Expanded Content Envelope for the Model 9977 Packaging

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ABSTRACT

An Addendum was written to the Model 9977 Safety Analysis Report for Packaging adding a new content consisting of DOE-STD-3013 stabilized plutonium dioxide materials to the authorized Model 9977 contents. The new Plutonium Oxide Content (PuO_2) Envelope will support the Department of Energy shipment of materials between Los Alamos National Laboratory and Savannah River Site facilities. The new content extended the current content envelope boundaries for radioactive material mass and for decay heat load and required a revision to the 9977 Certificate of Compliance prior to shipment. The Addendum documented how the new contents/configurations do not compromise the safety basis presented in the 9977 SARP Revision 2. The changes from the certified package baseline and the changes to the package required to safely transport this material is discussed.

Background

Savannah River National Laboratory (SRNL) developed the Model 9977 Radioactive Material (RAM) Packaging for the Department of Energy (DOE) National Nuclear Security Administration (NNSA) as a replacement for the Specification 6M packaging for which the authorization was withdrawn by the U.S. Department of Transportation (DOT) in 2006. The 9977 RAM shipping package Safety Analysis Report for Packaging (SARP) (S-SARP-G-00001) Revision 2 [Ref 1] was authorized by the DOE Environmental Management (EM) Packaging Certification Program (PCP) Certificate of Compliance (CoC) Revision 0 [Ref 2] issued on October 9, 2007 with an expiration date of October 31, 2012. The SARP and CoC authorized the shipment of a single Content, 100 grams of plutonium and/or uranium “Heat Source” material in either of two (2) container configurations. The initial SARP established the design of the packaging, demonstrated its compliance with the requirements of Title 10 of the Code of Federal Regulations (CFR) Part 71 [Ref 3] (documenting is ability of meet the Normal Conditions of Transport (NCT) and the Hypothetical Accident Conditions (HAC) without loss of containment), its design pressures, temperatures, shielding and criticality features, its minimum essential operating procedures, its acceptance and maintenance requirements, and its Quality Assurance program.

The 9977 Packaging design was authorized with:

- a maximum of 100 pounds of contents (RAM and packing materials),
- a radioactive decay heat rate of 19 watts,
- a "leaktight" 6-inch inside diameter Containment Vessel (6CV), with
- a design pressure of 800 psi at 300°F.

The 9977 RAM contents and configurations authorized for shipment have been expanded, since it was initially certified, by the issue of Addenda which are reviewed by the DOE Certifying Official and authorized through a new revision of the CoC. An Addendum establishes the safety of a Content/Configuration that the new content/configuration is bounded by the established safety basis, that is, it is within the limiting weights, temperatures and pressures, and is within the dose rate limits and is sub-critical. Or the Addendum demonstrates that the new content/configuration is within the limits established by the package design. I rare instances the Addendum will justify the extension of the Package design to accommodate the new Content/Configuration. An Addendum also includes the calculations and analyses needed to establish the new safety basis. The standard Addendum format excludes those sections of the SARP chapters which are not changed by safety analysis for the new content/configuration. The Addendum includes SARP text, figures, and tables that are revised, added, or deleted in establishing the safety basis for the new content/configuration. A Letter Amendment Request is an even simpler document, typically with only one or two minor changes to the existing safety basis.

In the Summer of 2012 SRNL was directed to develop and certify a new Plutonium Oxide Content that significantly challenged the 9977 Package safety baseline.

Discussion

The scope of the initial 9977 Packaging SARP was a single RAM Content, 100 grams of "Heat Source" plutonium and uranium, in either of two configurations, as assemblies of Radioisotopic Thermoelectric Generators or in Food-Pack Cans. The approved contents have been expanded several times since the initial CoC. Four (4) Addenda have added five (5) "Metals", Advanced Gas Reactor Type one (AGR-1) fuel pellets, Los Alamos National Laboratory Offsite Source Recovery Program (ORSP) oxides, and "Training Sources" for the Pacific Northwest National Laboratory (PNNL).

SRNL was directed and funded to add a new Plutonium Oxide Content envelope (to which uranium oxide was later added) to the 9977 authorized Contents. This Dual 3013 Pu Oxide Content would both exceed all existing content envelope limits but also challenge the baseline package design. The National Nuclear Security Administration (NNSA) requested SRNL justify the shipment in the Model 9977 of two (2) DOE-STD-3013 Containers loaded with Pu and U to the maximum limits permitted by the Standard. The 3013 configuration consists of a 3013 Outer Storage Can, an 3013 Inner Storage Can, and optional material container(s) designed to meet the requirements of the DOE 3013 Standard. Individual package users typically develop site-specific 3013 Inner and Product containers. The 3013 Containers used to contain the Plutonium Oxide contents, as defined in Tables 1 and 2, meet the requirements for the reference Outer 3013 Can as well as the inner and product containers found in DOE STD 3013, Section 6.2, "Containers."

Table 1 – Plutonium Oxide Content Envelope (Per 3013 Container)

	Material [a, b, c]	**Pu Oxide "Maximum Impurities"**
Fissile Material (Maximum Weight %)	^{236}Pu	1×10^{-7}
	^{238}Pu	0.05
	^{239}Pu [d]	95
	^{240}Pu [e]	9
	^{241}Pu [d]	1
	^{242}Pu	0.1
	U [f]	50
	$^{241}Pu + ^{241}Am$	1
	^{237}Np	0.05
Impurities (grams)	Be	0.44
	Al	0.66
	Mg	2.2
	Na	1.32
	F	1.1
	B	2.2
	Li	2.2
Total Mass (kg)	Radioactive Materials	4.4
	Impurities	0.082
	All Contents	5.0

Table Notes.

a	All contents shall be dry.
b	Pu/U content bulk density shall be no greater than 7 g/cc and no less than 2.0 g/cc.
c	Contents shall be stabilized in accordance with DOE-STD-3013, Section 6.1.1.
d	Nuclide classified as "fissile" per DOE Good Practices Guide, Criticality Safety Good Practices Program, Guide For DOE Nonreactor Nuclear Facilities, DOE G 421.1-1, 3.79 Fissile Nuclide, 8-25-99.
e	^{240}Pu shall be greater than ^{241}Pu.
f	All isotopes except ^{232}U, which is limited to 1×10^{-7} weight percent

Table 2 - Dual 3013 Configuration Plutonium Oxide Content Limits

Characteristic	Per 3013	Total per 9977	Previous 9977 Limit
RAM Isotopes		Pu and U	Pu and U
RAM Form		Oxide	Pu/U as metal U only as oxide
RAM Mass	4.4 kg metal as 5.0 kg of oxide	4.8 kg metal as 10.0 kg of oxide	4.4 kg metal as 5.0 kg of oxide
Decay Heat Rate	19 watts	38 watts	19 watts
Moisture	0.5 wt%/25 grams	50 grams	“dry” contents
Total Content Weight		as required	100 pounds

<u>Flammable Gas Mixtures</u>

Previously, only metal form plutonium had been authorized as a content within the 9977. Plutonium oxide has unique attributes which affect its packaging and which needs be accounted for in its shipment. The DOE 3013 Standard defines the process by which plutonium metal is converted into its oxide form, the benefits gained, and the final form of the oxide. The 3013 Standard permits up to 0.5wt% moisture in the final oxide, which results in 25 grams of water in 5000 kg of Pu Oxide. Pu will radiolyze this moisture, producing hydrogen and oxygen gases. The liberated oxygen recombines onto the Pu, but the hydrogen is free to mix with the ambient atmosphere within the 3013 Container and (if the 3013 Container should leak) with the atmosphere within the package’s Containment Vessel. A gas mixture with the correct ratio of hydrogen and oxygen and an ignition source (a spark) will deflagrate (combust). With the right spacing within the package the deflagration can progress to a detonation. The U.S. Regulatory Guide 7.9 [Ref. 4] states that the Package safety basis “Should demonstrate that hydrogen and other flammable gases will not result in a flammable mixture within any confined volume of the package.”

Plutonium oxide had previously been authorized as a content within the SRNL Model 9975 Packaging. The Primary Containment Vessel (PCV) of the 9975 has a 5-inch inside diameter which is an inherently safe configuration for precluding a criticality. The Pu Oxide Content in the 9975 also had a restrictive packing configuration to prevent a Deflagration to Detonation (DDT) event and required inerting of the PCV to limit the atmospheric oxygen to less than 5% to preclude a flammable gas mixture. It was determined that the atmosphere with the 9977 6CV, including within the 3013 Containers, would also have to be inerted such that at the time of shipment the oxygen would be no greater than 5%. Since the oxygen liberated from radiolysis of the moisture reacts with and reabsorbs onto the plutonium oxide, the initial inerting of the 6CV and Contents assures the gas mixture remains non-flammable.

Shielding

Package shielding performance was evaluated for the Plutonium Oxide Content Envelope with the aluminum 3013 Spacer (added for criticality control as discussed below) and two 3013 containers each with 5 kg of PuO_2 (4.4 kg Pu). Some of the impurity isotopes listed in Table 1 were added after the shielding evaluation was completed and, therefore, were not explicitly modeled. However, the shielding evaluation conservatively assumed that beryllium would bound any other impurity isotope. Adding any of the other impurity isotopes to the beryllium would only reduce the alpha-n neutron source.

The 9977 Packaging design does not include any specific features intended to provide significant shielding. Photon (gamma radiation) shielding is primarily provided by drum geometry. Some gamma attenuation provided by the containment vessel stainless steel body weldment and closure. Neither the package geometry nor its materials of construction are specifically designed to provide neutron shielding. Neutron dose rate attenuation is provided primarily by the distance between the source and points external to the package, with minimal attenuation provided by the materials of the 6CV, Fiberfrax®, polyurethane foam, and the drum. The presence of material containers inside the 6CV does not significantly reduce the dose rate.

The requirements of 10 CFR 71 for radiation dose rate limits are met for NCT (maximum of 72.51 mrem/hr on contact at the package side) and HAC (maximum of 6.24 mrem/hr one (1) meter from the package bottom). The Transport Index for shielding to be placed on the package label is determined by measurement at the time of transport and must be less than 10 (for Non-Exclusive Use) as required by 10 CFR 71.47(a).

Sub-Criticality

The total fissile material (i.e., ^{239}Pu) mass in Plutonium Oxide Content Envelope is limited to 8.8 kg (with 4.4 kg in each 3013 container). It is known from analyses previous done for the SRNL 9975 and 9978 RAM Packagings that their 5-inch diameter containment vessels were sub-critical with larger fissile material masses. For the Plutonium Oxide Content Envelope in the Model 9977, a “3013 Spacer” configuration, illustrated in Figure 1, was proposed and analyzed. This Spacer would provide the volume and spacing of a 5-inch diameter Containment Vessel, as demonstrated in the Model 9978 Packaging SARP, rather than the 6CV approved for the 9977 design. The Nuclear Criticality Safety Evaluation (NCSE) analyzed these contents in the 3013 Spacer in the 6CV in the 9977. It should be noted that the uranium isotopes listed in Table 1 were a later addition to the original content envelope and were added after the NCSE was completed and, therefore, were not explicitly modeled. However, the NCSE conservatively assumed the content to be 100% ^{239}Pu and substituting uranium gram for gram for ^{239}Pu will only reduce k_{eff}.

The 9977 was initially authorized for the shipment of contents in RTGs and Food-Pack Cans. The Addendum added the Dual 3013 content configuration. These content containers are used to prevent the inadvertent contamination of the package by providing a level of confinement for the radioactive material contents and to provide protection of the content being shipped. These content containers are also referred to as product containers. The Dual 3013 Content configuration is shown in Figure 1. The analyzed Dual 3013 configuration includes the 3013 Spacer and the Heat Dissipation Sleeve. It is noted that aluminum foil or peanuts may be used as dunnage in place of either 3013 Container. The results of the NCSE are listed in Table 3.

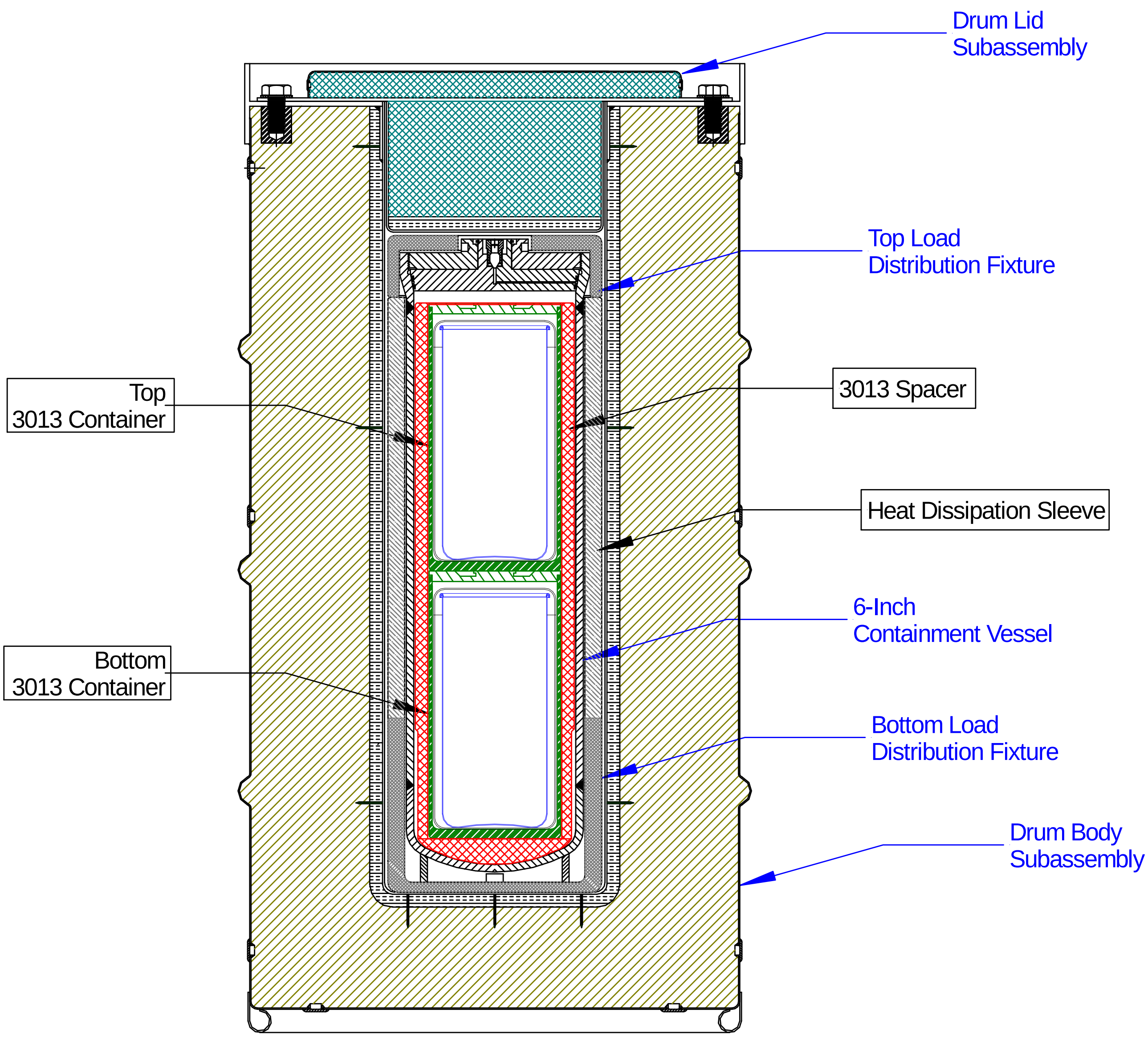

Figure 1 – The Heat Dissipation Sleeve in the Model 9977 Packaging

Table 3 - Summary of Criticality Safety Analysis Results (k_{safe}=0.945)

Content Envelope	**PuO_2**
Single Package Results	
Package calculated to be subcritical under conditions for maximum reactivity (for flooded conditions)	Max. k_{eff} = 0.8808
Most reactive configuration	^{239}Pu oxide (sphere), 11.46 g/cc, no 3013 container
Moderation for most reactive configuration	Flooded 6CV with 3013 Spacer
Reflection for most reactive configuration (package materials and/or 30 cm water)	30 cm water
Array Results	
NCT Array	Max. k_{eff} = 0.7199
Number of Packages	Analyzed: infinite
Water in drum voids	No
Most reactive fissile content	^{239}Pu oxide, 11.46 g/cc, in 3013 containers
Reflection surrounding array	Not applicable for infinite array
HAC Array	Max. k_{eff} = 0.9379
Number of Packages Analyzed	108 (6x6x3)
Most reactive fissile content:	^{239}Pu oxide, 3.28 g/cc, in flooded convenience can and inner 3013 containers
Moderation to credible extent	Dry
Reflection Surrounding Array	30 cm water

38 Watt Decay Heat Rate

The Viton GLT-S O-Rings used as part of the containment boundary in the 9977 6CV have a short term normal operating temperature limit of 400°F and a long term limit of 300°F. The challenge was to keep the O-Rings below this 300°F value while doubling the Content Decay Heat Rate to 38 watts. Ways in which to increase the heat conductance out of the 6CV and away from the O-Rings were investigated. It was decided to add an aluminum sleeve into the annular space between the outside the wall section of the 6CV liner of the Drum Overpack, essentially increasing the height of the Bottom Load Distribution Fixture until it met the Top Load Distribution Fixture. This Heat Dissipation Sleeve (HDS), illustrated in Figure 2, would have two benefits. Firstly, it would decrease the heat transfer resistance between the 6CV and the Overpack creating a more uniform heat transfer path out of the 6CV wall. This would more effectively dissipate the content heat load reducing the temperature of the O-Rings temperature and mitigate “hot spots” in the Overpack foam adjacent to the 6CV contents. The presence of the HDS kept the O-Ring temperature below 300°F. Secondly, the HDS provides mechanical support to the exterior of the 6CV, supporting it in the lateral HAC drop conditions. The structural calculations demonstrated that the presence of the DHS sleeve in the HAS Side Drops reduced the peak wall stresses and created an almost uniform stress distribution over the remainder of the 6CV walls. The down-side to the HDS is its weight. At 23 pounds the HDS consumes almost ¼th of the 9977s authorized Content weight, leaving 77 pounds for RAM, 3013s, and packing material inside the 6CV.

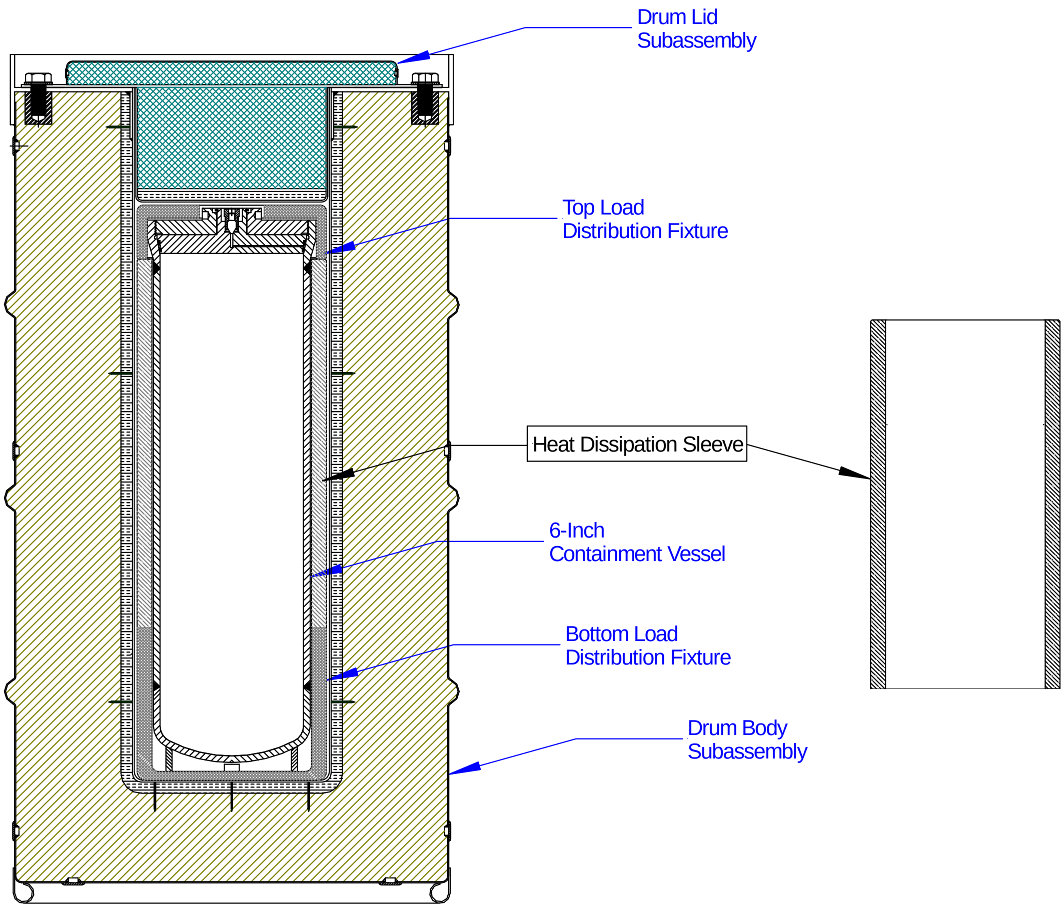

Figure 2 – The Heat Dissipation Sleeve in the Model 9977 Packaging

Maximum Normal Operating Pressure

The baseline 9977 SARP maximum pressure calculations are based on content configurations occupying no more than half of the volume inside 6CV. The internal volume of a 6CV is 608 in^3. The 9977 baseline Maximum Normal Operating Pressure (MNOP) was 41.2 psig, which is a 95% margin against the 6CV design pressure. The MNOP calculations are revised to evaluate the Plutonium Oxide Contents to ensure that the Dual 3013 content geometry configuration and their thermal characteristics are correctly evaluated.

The free volume calculations assume that the 3013 containers leak and the gases escape into the 6CV. The free volume of the 6CV is calculated by subtracting the volume of content configuration and the porosity of the Pu Oxide. From their respective drawings and an assumed 50% PuO_2 crystal density, the internal free volume of a 6CV in the Dual 3013 configuration is 246 in^3. Pressure rise within the 6CV was calculated due to heating of the gases container within the 6CV and the 3013 Containers, the off-gassing from plastics, the complete radiolysis of 50 grams of water, and the generation of helium.

Table 4 - Maximum Pressures for 6CV under NCT and HAC

Condition	**Max Pressure** (psig)	(Margin)
NCT	422	47%
HAC	485	39%
Design Limit	800	

Note: Margin is defined as [1-(actual/allowable)]×100%.

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References

1 *Safety Analysis Report for Packaging Model 9977 Type B(M)F-96*, S-SARP-G-00001, Revision 2, Savannah River National Laboratory, Savannah River Site (August 2007).
2 *DOE Certificate of Compliance No. 9977 for the 9977*, 9977, Revision 0, U.S. Department of Energy (October 2007).
3 *Packaging and Transportation of Radioactive Material*, Code of Federal Regulations, Title 10, Part 71, Washington, DC (December 2012).
4 U.S. Nuclear Regulatory Commission Regulatory Guide, *Standard Format And Content Of Part 71 Applications For Approval Of Packages For Radioactive Material*, Office Of Nuclear Regulatory Research, Revision 2, March 2005