m = \left[\frac{3.81\times10^3 * D^3}{aP_a} \times \sqrt{T/M}\right] * \left(P_u - P_d\right) = F_m\left(P_u - P_d\right)\ cm^3/s$$

$$L = \left[\frac{2.49\times10^6 * D^4}{a\mu} + \frac{3.81\times10^3 * D^3}{aP_a} \times \sqrt{T/M}\right] * \left(P_u - P_d\right)$$

$$1\times10^{-3} = \left[\frac{2.49\times10^6 * D^4}{0.381\times0.0185} + \frac{3.81\times10^3 * D^3}{0.381\times0.505} \times \sqrt{298/29}\right] * \left(1 - 0.01\right)$$

$$1\times10^{-3} = 3.50\times10^8 * D^4 + 6.28\times10^4 * D^3$$

The calculated aperture diameter is $1.26x10^{-3}\ cm$. This diameter is used in the equations for F_c and F_m where the average pressure is factored out. The equations for the constants, F_c and F_m have the following form:

$$F_c = \frac{2.49\times10^6 * D^4}{a\mu} = \frac{2.49\times10^6 * \left(1.26\times10^{-3}\right)^4}{0.381\times0.0232} = 7.10x10^{-4}$$

$$F_m = \frac{3.81\times10^3 * D^3}{a}\times\sqrt{T/M} = \frac{3.81\times10^3 * \left(1.26\times10^{-3}\right)^3}{0.381}\times\sqrt{429.1/29} = 7.69\times10^{-5}$$

These are the variables defined in equations B3 and B4 of ANSI N14.5 with the average pressure across a barrier factored out of equation B4 (for F_m) so that the leakage rates of the independent barriers (La_z - Lf_z) can be solved (e.g., Eq. 8 and Eq. 15). The variables $delNRTa_z$ - $delNRTj_z$ of the finite element matrix (e.g., Eq. 9 and Eq. 16) are from the perfect gas law, $NRT = PV$, and are set equal to the product of the carrier gas leakage rate into the barrier, the finite time element, and the pressure within the barrier minus the product of the carrier gas leakage rate out of the barrier, the finite time element, and the pressure within the barrier. The variables $NRTa_z$ - $NRTj_z$ of the finite element matrix (e.g., Eq. 10 and Eq. 17) accumulate the pressure-volume product of all the gas inside the given barrier. The variables Pa_z - Pj_z of the finite element matrix (e.g., Eq. 11 and Eq. 18) converts the pressure-volume product of the gas inside the barrier to the pressure inside the barrier by dividing by the volume inside the barrier. These sets of variable equations represent the mass balance of the carrier gas within the containment system.

The following variables represent the mass balance of the radioactive aerosol particles with the containment system. The variables ADa_z - ADj_z of the finite element matrix (e.g., Eq. 12 and Eq. 19) are the densities of radioactive aerosol particles within and leaking out of the barrier in grams per square centimeter. They are equal to the mass of radioactive aerosol particles inside the barrier divided by the volume inside the barrier. The variables $delRMa_z$ - $delRMj_z$ of the finite element matrix (e.g., Eq. 13 and Eq. 20) are the masses of radioactive aerosol particles, in grams, that stay within the barrier in the finite time element. They are equal to the difference between the mass of radioactive aerosol particles leaking in from the next inner barrier minus the mass of radioactive particles leaking into the next outer barrier. The variables $sumRMa_z$ - $sumRMj_z$ of the finite element matrix (e.g., Eq. 14 and Eq. 21) are the masses of radioactive aerosol particles, in grams, that has accumulated with the barrier in all preceding finite time elements. The variable $sumRMamb_z$ of the finite element matrix (see Eq. 22) is not part of the mass balance but is added to calculate the mass of radioactive material that escapes from the outermost barrier.

The constants are the volume inside the barrier, V_a for source barrier and V_b for other barriers, the finite time interval, *delt*, and the ratio of Specific Activity to A_2, A_2mass (i.e., $\#\ A_2/gram$). V_a is determined as follows.

$$V_a = V_{PCV} + V_{SCV} + V_{Shield} \qquad \text{(Eq. 24)}$$

where:

V_{PCV} is the free volume inside the PCV [*3389 cm*^{3} from Appendix 4.1 of SAR],
V_{SCV} is the free volume inside the SCV [*2741 cm*^{3} as calculated below], and
V_{shield} is the free volume inside the shield [*11400 cm*^{3} as calculated below].

The volume between the PCV and the SCV was determined by subtracting the free volume within the PCV from the free volume within the SCV as found on Lines 8 and 10, respectively, in Table 3.20 of the SAR. This volume is *2778 cm^3*. The air volume inside the lead shield by conservatively modeling the SCV as a series of right circular cylinders and subtracting the obtained external volume from the calculated internal volume of space within the shield region. The resulting volume is *11,400 cm^3*. Substituting these values into Eq. 24 yields a value of *1.75x10^4 cm^3* for V_a.

The volume inside the other barriers, V_b, is assumed such that the total air volume in barriers outside the source barrier is equal to the arbitrary constant of *4,500 cm^3*.

The finite time element is assumed to be *10,000* seconds, and the model was evaluated at *30,240* time steps or *3,500* days. The length of time considered was chosen to ensure proper model behavior for long times.

The constant A_2mass was calculated as follows.

Case 1

$$A_2mass = \frac{\left(\frac{Ci\ in\ oxide}{\text{Grams}\ in\ oxide}\right)}{\text{A}_2} = \frac{\left(\frac{2.59}{7.48}\right)}{5.41\times10^{-3}} = 64.0\ \ {}^{A_2}\!/\!_{gram}$$

Case 2

$$A_2mass = \frac{\left(\frac{Ci\ in\ oxide}{\text{Grams}\ in\ oxide}\right)}{\text{A}_2} = \frac{\left(\frac{16.3}{7.47}\right)}{5.38\times10^{-2}} = 40.6\ \ {}^{A_2}\!/\!_{gram}$$

In the previous equations for A_2mass, the values for A_2 and for curies and grams in oxide can be found in Table 3 and Table 5, respectively, for *Case 1*, and in Table 4 and Table 6, respectively, for *Case 2*.

Initial Values

The initial values for the majority of the variables in the finite element matrix are selected to be very small but to avoid element values of zero or less so that the results can be displayed on logarithmic plots. The exceptions are as follows.

The variable La_o (Eq. 16 and Eq. 20) is set equal to the leakage rate of the barriers as measured by a leak test conducted with pressure drop of *1 atm* as prescribed in ANSI N14.5. This value is assumed equal to *1x10^{-3} cm^3/s*. The variables $NRTa_o - NRTj_o$ (e.g., Eq. 10 - Eq. 11 and Eq. 17 - Eq. 18) are set equal to the product of the volume of the given barrier and its internal pressure. The initial pressure of the source barrier, Pa_o, is set equal to *1.6 atm*, while Pb_o - Pj_o, are set equal to atmospheric pressure incremented by *0.001 atm* from the outside barrier inward. It is noted that the initial pressure is conservative in that a full puncture of the 9975 through the food can would have

relieved this pressure prior to transportation. The variables $delNRTa_o$ – $delNRTj_o$ (e.g., Eq. 10 and Eq. 17) are set equal to the pressure-volume product divided by *10 times* the finite time element. Finally, the variable ADa_o (Eq. 20) is set equal the mass of radioactive aerosol particles, calculated in Table 5 for *Case 1* and Table 6 for *Case 2*, times the adherence factor, *0.15* divided by the volume of the source barrier, V_a, calculated previously by Eq. 24. The resulting value for ADa_o is $6.40x10^{-5}$ *grams/cm*^{3} for both *Case 1* and *Case 2*.

Results and Discussion

Figure 2 display the modeling results for calculations performed to determine the limiting composition case. The figures indicate that the composition used in *Case 1* bounds that used in *Case 2*. This is consistent with the calculations of Section 0. As a result, remaining modeling is limited to *Case 1* compositions. Included in the figures are results in which the volume of each barrier, outside the source barrier, has been doubled. Comparing the results for *Case 1* with the results of *Case 1* with larger barrier volumes indicates that the container volumes used in the model are maximum volumes. This allows greater flexibility in selecting or designing the individual barriers of the multiple barrier system. In order to evaluate the effect of finite time element size, *Case 1* was modeled with significantly shorter finite time elements size (*100 sec* versus *10,000 sec*, with the results included in Figure 2. These results indicate very little sensitivity to the size of the finite time element.

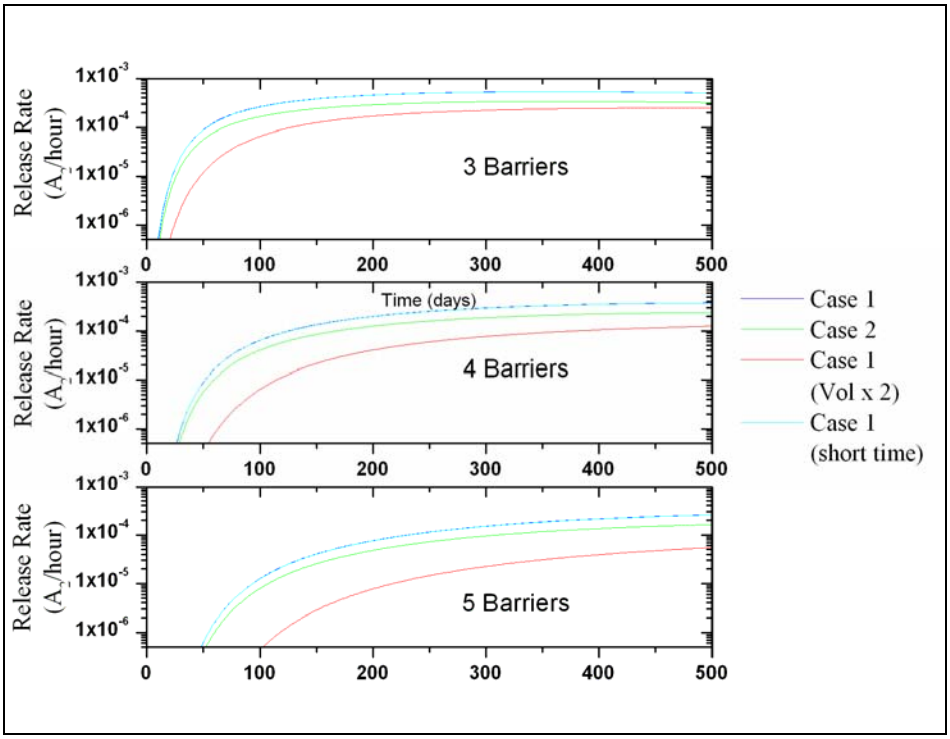

Figure 2 Modeling results comparing Case 1, Case 2, Case 1 with double barrier volume, and Case 1 with short time steps.

Figure 3 display the behavior of several model parameters. These data are for four leaky barriers. The carrier gas leak rate of the source barrier drops off rapidly as the pressure drop across its

boundary decreases. The leak rates of the individual boundaries expectedly reach equilibrium quickly and continue to decrease congruently as the pressure of the entire system is reduced toward atmospheric pressure. The density of radioactive particles within the containment system becomes uniform within a couple years. This is further illustrated in the final plot of these figures. This plot displays equal amounts of radioactive particles in the barriers surrounding the source barrier. They are equal because their volumes and particle densities are equal. The rate at which the masses of radioactive particles in the source barrier and in ambient air change continually decrease as the pressure of the source barrier approaches unity.

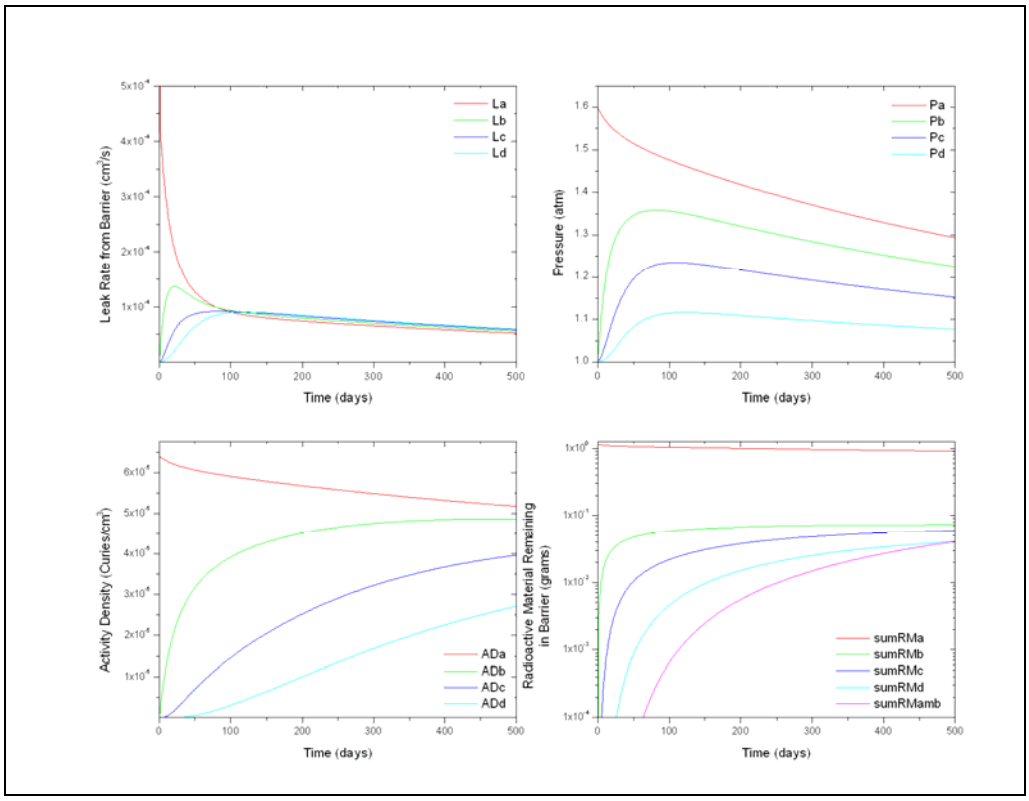

Figure 3 Carrier gas leak rates; Pressure inside boundary; Density of radioactive particles; and Radioactive particle accumulation inside boundary.

The release rates of radioactive material from the individual barriers of a four leaky barrier system are shown in Figure 4. As with the carrier gas leak rates, shown above, the radioactive material release rates quickly approach equilibrium and continue to decrease with time as the density of radioactive material available for transport and pressure drop across barriers decrease. The figures demonstrate that there is always a driving force for release, demonstrated by a positive release rate at times greater than zero. However, reduction in model conservatism allows credit for the reduction in overall release as the driving force for release, relative pressure, is reduced. The model does not consider diffusive releases that are overwhelmed by the convective releases modeled here for the time of interest for this analysis.

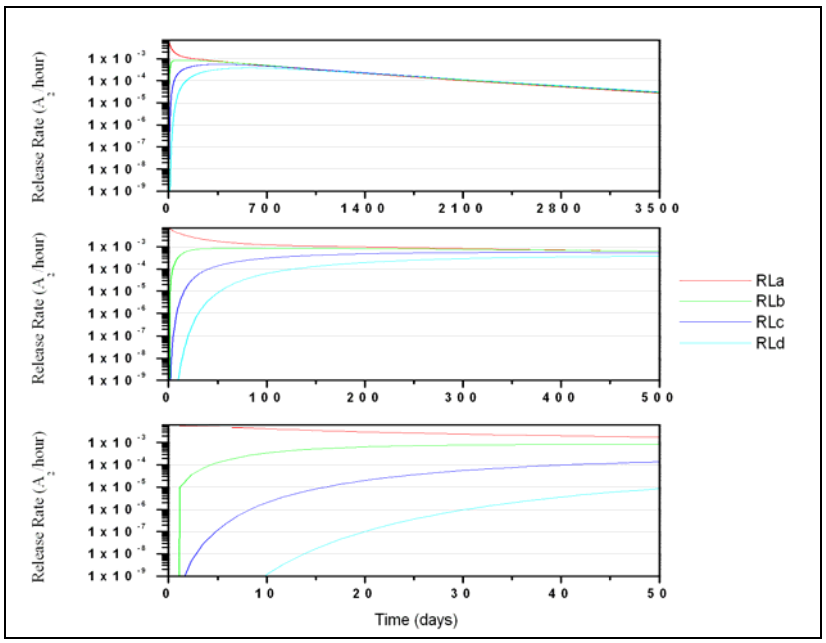

Figure 4 Radioactive material release rate for individual barriers.

Figure 5 display the radioactive material release rates for multiple leaky barrier systems. Systems of two to ten leaky barriers are considered in this analysis and are shown the the figures. The figures present data on the release of radioactive material from the outer barrier of the containment system. The regulations require a release rate of less than 10^{-6} A_2/*hour* for a shipment of radioactive material. Figure 5 shows that achieving this release rate is possible through the implementation of as few as three leaky barriers. This condition exists for about ten days, after which time, the release rate exceeds the release limit. Additional barriers will allow for significant increases in the time prior to exceeding the limit.

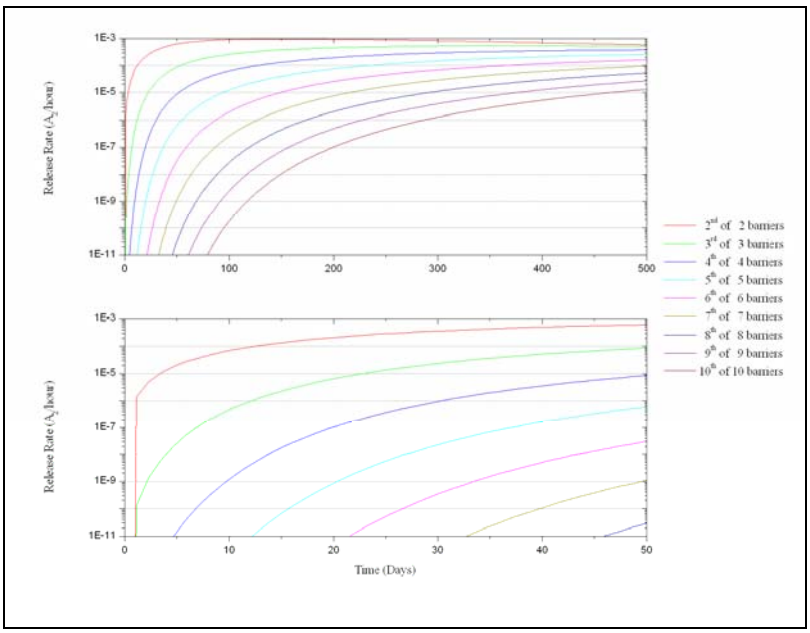

Figure 5 Radioactive material release rate for multiple leaky barrier systems.

CONCLUSIONS

Results of the containment evaluation of the 9975 package with plutonium metal contents indicate the necessity that a conventional containment system be leak-tight. In lieu of such a system, an alternative containment system has been evaluated. This system contains several redundant barriers that are individually unable to comply with the current transportation requirements. This report represents the theoretical basis for using such a containment system. The analysis provides a conservative estimation of the release rate of radionuclides from several multiple leaky barrier systems. However, it would be useful to benchmark the results of this analysis through an experimental program.

The analysis of multiple leaky barrier systems indicate that as few as three barriers with leak rates of less $1x10^{-3}$ cm^3/sec can be used successfully to maintain a containment system release rate less than $1x10^{-6}$ $A_2/hour$. The container volumes used in this analysis are nominal, less than $4{,}500$ cm^3, and increasing container volume results in longer times prior to exceeding the release limit of 10^{-6} $A_2/hour$.

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[3] Towell, R.H., Kapoor, A, Moses, S.B., and Oras, J.J., *"Method of Estimating the Leakage of Multiple Barriers in a Radioactive Materials Shipping Package,"* **Transportation, Storage, and Disposal of Radioactive Materials**, PVP-Vol. 348, ASME (1997).