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DOE NUCLEAR MATERIAL PACKAGING MANUAL: STORAGE CONTAINER REQUIREMENTS FOR PLUTONIUM OXIDE MATERIALS

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ABSTRACT
Loss of containment of nuclear material stored in containers such as food-pack cans, paint cans, or taped slip lid cans has generated concern about packaging requirements for interim storage of nuclear materials in working facilities such as the plutonium facility at Los Alamos National Laboratory (LANL). In response, DOE has recently issued DOE M 441.1 "Nuclear Material Packaging Manual" with encouragement from the Defense Nuclear Facilities Safety Board. A unique feature compared to transportation containers is the allowance of filters to vent flammable gases during storage. Defining commonly used concepts such as maximum allowable working pressure and He leak rate criteria become problematic when considering vented containers. Los Alamos has developed a set of container requirements that are in compliance with 441.1 based upon the activity of heat-source plutonium (90% Pu-238) oxide, which bounds the requirements for weapons-grade plutonium oxide. The pre and post drop-test He leak rates depend upon container size as well as the material contents. For containers that are routinely handled, ease of handling and weight are a major consideration. Relatively thin-walled containers with flat bottoms are desired yet they cannot be He leak tested at a differential pressure of one atmosphere due to the potential for plastic deformation of the flat bottom during testing. The He leak rates and He leak testing configuration for containers designed for plutonium bearing materials will be presented. The approach to meeting the other manual requirements such as corrosion and thermal degradation resistance will be addressed. The information presented can be used by other sites to evaluate if their conditions are bounded by LANL requirements when considering procurement of 441.1 compliant containers.

INTRODUCTION
The current inventory of LANL Special Nuclear Material Containers (SNMCs aka Hagan containers) were designed to provide a robust container for daily use, safe transport within the plutonium facility PF-4, and storage up to twenty years. The original design criteria are given below.[1]

Design Criteria of 1999

1. Set of four containers ranging in nominal size from one to twelve quart capacities.
2. Each container shall nest into the next larger size, and have a clearance of about two inches on the height and about ½ inch on the diameter.
3. Containers to be 304 stainless steel and have a body wall thickness on 0.020-0.032 inch to allow transmission of radiation for nondestructive assay.

4. Container body shall be seamless, have a flat bottom with rounded inner edges, and a 32 standard finish (easy to wipe clean to allow decontamination).

5. The lids shall be of 304 stainless steel and have a positive closure mechanism such as a clamp or screw.

6. The lid must contain a nuclear materials filter similar to the Nuclear Filter Technology Inc. model NUCFIL-030.

7. The assembled containers loaded to ½ volume with dry sand must withstand a nine-foot drop impacting on concrete (flat on bottom, inverted, side of body, 45° on lid). The container must not rupture or leak. Deformation of the container is expected. A test will be performed by pressurization through the filter hole to 2 psi with the container submerged in water. Container will pass if no bubbles are observed.

Figure 1. A complete set of Hagan Containers.

The DOE Manual for Packaging of Nuclear Material requires packages to have a Design Release Rate and Design Qualification Release Rate.[2] In order for existing SNMCs to be qualified as compliant with the DOE manual M441.1-1 and to establish criteria for procurement of new containers, criteria must be developed for the Design Release Rate and the Design Qualification Release Rate and testing conducted to demonstrate compliance. The testing must bound the conditions that the containers will experience while in use.

DESIGN RELEASE RATE

The Design Release Rate is $10^{-6}A_2$ at the maximum allowable working pressure (MAWP). The Design Release Rate depends upon the isotopic composition. The materials considered here are common plutonium containing materials. The nominal isotopic content is taken from DOE's 3013 Standard for Weapon Grade (WG), Fuel Grade, and Power Grade plutonium and from the LANL convention used in safety documentation for Heat Source (HS) plutonium (Pu-238 containing material). The isotopic compositions of the radionuclides are shown in Table 1.

Table 1. Isotopic composition for common plutonium containing material.

Nuclide	A_2 (Ci)	Specific Activity (Ci/g)	Pure ^{239}Pu	Weapon Grade	Fuel Grade	Power Grade	Heat Source
^{238}Pu	0.027	17	0.00%	0.05%	0.1%	1.0%	89.3%
^{239}Pu	0.027	0.062	100.00 %	93.50%	86.1%	63.0%	10.1%
^{240}Pu	0.027	0.23	0.00%	6.00%	12.0%	22.0%	0.6%
^{241}Pu	1.6	100	0.00%	0.40%	1.6%	12.0%	0.0%
^{242}Pu	0.027	0.0039	0.00%	0.05%	0.2%	3.0%	0.0%
^{241}Am	0.027	3.4	0.00%	0.00%	0.0%	0.0%	0.0%

The A_2 for a mixture is given by

$$A_2 = \frac{1}{\sum_i \frac{f(i)}{A_2(i)}} \qquad \text{Eq. 1}$$

Where f(i) is the activity fraction of radionuclide I that is available for release and $A_2(i)$ is the A_2 value for that isotope. Applying Eq. 1 to the material compositions in Table 1 yields the A_2 values for the mixtures, which are expressed in terms of Curies and grams of oxide. The material release rate of interest is 10^{-6} A_2 per hour expressed in grams per second. The air leak rate assumes a material density of the oxide in an air atmosphere of 1×10^{-7} g/cm^3 and is expressed in terms of the maximum container pressure.

Table 2. Mixture leak rate for the materials with isotopic composition given in Table 1. The material is assumed to be oxide.

	Pure ^{239}Pu	Weapon Grade	Fuel Grade	Power Grade	Heat Source
A_2 for mixture (Ci)	0.027	0.149	0.367	0.716	0.027
Total activity (Ci/g)	0.0547	0.4239	1.4989	10.8225	13.4014
A_2 for mixture (g)	0.4938	0.3518	0.2449	0.0662	0.0020
Material release rate (g/s)	1.37E-10	9.77E-11	6.80E-11	1.84E-11	5.60E-13
Air leak rate (cm^3/s)	1.37E-03	9.77E-04	6.80E-04	1.84E-04	5.60E-06

The MAWP for the SNMCs was not specified originally. In order to certify the containers as in compliance with the Design Release Rate, the leak rate must be determined at the MAWP. The existing SNMCs have been tested at a differential pressure of 1 atmosphere and the o-ring seals have failed. In addition, a structural analysis of the 8-qt container indicated that the yield strength of the container was approached at 1 atmosphere differential pressure and the report recommended that the containers not be subjected to that magnitude of differential pressure.[3, 4] Therefore a MAWP should be specified taking into account the fact that the containers are filtered.

There are two sources of pressure within a filtered container under ambient conditions and under normal operating conditions. The sources of pressure are gas generation and changes in atmospheric pressure. Gas generation is typically due to radiolysis of water although plastics also contribute. In this analysis, gas generation is calculated for radiolysis of water with a G-value equal to the maximum observed in the 94-1 Surveillance and Monitoring Program (300 nmol/s W) for material with a water content of 5wt% where the maximum Wattage of the container is a function of the volume at 4 W/L, which assumes 2 W/kg and that the bulk density of plutonium oxide powder is 2 g/ml. The volume of gas generated is given as:

$$\Delta V_g = \frac{G \cdot f_{H2O} \cdot SA \cdot R \cdot T}{P} \qquad \text{Eq. 2}$$

where G is the G-value, f_{H2O} is the fraction of water, SA is the specific activity in W, R is the gas constant, T is the temperature assumed to be 55°C, P is the local atmospheric pressure (78 kPa at Los Alamos), and ΔV_g is the gas generation rate in ml/s. The volume of gas associated with local atmospheric pressure changes is given by:

$$\Delta V_a = \frac{\Delta P}{P} \cdot V \qquad \text{Eq. 3}$$

where ΔP is the maximum change in atmospheric pressure, P is the local atmospheric pressure (78 kPa), V is the volume of the container, and ΔV_a is the volume of air that must be expelled in ml/s. The maximum change in atmospheric pressure can be estimated from local meteorological data that is available from the LANL Weather Machine. Records of the maximum and minimum pressure for every day over the period of Feb 1, 1990 to July 4, 2007 were used. The maximum one day pressure change was 2.01 kPa (the average one day pressure change was 0.46 kPa). In order to estimate the bounding case, it was assumed that the maximum pressure change in one day occurred over a 2 hour time. The pressure drop due to the flow of the gas is given by the filter flow capacity of 200 ml/min at 1" of water column assuming that the filter is 99% clogged.

$$\Delta P_g = \frac{\Delta V_g}{F \cdot 0.01} \qquad \Delta P_a = \frac{\Delta V_a}{F \cdot 0.01}$$

where ΔV_g and ΔV_a are the volume changes calculated from Eq. 2 and Eq. 3, F is the filter flow capacity, 0.01 is the fraction of the capacity assumed to be present, and ΔP_g and ΔP_a are the pressure drops across the container due to the gas generation and the changes in atmospheric pressure.

Table 3. The volume of LANL's SNMCs, the rate of gas generation, the pressure created by gas generation, the volume of gas due to atmospheric changes, the pressure created by atmospheric changes, and the total pressure change are given.

Vol of container	V_g (ml/s)	ΔP_g (kPa)	V_a (ml/s)	ΔP_a (kPa)	ΔP_{g+a} (kPa)
1 qt	0.0019	0.015	0.0034	0.025	0.04
3 qt	0.0059	0.044	0.01	0.076	0.121
5 qt	0.0099	0.074	0.017	0.127	0.201
8 qt	0.016	0.119	0.027	0.203	0.322
12 qt	0.024	0.178	0.041	0.305	0.483
5 gal	0.04	0.297	0.068	0.508	0.805
10 gal	0.079	0.593	0.136	1.017	1.61

The SNMCs show a maximum pressure generated by both mechanisms of less than 0.5 kPa and the drums show a maximum pressure generated by both mechanisms of 1.6 kPa. Therefore, in a conservative fashion, we assign a MAWP of 1 kPa to the SNMCs and a MAWP of 2 kPa to the drums.

The SNMCs are used for both Weapons Grade Pu (WG) and Heat Source Pu (HS). The appropriate testing conditions for the SNMCs will be 1 kPa differential pressure where the outside pressure is 0.01 kPa and the internal pressure is 1.01 kPa. The appropriate testing conditions for the drums will be a differential pressure of 2 kPa where the outside pressure is 2.01 kPa and the internal pressure is 0.01 kPa. The He leak rates allowed at 1 atm pressure need to be corrected to the units measured by the He leak detector, which is in atm $cm^3\ s^{-1}$, at the testing conditions. In addition, the pressure at which the containers are used needs to be considered. In order to be as general as possible, the pressure at which the containers are used is taken as 1 atmosphere. As the pressure at which the containers are used is lowered, the He leak rate criteria is reduced, therefore using a 1 atmosphere usage pressure is conservative with respect to the usage pressure at Los Alamos. The details of this calculation are given in Appendix 1. The recommended He leak rate criteria are given in Table 4.

Table 4. Design Release Rate criteria for helium leak checking containers. The first value is the He leak rate at the differential pressure across the container of 1 kPa and container pressure of 1 kPa and the second value is the He leak detector reading under the conditions of the first value.

	Weapons Grade Pu		Heat Source Pu	
	Air leak rate ($cm^3\ s^{-1}$)	He leak rate (atm $cm^3\ s^{-1}$)	Air leak rate ($cm^3\ s^{-1}$)	He leak rate (atm $cm^3\ s^{-1}$)
SNMCs	9.77E-4	6.81E-05	5.60E-06	1.29E-06
Drums	9.77E-4	8.46E-05	NA	NA

DESIGN QUALIFICATION RELEASE RATE

The Design Qualification Release Rate from the Manual can be measured either as a gas leak rate after drop or a material release amount after drop. Previous testing has shown that SNMCs release a puff of material during a drop test. When a container is dropped, the immediate action is to survey the container and take the container out-of-service. A test of material released is more appropriate under these conditions than a helium leak rate if the amount released is measurable. The allowed material release following a drop is 10^{-3} A_2. The amount of allowed release is 0.35 mg of oxide for weapons grade Pu and 2 μg of heat source Pu. This may be a measurable quantity under strictly controlled conditions. Experimentation is required to identify appropriate methods and materials to measure such a small amount and until demonstration of the technique in an industrial testing setting, it is suggested that to qualify the SNMCs for use with weapons grade and heat source Pu the gas leak check approach be used.

The required drop height is 1 x maximum height during operations. There are three heights that cover all operations. Four feet covers operations during transportation on carts in hallways, elevators, and laboratory rooms as well as handling required to place a container onto a cart or into hoods. Eight feet covers most vault operations and a few laboratory room storage locations. Twelve feet covers the worst case vault operations.

The payload should encompass the maximum allowed weight. In order to establish the maximum weight for testing, the MASS database was used to query the vault inventory for the packaged weight of all SNMCs. In some instances, one item had a significantly higher weight than the rest of the packages of that size. It was determined that the packages with significantly higher weights were sealed sources with lead pig shielding and out of scope of the Manual. Such items were not considered for this exercise as the nuclear material was intrinsically sealed and not available for leakage out of the container and as such not covered by the requirements of the manual. The largest packaged weight was then chosen for each size. The maximum package weight have been established, Table 5.

Table 5. Maximum package weights for SNMCs.

Package	Maximum package weight (kg)
1Q	10
3Q	15
5Q	18
8Q	20
12Q	22
5G	25
10G	40

The SNMCs must be tested with realistic total weight and under worst case orientations. The payload will be loaded into a slip lid can taped shut with a filtered lid to facilitate pump out during He leak testing. The slip lid can is placed into the SNMC (or Drum) and sealed as normal. The package is bubble leak tested both prior to and post testing. The Manual Design Qualification Release Rate criteria of 10^{-3} A_2 per 10 minutes is 6000 times greater than the Design Release Rate criteria. This translates to a gas leak rate of 29 cm^3/s for WG oxide and 0.22 cm^3/s for HS oxide for a pressure differential of 5 kPa. The appropriate gas leak test for this magnitude of leak rate is the "pressurized cavity bubble method". For these calculations a differential pressure of 5 kPa is chosen. The pressure is applied through the filter fixture. The leak rates are given in Table 6.

Table 6. Air leak rates at 5 kPa differential pressure for the Design Qualification bubble leak test.

Container class	WG Pu	HS Pu
SNMCs	29 $cm^3\ s^{-1}$	0.22 $cm^3\ s^{-1}$
Drums	19 $cm^3\ s^{-1}$	NA

RADIATION CRITERIA

The radiation dose to container construction materials is estimated for weapons grade plutonium oxide powder. Two cases are considered, one with an intact inner package and one with a failed inner package. The dose to steel and to plastic due to alphas is essentially the same. The dose rates per year are summarized in Table 1. The vendors are asked to estimate the lifetime of their design from the dose rates.

Table 1. Dose rates to container.

Case	Dose rate due to x-rays and gamma rays (rad/yr)	Dose rate due to alphas (rad/yr)	Total dose (rad/yr)
Intact inner package	6.6×10^4	0	6.6×10^4
Failed inner package	6.6×10^4	1.2×10^6 (in a ~ 45 micron layer)	1.2×10^6 (in a ~ 45 micron layer)

The radiation dose to the materials of construction comes from gamma and neutron irradiation regardless of whether the inner package is intact or not and from alpha radiation if the inner package has failed. The surface dose rates for gamma and neutron radiation have been derived for the case of a (gloved) hand in contact with plutonium material.[5, 6] The mass energy absorption coefficient for tissue and for stainless steel or viton will be different. However, it is the closest estimate I can find on short notice and will be used here. The neutron dose rate is given in terms of neutrons $s^{-1}\ gm^{-1}$ (where the grams refers to the mass of the nuclear material). I do not know how to convert this into Rads at this time. The definition of a rad is 1 joule of radiation energy deposited into 1 kilogram of material. The surface dose rates for the various radionuclides of interest are given in Table 2.

Table 2. Surface dose rates for gamma radiation and neutron emission for plutonium isotopes and americium.

Isotope	X,Gamma rays (rad/hr)	Oxide (neutrons/sec gm)
238-Pu	960	14000
239-Pu	0.98	45
240-Pu	4.7	170

241-Pu	.0090	---
242-Pu	.30	2.7
241-Am	0.19 t frac 241-Pu	---

IDENTIFICATION OF WORST CASE MATERIAL.
The worst case would be heat source Pu if large amounts of oxides were stored. However, the Hagan containers are authorized to store heat source Pu residues only. A more sophisticated analysis would be required to estimate the dose to the materials of construction from residues, therefore I will take as the worst case weapons plutonium with the isotopic ratios as Pu-238 0.01%, Pu-239 93.878%, Pu-240 6%, Pu-241 0.09%, and Pu-242 0.03%. Note that to get the surface dose rate as 50 grams of 238-Pu oxide a container would need to have 49 kg of 239-Pu.

The DOE Handbook on Design Considerations has the following information concerning materials of construction. "Most polymeric materials are suitable for cumulative radiation exposures of less than 1x104 rads in air. Most thermoplastics (with the exception of Teflon polytetrafluoroethylene (PTFE) and a few others) are suitable up to $1x10^6$ rads and may be usable up to $1x10^7$ rads."

Case 1: Inner package has not failed.

Dose due to alphas is zero. The dose due to X,Gamma rays is independent of the amount of material in the container and dependent only upon the isotopic fractions as long as the depth of the nuclear material in the container is greater than the penetration depth of the gamma rays. For kilogram amounts of material this should be true. The dose to the packaging due to X,Gamma rays is 7.5 Rad/hr or $6.6x10^4$ rads/yr. For 20 years in storage, this becomes 1.3×10^6 rads.

Case 2: Inner package has failed and inside of container is coated with a fine layer of plutonium oxide powder.

The alpha particle source is the plutonium oxide. The alpha range in plutonium oxide is about 15 microns. The energy deposited to a surface from alpha particle flux to the surface can be estimated assuming a 15 micron layer of plutonium oxide with a suitable void fraction to acknowledge that the coverage is not continuous. I will assume that 10% of the surface is covered, that 25% of the alphas emitted within that volume make it to the surface and carrying 50% of energy that they started with. This yields 0.37 joules per day impacting 1 cm^2 of surface. To convert this into rads, the volume of material that is affected must be known. For steel I assume the depth is 15 microns with a density of 8 and for plastic I assume 45 microns with a density of 2. For the steel the energy deposited is 125 rad/hr or 1.1×10^6 rad/yr and for plastic 138 rad/hr or 1.2×10^6 rad/yr. This dose is in addition to the x,gamma ray dose.

CONCLUSIONS
Containers used for packaging plutonium material at Los Alamos National Laboratory must have design criteria as specified by DOE's Nuclear Material Packaging Manual. The design criteria specified by the Manual were developed to protect workers from

excessive exposure to nuclear material under normal working conditions and during an accident condition specified as a drop of the container. The design criteria are a design leak rate and a design qualification leak rate. Because the containers are vented, the leak rates and testing conditions are not covered by an existing ASTM Standard. In this document, the approach to specifying the design leak rate and design qualification leak rate for vented containers is developed. Development of the design criteria required specification of (1) the material to be contained, (2) the maximum allowable working pressure, and (3) the maximum payload. The leak rates under testing conditions are determined using the allowed leak rates and specifying the testing conditions. The methodology of ANSI 14.5 is used. Requirements for resistance to radiation are also considered.

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