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Respirable Release Fraction Measurement Chamber

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ABSTRACT

The Respirable Release Fraction Measurement Chamber (RRFMC) is a 6.10 m tall, 1.42×1.02 m inside footprint, HEPA filtered, airtight drop tower. The maximum drop height is 4.88 m and maximum drop mass is 68.0 kg. This unique capability uses cerium oxide as a surrogate for plutonium oxide to measure aerosol release from a dropped nuclear material storage container (NMSC). The RRFMC directly measures Airborne Release Fraction (ARF) and Respirable Release Fraction (RRF) under various accident conditions. It has two hi-speed video cameras, located on orthogonal axes, at the impact zone. The measurements of the background particulate concentration, aerosol transmission ratio, test powder effective density and experimental uncertainty of the system have been reported previously, and these measurements are currently being further refined. The RRFMC operations are qualified for NQA-1 2008, 2009 addendum to sub part 2.4 Research and Development. The spilled mass (ms), RRF, Spilled Uptake Mass (SUM), and Spilled Uptake Factor (SUF) were measured for selected containers. The RRFMC results are described and compared to those obtained by Gao, Zhang and Byington (2013), who studied the uptake factor of tungsten oxide (a surrogate for uranium) particles resulting from dropped containers.

KEYWORDS: nuclear material, storage container, respirable release fraction, drop tower, spilled uptake mass, spilled uptake factor, cerium oxide

INTRODUCTION

Thousands of nuclear material storage containers (NMSC) are currently used throughout the US Department of Energy complex. These containers are designed to protect nuclear facility workers from an internal exposure to radioactive material during usage and in case of an accident. One accident scenario is that a dropped NMSC releases respirable radioactive material that could be inhaled by co-located worker. The U.S. DOE M441.1-1 requires the container package to have a post drop design qualification release rate that will prevent the exposure of the worker to greater than 5 rem CED[1]. The RRFMC is designed to perform drop tests to evaluate container packages to ensure that they meet this post drop design qualification release rate. The purpose of this study is to (1) describe the current status of the RRFMC capability, (2) report preliminary results for drops of containers loaded with cerium oxide as a surrogate for plutonium, and (3) compare study results with for cerium oxide to those obtained by Gao et al[2] for tungsten oxide (uranium oxide surrogate).

METHODS

Respirable Release Fraction Measurement Chamber (RRFMC)

The RRFMC is an airtight 6.10 m tall PermaCon^{TM} enclosure (Pajarito Scientific Inc, Santa Fe, NM) integrated into a closed loop wind tunnel (Figure 1). The impact zone (1.42×1.02 m inside footprint) has upstream and downstream HEPA filters and operates at a neutral pressure compared to ambient.

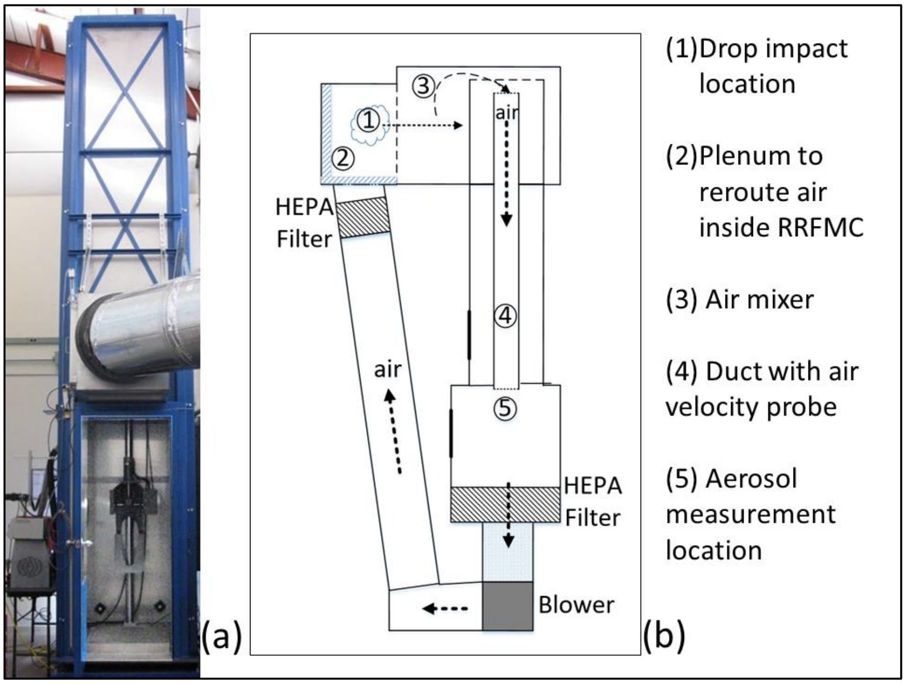

Figure 1. (a) Lansmont PDT 80 drop tester inside RRFMC PermaCon^{TM} enclosure, (b) Top view of the schematic floor plan and wind tunnel with RRFMC.

A remote controlled drop tester (Model PDT 80, Lansmont Inc, Monterey, CA) in the enclosure (Figure 1a) provides a variable test height, h, up to 4.88 m, and test items can weigh up to 68.0 kg. Two MEMRECAM HX-7 cameras (nac Image Technology USA, Simi Valley, CA) provide dual-axis high-speed videography. The cerium oxide test powder was selected as a plutonium oxide surrogate based on previous work at several US national labs[3] [note: Los Alamos Unrestricted Release are available at www.osti.gov.].

Containers are dropped in the RRFMC during active filtration of airflow. For released powder at location #1, entrained powder in the airflow passes into a generic mixer[4] #3 and through a duct #4 for single point sampling[5,6]. Real-time aerosol measurements for aerodynamic equivalent diameter (AED) from 0.5 to 20 µm are performed with an Aerodynamic Particle Sizer (APS, TSI Inc., Shoreview, MN) at location #5. The test air passes into a bank of HEPA filters, through the fan, through another HEPA filter and returns into the RRFMC. The airflow is routed through a plenum #2 that sweeps a curtain of purified air across the impact zone surface #1.

Calculation Method

Before a test, the RRFMC and wind tunnel are vacuumed and cleaned with a damp wipe to reduce background dust (dust is either natural or artificial background aerosol; powder is the analytical product input). The tested container is loaded in the desired drop orientation and the RRFMC door is closed. The aerosol concentration inside the RRFMC is monitored until it reaches the defined equilibrium background concentration of 10^{-8} g m^{-3}. The tested container is raised to the drop height and dropped by remote control. An increase of aerosol concentration is detected by the APS, whether there is a release of test powder or not, since even the drop of a clean control container (with no test powder) will dislodge and re-entrain dust inside the RRFMC. After impact, aerosol concentration is monitored until the background value is reached. Internal deposition losses as a function of AED were separately determined by measuring aerosol transmission from the impact area to the measurement location with monodisperse particles[7]. The transmission ratio for every APS sampling channel, P_j, was then calculated by fitting the experimental data[7].

The respirable mass (AED<10 μm) of aerosol released from a tested container, *mr(g)* is:

$$mr = FR * Ts * \sum_{i=1}^{t}\left[\sum_{j=1}^{D_a=10}\left(\frac{C_{ij}}{P_j}\right)\right] \quad (1)$$

where:
C_{ij} = the measured aerosol concentration at $t=i$ in APS sampling channel j (g m^{-3}),
P_j = the transmission ratio in each APS sampling channel j,
FR = RRFMC airflow at location #4 (m^3 min^{-1}), (Velocicalc 9565-P, TSI Inc., Shoreview, MN),
Ts = the APS sampling interval (1 min),
$t=1$ is when the container is dropped, and
$t=t$ is when the measured concentration returns to the background concentration.

The airborne mass (AED<20 μm) of aerosol released from a tested container, *ma(g)* is:

$$ma = FR * Ts * \sum_{i=1}^{t}\left[\sum_{j=1}^{D_a=20}\left(\frac{C_{ij}}{P_j}\right)\right] \quad (2)$$

The APS is a time-of-flight spectrometer that measures the velocity of particles in an accelerating air flow through a nozzle. Particles with different density or shape will be accelerated and therefore sized differently in the APS instrument. The APS converts each time-of-flight measurement to an aerodynamic particle diameter, D_a. Mass concentration is calculated from the particle number concentration in each APS sampling channel and the particle effective density[8]. The measured effective density value for CeO_2 is 0.22 ± 0.10 g cm^{-3}, from combined APS and cascade impactor measurements[9]. The effective density is the ratio of the particle density to the particle dynamic shape factor[10], and the dynamic shape factor is the ratio of the actual resistance force of a nonspherical irregular particle to the resistance force of a sphere having the same terminal settling velocity and volume[11].

Previous DOE guidance for analysis of discrete events are determined using a five-component equation[12]:

$$Source\ Term = S = MAR * DR * ARF * RF * LPF \quad (3)$$

The different terms are MAR = material at risk, ARF = airborne release fraction, DR = damage ratio, RF = respirable fraction, and LPF = leak path factor. Given these definitions, it is useful to group DR, ARF, and RF as the Respirable Release Fraction[13]:

$$RRF = DR \cdot ARF \cdot RF = \frac{mr}{MAR} \qquad (4)$$

The spilled mass, *ms(g)*, is calculated as the weight difference of the tested container before and after the drop test.

RRFMC Aerosol Background Due to Physical Impacts

In a drop test of a clean control container (with the same potential energy, PE=mgh, of the tested container but without test powder), background dust on the internal wall surfaces of the RRFMC is re-suspended (Figure 2). For each tested container, this increase of re-suspended background aerosol was measured by dropping three replicate controls. The dust background uncertainty was calculated from the system uncertainty and the replicate control standard deviation. Corresponding background masses were subtracted from the measured respirable and airborne masses for tested containers (with test powder), matching the potential energy to yield net respirable mass, *netmr(g)*, and net airborne mass, *netma(g)*.

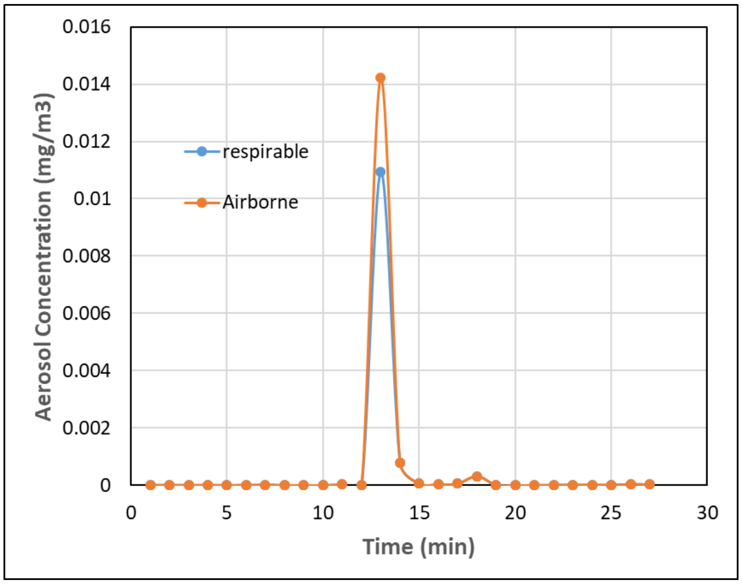

Figure 2. Aerosol concentration spike measured at a 324 J clean control drop

Spilled Uptake Mass (SUM) and Spilled Uptake Factor (SUF) Calculation

The SUM (spilled uptake mass) estimates worker inhalation of respirable particles from a dropped container[2]. In a room of volume, $V(m^3)$, with a mass concentration, $C_t\ (g\ m^{-3})$, for a standard breathing rate, $Q(m^3\ h^{-1})$, over time, $t\ (h)$, the SUM is:

$$SUM = Q \cdot \int_0^t C_t\, dt \qquad (5)$$

The aerosol concentration in a ventilated room decreases exponentially over time, based on the volume and ventilation rate. The number of air changes per hour in a room, $A(h^{-1})$, is a measure of the air volume added to or removed from a space per unit time[14].

The net respirable mass, *netmr(g)*, is assumed to instantly disperse and mix throughout a volume, V, to produce an initial mass concentration, C_0, in the room. The aerosol concentration, C_t, (at time, t) is then[14]:

$$C_t = C_0 \cdot \exp(-At) = \frac{netmr}{V} \cdot \exp(-At) \qquad (6)$$

By substitution and integration, SUM(g) is therefore:

$$SUM = Q \cdot \frac{netmr}{A \cdot V} \cdot (1 - \exp(-At)) \qquad (7)$$

For calculations with the Los Alamos RRFMC:

$netmr$ = the net released respirable mass (g),
Q = standard person breathing rate (1.2 m^3 h^{-1}),
A = ventilation rate of the room, selected for LANL operational reasons, (8.0 h^{-1})
V = room volume (70.8 m^3), selected for LANL operational reasons, and,
t = time duration for a person in the room after the container is dropped (0.167 h).

The calculation is therefore a time-averaged, volume-averaged uptake mass. This accounts for container damage, room volume, room ventilation, and standard worker respiration, but does not account for non-uniformity of aerosol dispersal in a room after a drop, or details of air movement patterns in the room. Note the SUM calculation method and test configuration in this study were different from Gao et al[2]. Their calculation method used the directly measured airborne concentration at a nearby location of the drop event (1.5 m above the floor, 0.15m away from the falling path)[2].

The spill uptake factor (SUF) is calculated by dividing the SUM by the spilled mass[2]:

$$SUF = \frac{SUM}{\mathrm{ms}} \qquad (8)$$

DROP TEST CONDITIONS

A 2-gal pail (The Cary Co., Addison IL) was used as container (0.23 m diameter and 0.28 m height) for the results reported in this paper. Figure 3a shows a picture of the 2-gal Cary pail and lid. The lid has 12 tabs, and in each drop with lid, only three of the tabs were closed to ensure a significant release of cerium oxide from the container.

The particle size of the CeO_2 powder is AED ≤ 1 µm (CAS#:1306-38-3; American Elements, Los Angeles, CA; Lot#: 11815114547-915) according to the manufacturer's Certificate of Analysis. All the tested containers were loaded with payload and dried CeO_2 test powder. Before each test, the CeO_2 test powder was dried at 110 ^{o}C for 2 hours to remove moisture. The payload was an 8.66 kg steel cylinder. Dried test powder (between 3.4 to 5.5 g CeO_2) was placed into the test pail on the top of the pay load. The gross weight of tested object was about 9.46 kg. After test powder was loaded, except for Test 4, the lid was put on and only 3 of 12 tabs of the lid were crimped down. The container to be tested was then loaded on the drop tester (Figure 3b).

The particle size distributions of powder dispersed from a Wright Dust Feeder (CH Technologies Inc, Westwood, NJ) and a dropped container (Test 4, the only no-lid test) were measured. The Wright Dust Feeder is an instrument that uses a Venturi outlet and carbide cutter head for superior de-agglomeration at the time of aerosol generation.

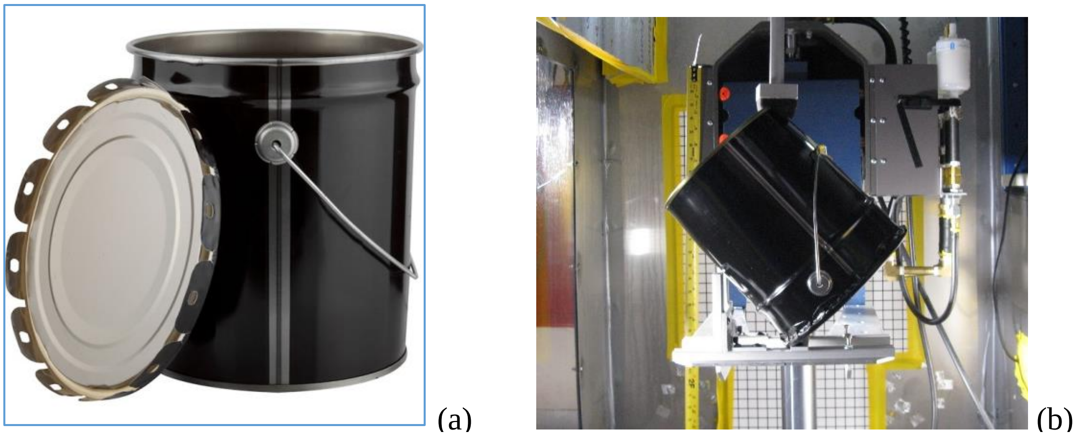

Figure 3. Container that was drop tested in the RRFMC: (a) test pail (b) loaded on drop tester, CG over top corner orientation.

The drop height was directly set in the Lansmont PDT-80 drop tester and verified by a NIST traceable tape measure (resolution 0.00254 m). The mass of test powder (CeO_2, MAR), pre-drop gross mass, and post-drop gross mass were measured on a Mettler SR64001 mass balance (resolution 0.1 g). All the containers were dropped in the orientation of CG (center of gravity) over the top corner (most vulnerable) orientation with lid on, except for Test 4 which was dropped without a lid at CG over bottom corner orientation.

The CG of tested containers was manually determined by the operator before the drop test. The spilled mass was measured according to the mass difference of pre-drop gross mass and post-drop gross mass. After a drop, the lid and payload were put back into the container and weighed again for the post-drop gross mass.

The respirable mass, *mr*, and airborne mass, *ma*, were calculated (Eq 1 and 2) with the correction factor for aerosol transmission and 0.22 ± 0.10 g cm^{-3} effective density. Average backgrounds for three control drops at the same drop potential energy were subtracted from the gross to yield the net values. The RRF was calculated (Eq 4) using the net respirable mass, *netmr*, values (Table 1) and the SUM and SUF (Eq 7 and 8) were calculated. (Table 1).

TEST RESULTS

The particle size distribution of CeO_2 test powder released (Figure 4, orange dash curve) from the dropped container (Test 4 Peak) indicates agglomeration of the primary particles into larger sizes compared to the particle size distribution from the Wright Dust Feeder which breaks the particle agglomeration apart by mechanical grinding and pressurized air jets.

In comparing the measured results (Table 1), the spilled masses from the dropped containers varied from 0.2 g to 2.8 g. Note that Test 2 has the lowest spilled mass (the case when the lid remained attached, Figure 5b). Test 4 has less spilled mass than Test 1, 3, 5 and 6, although it was dropped without a lid with the CG over the bottom corner orientation. For tests with the CG over top corner orientation, the test powder would be in direct contact with the inner lid surface and therefore more available for release from the container during the impact.

For tests with a lid on the pail (Test 1-3, 5-6), the lid generally popped open during the impact (Figure 5a), except for Test 2 (Figure 5b) where the lid remained attached and the container stayed upright after the drop.

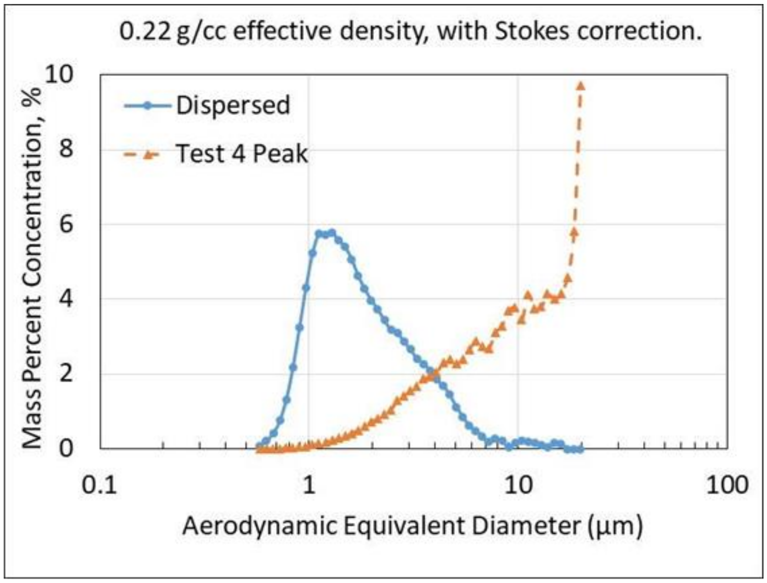

Figure 4. Particle size distribution of CeO_2 test powder: Mechanical dispersion by a Wright Dust Feeder, compared to the airborne aerosol from the drop Test 4 .

Table 1. Summary of drop test conditions and test results

	Drop Height, m	**Gross Mass, g**	**Test powder MAR, g**	**Spilled mass, g**	**RRF**	**SUM, g**	**SUF**
Test 1	4.78	9459.9	3.4	2.3	8.74E-03	4.64E-05	2.02E-05
Test 2[1]	4.78	9455.1	5.1	0.2	2.61E-03	2.07E-05	1.04E-04
Test 3	4.78	9459.2	4.1	2.8	1.86E-02	1.19E-04	4.25E-05
Test 4[2]	4.78	9310.9	4.6	1.3	2.88E-03	2.07E-05	1.59E-05
Test 5	4.78	9471.3	4.7	1.8	5.52E-03	4.04E-05	2.25E-05
Test 6	4.78	9472.1	5.5	2.7	9.18E-03	7.88E-05	2.92E-05

1. **Lid stayed on after drop (Figure 5b).**
2. **Drop tested without lid, CG over bottom corner orientation.**

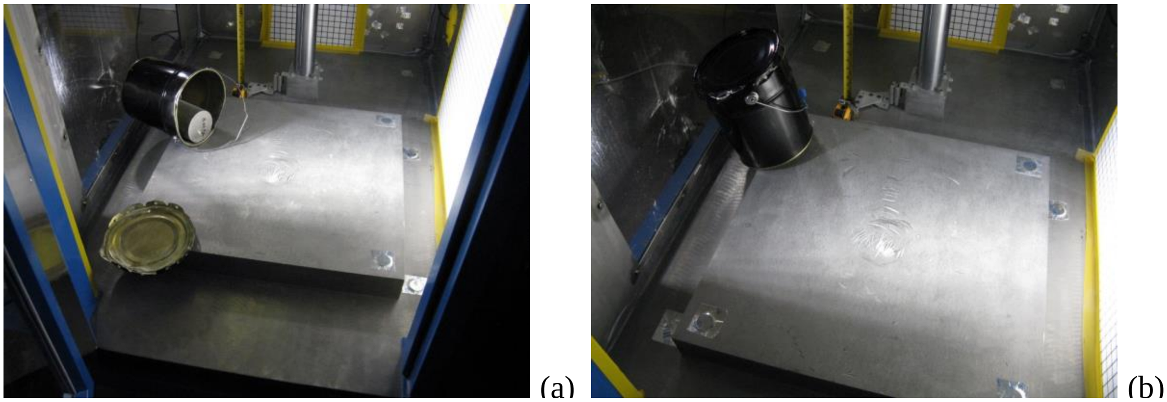
(a)
(b)

Figure 5. Positions of the container after dropping: (a) Test 1; (b) Test 2.

The test conditions of Test 1-3, and 5-6 were the same except for the minor difference in the loaded test powder mass. However, the spilled mass varied from 0.2 g to 2.8 g and the measured RRF varies from 2.61E-03 to 1.86E-02, indicating considerable variability of drop test data. This variability is observed not only in this study, but also in the data of other researchers[2] (Figure 6).

Comparison of LANL Test Results to Y12 Test Results

Gao et al[2] (Y12) studied airborne concentrations of particles resulting from drop tests of containers loaded with tungsten oxide (WO_3) (a surrogate for uranium oxide). This research was conducted at Syracuse University for the DOE Y-12 Site at Oak Ridge Tennessee. The experimental conditions for their tests differed from those done at LANL in the following ways: (1) test powder; (2) loaded powder mass; (3) container and payload; (4) drop height and orientation; (5) measurement system/configuration.

Figure 6 plots the measured SUMs in this study (LANL) with Y12 results. There is considerable variability in both LANL and Y12 measurements.

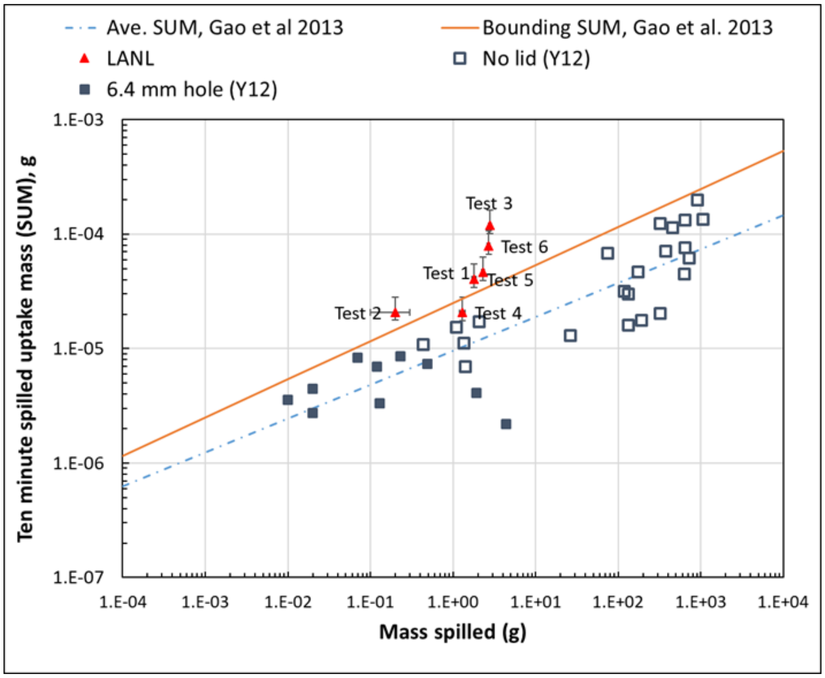

Figure 6. SUM comparison to Gao et al. 2013 (Y12).

Although there are many experimental differences, it is of interest to compare the LANL results with the Y12 tests that drop tested paint cans with a 6.4 mm hole in the lid of the can. (Their other tests were performed with no lids on the paint cans.) Figure 7a shows boxplots of the SUM measurements for LANL and Y12 (6.4 mm hole data). There are six observations for LANL and ten for Y12. The LANL SUM values have a median that is statistically significantly higher than the Y12 median and the LANL values have greater variability. These differences are likely due to the smaller particle size of the CeO_2 test powder, higher drop height, higher drop potential enegy.

Figure 7b contains boxplots comparing the SUF values for LANL and Y12. The LANL SUF measurements are within the distribution of the Y12 SUF measurements. The Y12 values have greater variability, but that might be expected, since the experimental conditions have more variability (e.g., different air speeds, different drop heights) resulting in greater variability of the spilled mass. Both data sets have one observation that looks different than the rest (statistical outliers). For the LANL data the outlier is from Test 2 where the lid stayed intact (unlike the other tests), resulting in low spilled mass, which resulted in a higher SUF than the other tests (SUF is SUM divided by the spilled mass).

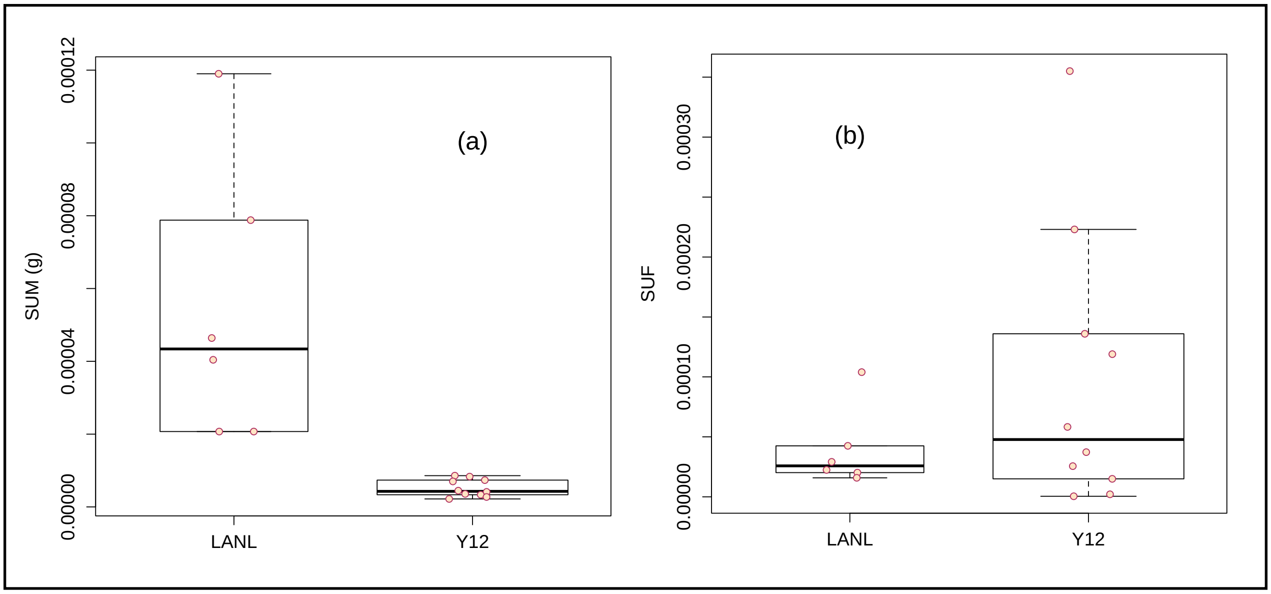

Figure 7. Boxplots comparing LANL to Y12 (6.4 mm hole) data for (a) SUM and (b) SUF.

DISCUSSION AND CONCLUSIONS

The RRFMC is operational and proving to be a valuable tool for evaluation of container performance. It will continue to be developed for this purpose. The RRFMC allows for an actual measurement of released powder during the dynamic phase of an impact event. By comparison, a helium leak test is performed under static, post-impact conditions. To verify compliance with DOE M441.1-1, multiple drops of the container must be performed, due to variations in test results.

In the near future, tests replicating the Y12 drop tests will be carried out in LANL RRFMC. These tests will use the same paint can , similar test powder (tungsten oxide), the same drop height and orientation, and a similar gross weight. These experiements will not only provide validation of the RRFMC measurements, they will also increase the understanding of the variability of uptake measurements and the implications of this variability for future studies and risk assessments. The effect of different MAR, with or without inner container, and different container configuration will be further investigated in RRFMC.

In addition applications other than container drop tests are being considerd for the RRFMC. One possibility would measure test powder release from a pressurized, static container. The RRFMC is also suitable for drop testing other items with or without aerosol release measurements.

ACKNOWLEDGEMENTS

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