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## A SIMPLE METHOD TO ESTIMATE AIRBORNE RELEASE FRACTIONS ASSOCIATED WITH THE PRESSURIZED RELEASE OF RADIOACTIVE MATERIALS

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### ABSTRACT

Pressurized release of radioactive materials, such as plutonium bearing oxides, could occur during a fire accident event where a robust container holding the radioactive materials ruptures catastrophically due to excessive pressurization. The common source of pressurization is the vaporization of water adsorbed onto the plutonium bearing oxides. A simplified pressure based approach derived from NUREG/CR-6410 is presented which allows calculation of airborne release fractions for pressures up to and above 500 psig. A parametric study is used to determine a bias (conservative or non-conservative) when the approach is applied to DOE-STD-3013 welded stainless steel containers which are used to store plutonium bearing oxides at the US Department of Energy Savannah River Site. The event considered is a 3013 container subjected to a hypothesized facility fire where the container fails catastrophically and ruptures resulting in a vented high pressure release of the plutonium bearing oxide materials. The parametric study shows that the simple correlation for calculating airborne release fractions is biased high by about 40% compared with the detailed NUREG/CR-6410 method. Care should be taken in using the simplified approach where the ratio of vessel net free volume to maximum allowed oxide mass is greater than 0.66 cc/g.

### INTRODUCTION

Radioactive materials, such as plutonium bearing oxides, are stored in 3013 containers at the US Department of Energy Savannah River Site (SRS) near Aiken, South Carolina. The 3013 containers are packaged in 9975 shipping packages which are palletized for long term storage. Figure 1 provides a photograph of typical 3103 containers used to package plutonium oxide bearing materials. The triple nested 3013 containers are each made of stainless steel. The outermost 3013 container is designed to the ASME BPV Code with a 699 psig design pressure and utilizes a welded closure [1]. The second layer container, referred to as the inner 3013 container, also utilizes a welded closure, however, the innermost container, referred to as the 3013 convenience container, contains the plutonium oxide materials and utilizes a screw lid closure. The innermost container is designed to be leaky to allow for purging and backfilling of helium gas in both the convenience and inner container prior to the inner container being welded. After welding, the inner container is then leak tested. Subsequently, the welded inner container is placed inside the outer most container which is purged and backfilled with helium prior to being welded. The outer container is also leak tested and radiography is used to verify that the outer container weld is free of defects.

The 3013 containers are stored in 9975 shipping packages until the plutonium oxide materials are dispositioned via down blending with an inert adulterant at SRS, packaged in authorized waste containers, and transported to the Waste Isolation Pilot Plant (WIPP) for final geologic disposal. Figure 2 illustrates the 9975 shipping container by showing its two nested containment vessels, lead shielding, and fiberboard overpack within an outer stainless steel 35 gallon drum; the 3013 container is not shown. These 9975 shipping packages are palletized and stacked to store 3013 containers at the Savannah River Site as seen in Figure 3. In order to initiate the down blending process, the 3013 must be removed from the 9975 and introduced into a glove box where each welded 3013 container is cut open.

Figure 1: 3013 Outer, Inner, and Convenience Containers

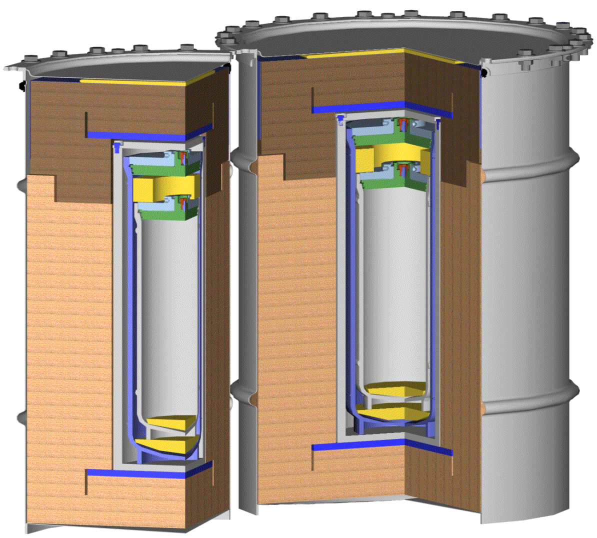

Figure 2: 9975 Shipping Package

A hypothesized pressurized release of plutonium bearing oxide could occur should the 3013 container outside of the 9975 shipping package and prior to being cut open in the glove box be subjected to a fire event. During a hypothetical fire event the 3013 container would pressurize due to evaporation of adsorbed moisture on the plutonium oxide materials and heating of the 3013 fill gases. Additionally, the fire would severely weaken the 3013 outer container to the extent that it may fail (e.g. rupture) catastrophically. The purpose is to provide the first publication of a simple method to estimate the airborne release fraction associated with a pressurized release from a 3013 container.

**PRESSURIZED RELEASE OF PLUTONIUM OXIDE**

Pressurized release of plutonium oxides could occur during a fire accident event where the container holding the radioactive materials ruptures catastrophically due to excessive pressurization. Limited data exists regarding the fraction of oxide that becomes airborne upon pressurized failure of containers. However, the US Nuclear Regulatory Commission has published a regulatory document, NUREG/CR-6410, which provides a first principle framework for estimating airborne release fractions due to a pressurized release [2]. The US Department of Energy Standard DOE-HDBK-3010-94 discusses venting of pressurized powders or pressurized gases through a powder and provides bounding airborne release fractions (ARF) and respirable fractions at 25 and 500 psig [3]. The first principal basis from NUREG/CR-6410 will be used to develop a simple pressure based equation for the ARF as a function of vessel failure pressure, and the equation will necessarily include the values of the bounding airborne release fraction from DOE-HDBK-3010-94 at 25 and/or 500 psig which are 5E-03 and 1E-01, respectively.

Figure 3: 9975 Shipping Packages At Savannah River Site

**NUREG/CR-6410**

A correlation for the ARF as a function of potential powder velocity is provided in NUREG/CR-6410 where the potential powder velocity is based on the energy of the pressurized gas,

$$v_p = \left(\frac{2PV}{m}\right)^{1/2} \qquad (1)$$

where $v_p$ is the potential powder velocity, $P$ is the container failure pressure, $V$ is the container gas volume, and $m$ is the total mass of the plutonium oxide contents. The NUREG references a limited amount of pressurized release experimental data to establish the following upper bound correlation,

$$ARF_{ub} = 2.74E - 04\ v_p^{1.4} \qquad (2)$$

and a best fit or best estimate correlation.

$$ARF = 1.0E - 04\ v_p^{1.4} \qquad (3)$$

Both correlations are power law based with an exponent of 1.4.

## DERIVATION OF SIMPLIFIED EQUATION

Equations 1 and 2 (equation 3 could also be used) are used to determine the ARF for a failed container due to over pressurization. Consider the ARF for a container whose failure pressure $P_1$ (psig) and the ARF if the failure pressure is 500 psig. The ratio of the two ARF's is considered in equations 4 and 5.

$$\frac{ARF_{P1}}{ARF_{500}} = \frac{2.74E{-}04\left(v_{p1}\right)^{1.4}}{2.74E{-}04(v_{500})^{1.4}} \qquad (4)$$

$$\frac{ARF_{P1}}{ARF_{500}} = \frac{(2P_1V/m)^{0.7}}{(2*500*V/m)^{0.7}} = \left(\frac{P_1}{500}\right)^{0.7} \qquad (5)$$

However, the ARF at 500 psig is 0.1 per DOE-HDBK-3010-94.

$$\frac{ARF_{P1}}{ARF_{500}} = \frac{ARF_{P1}}{0.1} \qquad (6)$$

A simplified equation for the ARF at $P_1$ is readily determined by equating the right hand sides of equations 5 and 6.

$$ARF_{P1} = 1E - 01\ (\frac{P_1}{500})^{0.7} \qquad (7)$$

The same analysis can be performed using the ARF value of 5E-03 at 25 psig rather than 1E-01 at 500 psig.

$$ARF_{P1} = 5E - 03\ (\frac{P_1}{25})^{0.7} \qquad (8)$$

This approach assumes the content mass and container volume are relatively constant at increased pressure.

## ASSESSMENT OF SIMPLIFIED EQUATION

Substituting a pressure of 25 psig into equation 7 and 500 psig into equation 8 yields the seemingly contradictory results. An ARF of 1.23E-02 at 25 psig results when using equation 7, rather than 5E-03, and an ARF of 4.1E-02 at 500 psig results when using equation 8 rather than 1E-01. Mathematically, the result indicates that the two points, 5E-03 at 25 psig and 1E-01 at 500 psig do not both fall upon the same curve where ARF is a function of the pressure raised to the 0.7 power. An ARF of 1.23E-02 at 25 psig should provide a similar level of conservatism as 1E-01 at 500 psig since these two points do fall upon a power curve where ARF is a function of the pressure raised to the 0.7 power. Note that the 0.7 exponent in equation 7 is an artifact of the curve fit of ARF data with velocity shown in equations 2 and 3. The NUREG does not propose a greater exponent in the correlation for conservatism, but rather proposes different coefficients to the correlations in equations 2 and 3 in order to provide additional conservatism for the upper bound correlation. Therefore, equation 7 is judged to be an appropriately conservative correlation for ARF as a function of container rupture pressure in so much as the ARF value of 1E-01 is deemed to be appropriately conservative for a container rupture pressure of 500 psig.

## APPLCIATION TO 3013 A CONTAINER RUPTURE

Equations 1 – 3 and equation 7 can be used to determine the ARF for a 3013 container that ruptures during an accidental fire event. The free volume of an outer 3013 container varies depending on the inner and convenience containers used. The largest free volume is 2.432 liters or 2432 cc [1], and the theoretical density or crystalline density of pure plutonium oxide is 11.5 g/cc [1]. The free volume of a 3013 container as a function of plutonium oxide mass is given in equation 9,

$$V = 2432 - m/11.5 \qquad (9)$$

where $V$ is the net container free volume in cc and $m$ is the mass of plutonium oxide in grams (M denotes mass in Kg).

The potential powder velocity can be determined as a function of plutonium oxide mass and 3013 rupture pressure using equation 1. An upper bound and best estimate ARF can then be determined based on the powder velocity using equations 2 and 3. These results are presented in Table 1 for pressures ranging from 250 to 1000 psig. As seen in Table 1, the difference between the upper bound ARF and the best estimate ARF is substantial. As expected, a higher rupture pressure results in substantially higher ARF for the same plutonium oxide mass, while increasing plutonium oxide mass decreases the ARF for a given rupture pressure.

In applying these results, the parameter of interest is the product of the plutonium oxide mass, referred to as the material at risk, and the ARF. This product provides a mass quantity of material released. Table 2 shows how the mass of plutonium oxide material released varies with container release pressure and material mass using both the upper bound (equation 2) and best estimate (equation 3) correlation equations.

As seen in Table 2, the amount of material released (product of ARF and oxide mass) due to a pressurized rupture increases with increasing oxide mass in the container even though the ARF decreases with increasing oxide mass. Based on equations 1 and 2 or 3 the ARF varies approximately as shown in equation 10. The variation is approximate and not exact due to the effect of decreasing net free volume with increasing oxide mass per equation 9.

$$ARF\ \alpha\ M^{-0.7} \qquad (10)$$

Therefore, the product of ARF and M increases with plutonium oxide mass, and the calculation of interest for a safety evaluation should be based on the maximum allowed plutonium oxide mass in the container. Per DOE-STD-3013 the 3013 container is restricted to no more than 5 Kg of plutonium oxide.

Table 1: ARF As A Function of 3013 Rupture Pressure

| $P$ (psig) | M (Kg) | $V_p$ (m/s) | $ARF_{ub}$ | $ARF_{be}$ |
|---|---|---|---|---|
| 250 | 1 | 89.9 | 0.149 | 0.054 |
| 250 | 2 | 62.4 | 0.089 | 0.033 |
| 250 | 3 | 49.9 | 0.065 | 0.024 |
| 250 | 4 | 47.8 | 0.062 | 0.023 |
| 250 | 5 | 37.1 | 0.043 | 0.016 |
| 500 | 1 | 127 | 0.242 | 0.088 |
| 500 | 2 | 88.2 | 0.145 | 0.053 |
| 500 | 3 | 70.6 | 0.106 | 0.039 |
| 500 | 4 | 59.9 | 0.084 | 0.031 |
| 500 | 5 | 52.5 | 0.070 | 0.026 |
| 750 | 1 | 156 | 0.321 | 0.117 |
| 750 | 2 | 108 | 0.193 | 0.070 |
| 750 | 3 | 86.5 | 0.141 | 0.052 |
| 750 | 4 | 73.4 | 0.112 | 0.041 |
| 750 | 5 | 64.3 | 0.093 | 0.034 |
| 1000 | 1 | 180 | 0.393 | 0.143 |
| 1000 | 2 | 125 | 0.236 | 0.086 |
| 1000 | 3 | 99.9 | 0.173 | 0.063 |
| 1000 | 4 | 84.8 | 0.137 | 0.050 |
| 1000 | 5 | 74.2 | 0.114 | 0.042 |

Table 2: Oxide Mass Released

| $P$ (psig) | M (Kg) | M *$ARF_{ub}$ (Kg) | M *$ARF_{be}$ (Kg) |
|---|---|---|---|
| 250 | 1 | 0.149 | 0.054 |
| 250 | 2 | 0.179 | 0.065 |
| 250 | 3 | 0.196 | 0.072 |
| 250 | 4 | 0.199 | 0.073 |
| 250 | 5 | 0.216 | 0.079 |
| 500 | 1 | 0.242 | 0.088 |
| 500 | 2 | 0.290 | 0.106 |
| 500 | 3 | 0.319 | 0.116 |
| 500 | 4 | 0.338 | 0.123 |
| 500 | 5 | 0.351 | 0.128 |
| 750 | 1 | 0.321 | 0.117 |
| 750 | 2 | 0.385 | 0.141 |
| 750 | 3 | 0.423 | 0.155 |
| 750 | 4 | 0.449 | 0.164 |
| 750 | 5 | 0.466 | 0.170 |
| 1000 | 1 | 0.393 | 0.143 |
| 1000 | 2 | 0.471 | 0.172 |
| 1000 | 3 | 0.518 | 0.189 |
| 1000 | 4 | 0.549 | 0.200 |
| 1000 | 5 | 0.569 | 0.208 |

The simplified approach based on equation 7 is compared with the results from Table 2. Recall, the simplified approach is based on the conservative ARF of 0.1 at 500 psig and the exponential variation with pressure derived from equations 1 and 2 from NUREG/CR-6410 while Table 2 results are based solely on the NUREG/CR-6410 approach (equations 1 - 3). The comparison is provided in Table 3 of the material released using the upper bound results, equation 2, and the simplified approach, equation 7, for a plutonium oxide mass of 5 Kg.

Table 3: Oxide Mass Released As A Function of Pressure

| $P$ (psig) | M *$ARF_{ub}$ (eq. 2) (Kg) | M *$ARF_{P1}$ (eq. 7) (Kg) |
|---|---|---|
| 250 | 0.216 | 0.308 |
| 500 | 0.351 | 0.500 |
| 750 | 0.466 | 0.664 |
| 1000 | 0.569 | 0.812 |

Note: mass of oxide evaluated is 5 Kg

As seen in Table 3 the simplified approach results in a greater ARF compared with the upper bound correlation methodology of NUREG/CR-6410 by approximately 40% in determining the quantity of plutonium oxide released when 5 kg of oxide is evaluated. Care should be taken in using the simplified approach for other containers where the ratio of vessel free volume to maximum allowed oxide mass is higher. An estimate for the point where equations 1 and 2 and equation 7 provide the same resulting amount of oxide released is about 3.2 Kg of oxide within a 3013 container which is a net free volume to mass ratio of approximately 0.66 cc/g. A greater net free volume to oxide mass ratio results in the simplified approach (equation 7) underpredicting the ARF relative to the upper bound correlation methodology of NUREG/CR-6410.

## CONCLUSIONS

A simplified approach, based upon NUREG/CR-6410, is presented which allows calculation of airborne release fractions due to a catastrophic failure of a container for pressures up to and above 500 psig. A parametric study of 3013 containers demonstrates the simple methodology over estimates the ARF for a 3013 container with 5 Kg of plutonium oxide by approximately 40%. Care should be taken in using the simplified approach where the ratio of vessel net free volume to maximum allowed oxide mass is greater than 0.66 cc/g.

## REFERENCES

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