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RADIOACTIVE MATERIAL RELEASE FROM A CONTAINMENT VESSEL DURING A FIRE ACCIDENT

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ABSTRACT

A methodology is presented to determine the source term for leaks and ruptures of pressurized vessels. The generic methodology is applied to a 9975 Primary Containment Vessel (PCV) which losses containment due to a hypothesized fire accident. The release due to a vessel rupture is approximately two orders of magnitude greater than the release due to a leak.

INTRODUCTION

Plutonium oxides are handled at Savannah River Site (SRS) facilities in robust containers that may pressurize under certain accident conditions such as fires. An example of such a container is the 9975 PCV shown in Figure 1. The plutonium oxide has adsorbed moisture on its surface. During a hypothesized fire accident, where the PCV is assumed to be outside of a shipping package, the container will pressurize as adsorbed moisture on the plutonium oxide evaporates. The maximum pressure within the PCV can be readily determined using a simplified analysis where the plutonium oxide and PCV are conservatively assumed to be at the fire temperature. The PCV may be structurally evaluated at this fire temperature to determine if rupture occurs. If the maximum pressure within the PCV is below the rupture pressure, then a rupture does not occur. However, the closure of the PCV is via a threaded closure and an elastomeric o-ring which will readily fail at elevated temperatures. Therefore, the PCV is assumed to release radioactive material via a leak. The subject of this paper is to compare radioactive material releases from ruptures and leaks at comparable pressures.

For the same container pressure, the respirable airborne release fraction would be expected to be reduced due to the smaller flow velocities associated with leakage through the threaded closure as opposed to a container rupture. However, the DOE handbook does not provide a means for calculating this release (i.e. leak vs. rupture) [1]. This analysis will consider the potential release from a container for pressurized rupture and leak release mechanisms.

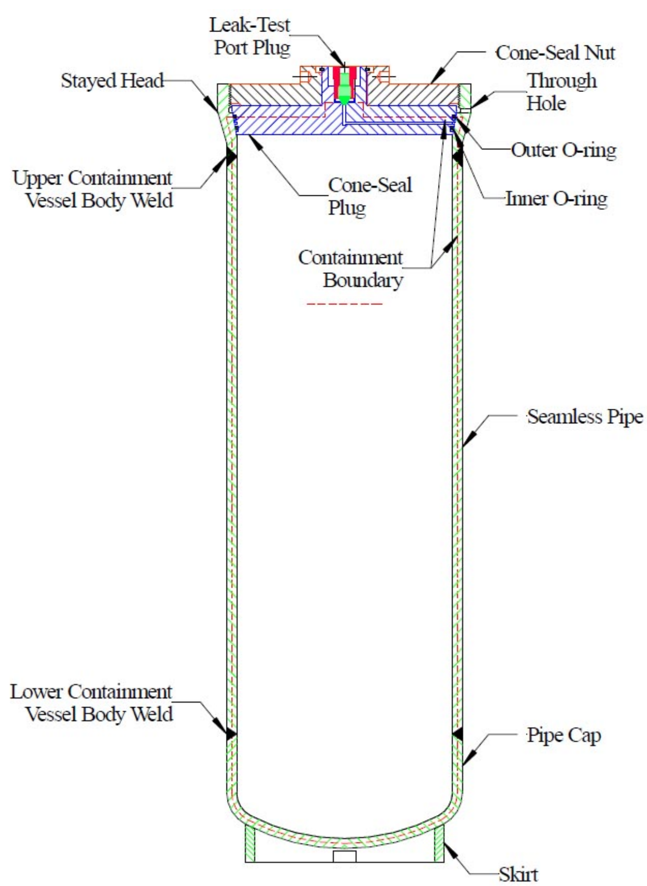

FIG 1. 9975 PRIMARY CONTAINMENT VESSEL (PCV)

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NOMENCLATURE

ARF	airborne release fraction
m	mass
P	container pressure
V	volume
v	velocity

CONTAINER RUPTURE ANALYSIS

The United States Department of Energy (DOE) Handbook provides airborne release fractions (ARF) and respirable fraction (RF) values for discrete rupture pressures (25 psig and 500 psig), and the range of these respirable airborne release is significant, 2.0E-03 and 7.0E-02, respectively [1]. These values are appropriately used to analyze events involving container ruptures. The apparatus used in the experiment to develop these values consisted of a 6-in diameter cylinder sealed with a double rupture disk. With the cylinder loaded with powder, the cylinder and the space between the two rupture disks were slowly pressurized. When the desired pressure inside the cylinder was attained, the pressure between the two rupture disks was relieved which caused them to fail catastrophically. These experiments more closely represent a container rupture than a container leak.

The United States Nuclear Regulatory Commission (NRC) developed a correlation for estimating ARFs from pressurized releases in NUREG/CR-6410 [2]. They related the ARF to the velocity of the escaping gas [Reference 2, Equation 3.17]:

$$ARF_{ub} = 2.74 \times 10^{-4} v^{1.4} \qquad (1)$$

where:

ARF_{ub} = Upper bound airborne release fraction (ARF)
v = Potential powder velocity, kinetic energy equivalent of pressurizing gas, m/s

NUREG/CR-6410 also provided a correlation for calculating the exit velocity of the escaping gas (equation 3.15):

$$v = \left(\frac{2PV}{m} \right)^{0.5} \qquad (2)$$

where:

P = Pressure above ambient, Pa, gauge
V = Container volume, including gas, m^3
m = Total mass of container contents (powder + gas), kg

Combining equations 1 and 2 shows that the ARF for a pressurized release is a function of container pressure, volume and mass. A comparison method may be used to estimate an ARF at any pressure based on a known ARF and pressure. The simple relationship is shown in equation 3.

(3)

$$\frac{ARF_2}{ARF_2} = \frac{2.74 \times 10^{-4} \left[\left(\frac{2P_2 v_{t2}}{m_2} \right)^{0.5} \right]^{1.4}}{2.74 \times 10^{-4} \left[\left(\frac{2P_1 v_{t1}}{m_1} \right)^{0.5} \right]^{1.4}} \qquad (3)$$

For the same container, the mass of the container contents would not change with an increased temperature or pressure. Note, NUREG/CR-6410 specifically refers to the container mass consisting of the powder and gas. However, the correlation (equation 2) was developed to determine the kinetic energy of the escaping gas and particles, therefore, the container mass also includes other items in the container, including any moisture on the powder. Using these concepts, equation 3 is simplified to:

$$ARF_2 = ARF_1 \left(\frac{P_2}{P_1} \right)^{0.7} \qquad (4)$$

ARFs for pressures greater than 25 psig are scaled off of the published ARF of 0.1 for 500 psig, those at or below 25 psig are scaled from 5.0E-3. Thus, equation 4 may be used to calculate a general ARF for rupture at pressure.

$$ARF = \begin{cases} 0.1 \left(\dfrac{p}{500\ psig} \right)^{0.7} & p > 25\ psig \\ 5.0 \times 10^{-3} \left(\dfrac{p}{25\ psig} \right)^{0.7} & p \le 25 psig \end{cases} \qquad (5)$$

CONTAINER LEAK ANALYSIS

Robust containers with o-ring closures, such as a 9975 PCV, may not rupture in a fire event but instead undergo a pressurized leak through a small opening or aperture (e.g., the threads of a container). It is overly conservative to analyze the release from these types of scenarios using the rupture ARF in equation 5. Instead, building upon a method described in ANSI N14.5-1997 Annex B may be used to estimate the release from a leak through a small aperture [3].

The standard provides several correlations used to calculate the upstream volumetric flow rate of gas. With a known powder aerosol mass density (concentration), the total amount powder released may be calculated using the volumetric flow rate. Note that the ANSI N14.5-1997 standard was developed primarily for shipping package leak rate determination and testing. The theoretical development of radioactive material release is based upon a maximum leak rate as the transportation regulations provide requirements for type B packages in terms of maximum allowable leak rates. In this application to a facility accident the integrated release or total radioactive material release is of primary interest. Rather than attempting to determine the duration of an accident, the approach presented here relies on an integrated volumetric release, which by definition is independent of time. An inherent conservatism in this approach is that the concentration of radioactive material in the leaking gas from the vessel is assumed constant (i.e. no credit for particulate settling or deposition), and it is assumed to be the maximum value as prescribed in ANSI N14.5-1997 of 1.0E-05 g/cm^3.

The following general steps may be used to estimate the release from a leak through a small aperture:

1. Verify the release mechanism of the event is a pressurized leak and that the container will not rupture during the event, and the release conditions are within the assumptions of ANSI N14.5-1997,
2. Obtain maximum vented gas volume from ideal gas law
3. Multiply the maximum vented gas volume by the aerosol mass density of the radioactive, material to determine the amount of released material, and
4. (Optional) Calculate the ARF for the event by dividing the amount of released material calculated in Step 3 by the total amount of material in the container.
5. With the total vented gas volume known, the amount of powder released from a container may be calculated. The mass of released powder is simply the product of the maximum vented gas volume and powder aerosol mass density. Based on experimental data, a reasonably bounding assessment of the mass density of powder aerosol is 1.0E-5 g/cm^3 [3].

$$m_R = \rho V_v = 10^{-5}\ {}^{gm}\!/\!_{cm^3}\, V_v \qquad (6)$$

where:

m_R = Mass of powder aerosol released from the container, g
ρ = Powder aerosol mass density, 1.0E-5 g/cm^3
V_v = Total vented gas volume, cm^3

Alternatively, if the desired output is the ARF, the following equation may be used:

$$ARF = 1.0E\text{-}05\ V_v/m_T \qquad (7)$$

where:

m_T = Total initial mass of powder inside the container, g

DISCUSSION

The aerosol mass density, identified in ANSI 14.5-1997, used to estimate ARFs for leaks through small apertures is based on experimental data from Curren and Bond [4]. The bulk powder used in the experiment was UO_2 with particle sizes in the range of 1 μm to 20 μm, however, an attempt was not made to quantify the particle size of the particles suspended in air. It is conservatively assumed that the aerosol mass density is composed of only particles in the respirable range (≤ 10 μm Aerodynamic Equivalent Diameter (AED)). Therefore an RF of 1 is used for leaks through small apertures, when equation 7 is utilized. Whne equation 6 is utilized the source term inherently includes the RF.

There are several conservatisms involved with this analytical method. The aerosol mass density is based on experiments simulating vehicle transportation conditions performed by Curren and Bond [4]. The value used in this analysis is based on the maximum recorded value in the Curren and Bond experiments in which "the UO_2 container was vibrated far more energetically than would be conceivable in normal transport". It is conservative to assume these conditions for pressurized release scenarios during thermal stress events.

Additionally, a constant aerosol mass density is applied in the evaluations. Schwendiman developed an empirical correlation fit to the Curren and Bond data to calculate the aerosol mass density as a function of settling velocity [5]. During an accident scenario the duration of venting may be in the range of seconds to minutes. When using the Schwendiman equation, the aerosol mass density is less than 40% of the maximum value after 10 seconds of settling. After a minute of settling the aerosol mass density is less than 1% of the maximum. If it was assumed the particles began to settle at the beginning of venting, the overall release of powder would be much less. Schwendiman also reviewed other mechanisms which reduce the amount of powders released through small apertures. In his review he discusses gravitational settling, turbulent impaction, Brownian Diffusion, electrostatics and thermophoretic deposition. These phenomena are conservatively not considered in the method presented. It should be noted that settling is not credited in the ANSI 14.5 methodology [3].

Finally, the leakage paths provide very small gaps in which the particles have to transverse. It is likely that these small particles will impact the gap surface and begin to plug the release path and reduce the particle leakage from the container. No credit for plugging has been taken. Similarly, the pressurization experienced by the vessel is not subject to a reduction due to leakage. A typical application of this method is to first conservatively raise the temperature of the plutonium oxide and vessel to the fire temperature and then determine the pressure due to evaporated moisture and initial fill gas pressurization without regard for any leakage. Once it is determined that the vessel does not rupture, then the release due to leak methodology is applied.

APPLICATION TO 9975 PCV

The 9975 is used to transport and store plutonium oxide materials at SRS. In order to retrieve the materials for processing the 9975 must be opened and the PCV removed. A hypothesized facility fire accident is assumed to occur while the unopened PCV is outside the 9975 shipping package. In general, there are four different packaging configurations utilized at SRS for storage. Three involve bulk plutonium oxide quantities while the fourth involves sources/standards. The facility fire of interest is an 800°C fire. Although the fire is of limited duration, for simplicity, the PCV and plutonium oxide contents are conservatively assumed to be at 800°C. The PCV is first shown not to rupture (rupture pressure is 2033 psig at 800°C). Subsequently, the leak methodology is applied. The summarized results are presented in Table 1.

As seen in Table 1, the ratio of burst based ARF to leak based ARF is consistently greater than two orders of magnitude. Note that the maximum quantity of plutonium oxide was analyzed for each packaging configuration. This results in the highest PCV peak pressure (pressure is dependent on total moisture content).

Table 1 Ratio of ARF Reduction

Configuration	Pu oxide mass (gm)	PCV Peak Pressure (psig)	PCV Free Volume (cm^3)	Vented Volume (cm^3)	Aerosol vented (gm) [Eqn. 6]	ARF [Eqn. 7]	Burst ARF [Eqn. 5]	Ratio (Eqn. 5/Eqn. 7)
Type 1 can in PCV	5000	1163	4433	3.55E+05	3.552	7.10 E-04	0.181	254.20
Type 2vented cans in PCV	5000	1474	3631	3.68E+05	3.677	7.35 E-04	0.213	289.817
Type 3 vented cans in PCV	5000	1965	3963	5.34E+05	5.337	1.07 E-03	0.261	244.196
Source/Standard in PCV	2000	256	4726	8.70E+04	0.870	4.35 E-04	0.063	143.832

CONCLUSIONS

A methodology has been presented to analyze a release resulting from the depressurization of a container by the rupture or leaking of high pressure gases. Using this method, it was demonstrated that the ARF×RF from a leak for a PCV is bounded by the rupture ARF×RF reduced by a factor of over 140 for all the cases evaluated. It must be shown that the container does not rupture due to pressurization from the fire event in order to use this methodology. In addition, an RF of unity must be used when applying this methodology utilizing a leak based ARF.

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