SRNS-STI-2016-00086

Proceedings of the ASME 2016 Pressure Vessels & Piping Division Conference
PVP2016
July 17-21, 2016, Vancouver, BC, Canada

PVP2016-63023

THERMAL GRADIENT WITHIN DOE 3013 CONTAINERS DURING STORAGE

Steve J. Hensel
Savannah River Nuclear Solutions
Savannah River Site, Aiken, SC 29808
Email: steve.hensel@srs.gov

Si Lee
Savannah River Nuclear Solutions
Savannah River Site, Aiken, SC 29808
Email: si.lee@srnl.doe.gov

ABSTRACT

The Department of Energy (DOE) Model 9975 Package is used to store plutonium bearing materials at the Savannah River Site (SRS). The plutonium is packaged in a 3013 container which consists of a convenience container, an inner welded container, and an outer welded container [1]. All containers are made of stainless steel. A portion of the population of 3013 containers have plutonium dioxide with alkeline earth chloride salts. Temperatures and temperature gradients within the 3013 container are useful to evaluate the transport of water vapor and potential corrosion in the inner container, particularly near the weld region. Thermal analyses of plutonium oxide packaged in 3013 containers which are stored in 9975 shipping packages were performed with emphasis on the temperatures and temperature gradients between the plutonium oxide and the inner container weld region. Thermal analyses investigated various plutonium oxide fill levels, heat generation, plutonium oxide thermal conductivity and 9975 package boundary conditions.

Not surprisingly, the results suggest the source heat is mostly dissipated radially outward as opposed to the vertical direction, and the 9975 package is insensitive to an adiabatic top and bottom surface boundary condition versus an adiabatic bottom surface assumption. The temperatures at locations of interest, such as the inner container weld region and the plutonium oxide centerline temperature may be correlated as a linear function of thermal loading and ambient temperature.

INTRODUCTION

This document describes the computational models for the thermal analyses of the storage of plutonium oxide in the Model 9975 package where the plutonium oxide is packaged inside in the 3013 container. Previous thermal analyses focused on calculating temperatures at locations of interest and their variability with different plutonium materials [2]. This analysis is focused on temperatures and temperature gradients between locations of interest in the 3013 container. The analysis considers a combination of plutonium oxide fill levels, ambient temperature and thermal loadings to evaluate the temperature sensitivity. The heat generation results in elevated temperatures within the 3013 container and a temperature gradient between the plutonium oxide and the Inner Container Closure Weld Region (ICCWR); depicted in Figure 1. The temperatures are of interest due to the potential for water vapor and chloride chemical reactions inside the inner 3013 container. All computational analyses and modeling geometry creations were performed by commercial software, ANSYS Fluent and GAMBIT.

The 9975 package consists of a 35-gallon drum with a removable lid, both constructed from 304L staineless steel sheet. Celotex, a fiberboard insulation, shields the Primary containment vessel (PCV), Secondary Containment Vessel (SCV), and 3013 container from external thermal loads such as fire. A lead shielding is located between the SCV and Celtotex insulation providing a means of gamma radiation shielding. The PCV and SCV provide a double containment of the 3013 container which contains the RAM-filled convenience can. The 3013 container is shown in Figure 1. A 6061-T6 aluminum spacer is placed between the PCV and the top of the 3013 container to provide impact damage protection and to augment the heat transfer from the heat source; plutonium oxide powder. Inside the 3013 container is the RAM-filled convenience can. Aluminum honeycomb placed between the bottom of the PCV and SCV as well as the PCV and 3013 container provides impact damage protection along with enhanced heat dissipation.

MODELING APPROACH

The geometry was created and meshed using the ANSYS GAMBIT software. The surface of the modeling domain is decomposed into component surfaces that allow greater mesh accuracy and efficiency. In regions that are expected to experience larger temperature gradients and/or increased molecular interaction complexity, the component surface is assigned a greater mesh density to increase solution accuracy. The gas-filled regions of the container are examlples of component surfaces with greater mesh density, as well as regions on and near conducting/convecting/radiating surface boundaries. Conversly, the mesh desensity is smaller on component surfaces that are expected to experience smaller temperature gradients; such as the thick celotex insulation portion of the 9975 package. This approach to modeling/meshing the 9975 package decreases the time of computation and increases the solution accuracy. The resulting number of computational mesh nodes over the modeling domain is approximately 27,000 nodes. Figure 2 illustrates the computational mesh over the modeling domain containing the geometry of the 3013 container.

Governing Equation

The steady-state equation governing the conduction-radiation heat transfer problem of the 9975 package under the axi-symmetric cylindrical coordinate system is shown below.

$$\rho C_p \frac{\partial T}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left(r q_{cond}\right) + \frac{\partial q_{cond}}{\partial z} + \nabla \cdot \bar{q}_{rad} - q''' = 0$$

Total enthalpy term T is local temperature in the computational domain. q_{cond} is the conductive heat flux term calculated by the product of spatial temperature gradient and thermal conductivity k. The radiation heat flux term $\bar{q}_{rad}$ was calculated by the Discrete Ordinate (DO) method. Decay heat source term* q''' is provided to the energy equation as a model input.

Boundary Conditions

Initially, the top and bottom surfaces of the 9975 package are assumed to be adiabatic. To analyze the solution's sensitivity to the initial assumption, the solution is also obtained assuming only the bottom surface to be adiabatic. For the calculations, the package is in an upright position and is always stored in the shaded facility. The initial assumption considers the containers being stacked in storage, whereas the adiabatic bottom surface assumption considers an isolated container resting on a concrete floor. The floor-container interface is assumed to be adiabatic due to concretes low thermal conductivity and due to relatively small temperature gradients at the bottom surface of the 9975 package. Since the analyses considers only the case of 9975 package storage, and not transport, the package is not exposed to direct solar radiation. Therefore, solar radiation on the surface of the package can be assumed negligible. Because the 9975 package is a cylindrical container, all energy transfer within the package is symmetric about the central axis of the container, axisymmetric. This allows for an analysis of only one half of the container resulting in reduced time of calculation. Cooling mechanisms for the 9975 package are conduction/radiation within the package and natural convection to the ambient for the outer surface of the package.

Prandtl number Pr in Table 1 is ratio of viscous momentum diffusion ν to thermal diffusion α. Rayleigh number, Ra, is defined as ratio of natural convection to viscous diffusion in terms of temperature drop of concrete wall surface ΔT and length scale L, that is,

$$Ra = \left(\frac{\Delta T g \beta L^3}{\nu \alpha}\right)$$

The parameter, β, is thermal expansion coefficient of air, and it is typically inverse of absolute temperature following the air to be ideal gas, and g is the gravitational acceleration.

Material Properties

Material properties for the thermal analyses of the 9975 package are listed in Table 2 and the emissivity associated with each component of the package are listed in Table 3**.** The cavity between each containment vessel is filled with carbon dioxide and the 3013 container is filled with helium for the nominal case thermal calculations. Based on previous work on plutonium oxide thermal conductivity [3] and discussuions with the principal author of this work, a simplified constant thermal conductivity of plutonium oxide with values of 0.1 and 0.21 W/m-K were chosen.

COMPUTATIONAL ANALYSES

The thermal limit for the 3013 and 9975 package is 19W. The majority of 3013 containers are packaged with 5 – 12 Watts depending on plutonium isotopics and density of the plutonium oxide. To gauge the solution's sensitivity to changes in thermal load, ambient temperature, and adiabatic assumptions, several parametric analyses were performed. These analyses include; a bounding 19 Watt thermal loading with assumed adiabatic top and bottom surfaces, 19 Watt thermal loading with an assumed adiabatic bottom surface, 15 Watt thermal loading with assumed adiabatic top and bottom surfaces, 10 Watt thermal loading with assumed adiabatic top and bottom surfaces, and a 5 Watt thermal loading with assumed adiabatic top and bottom surfaces. Ambient temperatures of 37.78°C (100°F), 32.22°C (90°F) and 26.67°C (80°F) were investigated where the ambient temperature is the temperature of the atmosphere surrounding the 9975 package. Lastly, a parametric evaluation

of plutonium oxide fill level was investigated. The fill-level refers to the physical level of plutonium oxide powder present in the 3013 convenience can.
Figures 3 and 4 provide temperature contours for 19 Watts in a full filled and half-filled 3013 container. Figures 5 and 6 show the linear relationship between heat loading and temperatures of interest. This relationship holds for the plutonium oxide and the inner container weld temperature, at least up to 19 Watts which is the maximum heat loading permitted in a 3013 container. Note that the plutonium oxide thermal conductivity in these analyses was 0.23 W/m-K.

From the calculation results, temperature for each of key components such as the inner 3013 weld region temperature, $T_{weld,}$ head space temperature, T_{HS}, and plutonium oxide region, T_{maxPu} and T_{avePu}, may be simply calculated as function of thermal loading Q in a linear form at ambient temperature T_{amb}.

$$T_i = A_i Q + T_{amb}$$

where i is the location of interest such as inner 3013 weld region, head gas space, and plutonium oxide. The constants A_i are unique for plutonium oxide fill level and plutonium oxide thermal conductivity.

The primary results of interest are presented in Table 4 where the plutonium oxide thermal conductivity of 0.1 and 0.21 W/m-K were utilized. Cases of fully filled, three-quarter filled and half-filled 3013 containers were evaluated at 19 Watts. Not surprisingly, the inner 3013 weld and head space temperatures are relatively independent of plutonium oxide thermal conductivity.

The factors A_i as a function of fill height and thermal conductivity are provided in Table 5. These correlations provide a fast and simple manner to calculate the temperature t locations of interest for a specified 3013 container heat loading and ambient storage temperature.

The application of the factors in Table 5 to 3013 inner container moisture conditions will be the subject of a future paper. In short, head space relative humidity data is obtained during 3013 destructive examination. During destructive examination the outer and inner 3013 containers are puncture and cut open. The lid on the convenience container is replaced with one with thermocouples and a humidity probe. Humidity measurement of the head space is obtained and represents the water vapor partial pressure during storage. Assuming constant water vapor pressure within the inner 3013 container, the relative humidity at the inner container weld region and the relative humidity in the plutonium oxide may be determined using the temperature correlation. The humidity conditions are considered important to determining potential corrosion mechanisms which may degrade the inner container weld region [4, 5].

CONCLUSIONS

Thermal analyses of plutonium oxide packaged in 3013 containers which are stored in 9975 shipping packages were performed with emphasis on the temperatures and temperature gradients between the plutonium oxide and the inner container weld region. Thermal analyses investigated various plutonium oxide fill levels, heat generation, plutonium oxide thermal conductivity and 9975 package boundary conditions. Not surprisingly, the results suggest the source heat is mostly dissipated radially outward as opposed to the vertical direction, and the 9975 package is insensitive to an adiabatic top and bottom surface boundary condition versus an adiabatic bottom surface assumption. The temperatures at locations of interest, such as the inner container weld region and the plutonium oxide centerline temperature may be correlated as a linear function of thermal loading and ambient temperature. The application of the linear correlation to 3013 inner container moisture conditions will be the subject of a future paper.

REFERENCES

1. DOE-STD-3013-2012, Stabilization, packaging, and Storage of Plutonium-Bearing Materials, March 2012.

2. Gupta, N. K., "Thermal Analysis of 3013/9975 Configuration", Journal of Nuclear Materials Management, Winter 2010, Vol. 38, No. 2.

3. Veirs, D. K., and Prenger, F. C., "Thermal Conductivity of high-Purity and Impure Plutonium Oxide Materials", Journal of Nuclear Materials Management, Winter 2010, Vol. 38, No. 2.

4. Berg, J. M., et. al., "Thermal Gradients and the Potential to Form Liquids in 3013 Containers", Journal of Nuclear Materials Management, Spring 2010, Vol. 38, No. 3.

5. Zapp, P. E., et. al., "Relative Humidity and the Susceptibility of Austenitic Stainless Steel to Stress –Corrosion Cracking in an Impure Plutonium Oxide Environment", Journal of Nuclear Materials Management, Spring 2010, Vol. 38, No. 3.

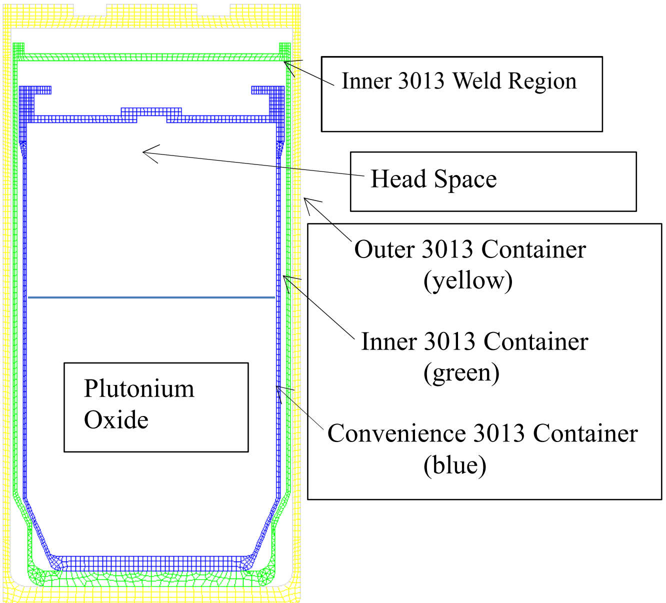

Fig. 1: 3013 Container

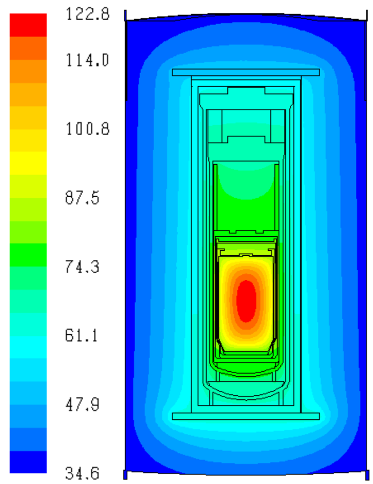

Fig. 3: 19W, full-filled 90°F (32.2°C) ambient

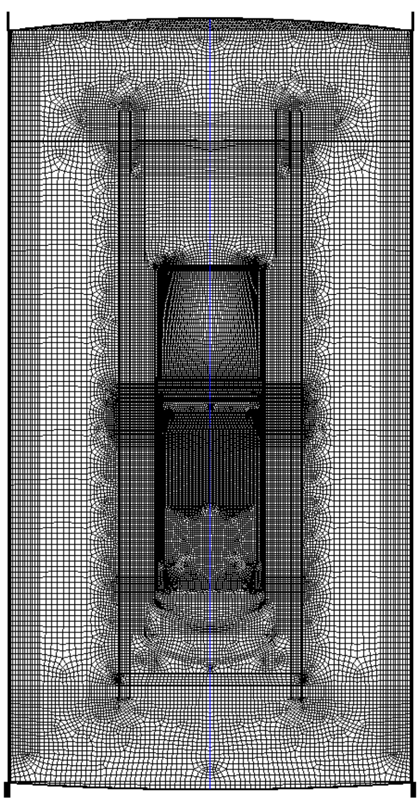

Fig. 2: Computational Doman
27,000 Elements

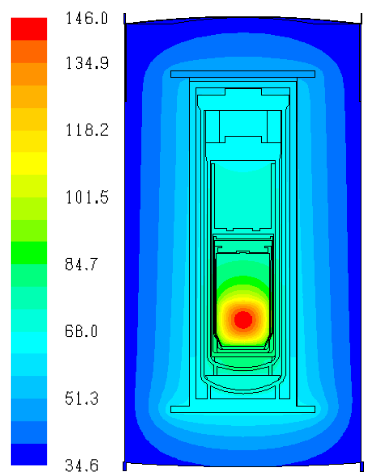

Fig. 4: 19W, half-filled 90°F (32.2°C) ambient

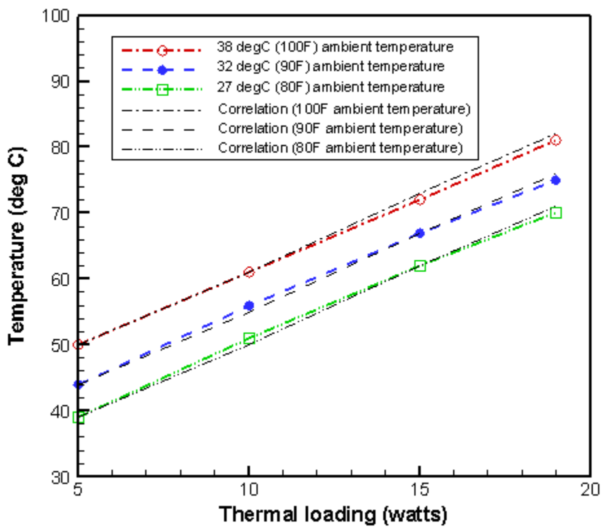

Fig. 5. Inner 3013 weld temperatures as function of heat generation for half-filled package

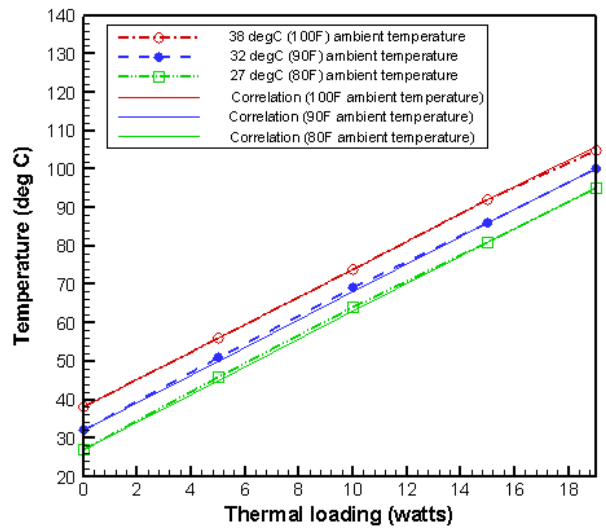

Fig. 6. Average plutonium oxide temperature as function of heat generation for full-filled package.

Table 1. Natural convective heat transfer coefficient used for the calculations

Geomtery orientation	Wall surface boundary	Natural convective heat transfer coefficient
Vertical	Isothermal plate with $Ra < 1.0\times10^9$	$h = \left(\frac{k}{L}\right)\left(0.68 + \frac{0.670\times Ra^{0.25}}{\left[1.0+\left(\frac{0.492}{Pr}\right)^{9/16}\right]^{4/9}}\right)$
Vertical	Isothermal plate with $1.0\times10^9 < Ra$	$h = \left(\frac{k}{L}\right)\left(0.825 + \frac{0.387\times Ra^{1/6}}{\left[1.0+\left(\frac{0.492}{Pr}\right)^{9/16}\right]^{8/27}}\right)^2$
Horizontal	Hot isothermal plate facing upward with $2.6\times10^4 Ra < 1.0\times10^7$	$h = \left(\frac{k}{L}\right)0.54\times Ra^{0.25}$
Horizontal	Hot isothermal plate facing upward with $1.0\times10^7 < Ra < 3.0\times10^{10}$	$h = \left(\frac{k}{L}\right)0.15\times Ra^{1/3}$

Table 2: Thermal Conductivity

Solid/Gas	Thermal Conductivity (°C)	(W/m-K)
304 Stainless Steel	0 100 500	13.40 16.32 21.78
1100 Aluminum	All	218.1
5052 Aluminum Honeycomb	All	6.612 (radial) 13.19 (axial)
Aluminum Spacer (6061-T6)	21.11 37.78 93.33 148.9	166.3 167.7 171.4 174.1
Kaowool Blanket	37.78 260	0.03479 0.06786
Lead	98.33 204.4	33.68 31.68
Plutonium Oxide	All	0.1 and 0.21
Air	0 100 200	0.0242 0.0318 0.0387
Carbion Dioxide (CO_2)	26.67 76.67 126.7	0.0166 0.0205 0.0246
Helium (He)	26.85 76.85 126.9 176.9	0.145 0.152 0.170 0.187
Celotex (radial)	All	.125
Celotex (axial)	-8.33 86.11 146.11	.0537 .0589 .0623

Table 3: Component Surface Emissivity

Surface	Emissivity
PuO_2	0.90
PCV/SCV/3013 Outer Can & Inner Can	0.30
Air shield/3013 Convenience Can	0.20
Drum Inner Surface	0.30
Drum Outer Surface	0.21
Aluminum Bearing Plates/Shield Lid/Spacer	0.20
Aluminum Honeycomb*	0.20
Lead Shielding	0.30
Celotex Fiber Board	0.50

Table 4: Temperature of the 3013 Locations of Interest

Cases	$K_{pu\ oxide}$ W/m-K	T_{weld} °C	T_{maxpu} °C	T_{HS} °C	T_{avePu} °C
Half fill	0.21	81	157	90	119
	0.10	81	223	91	145
Three-quarter fill	0.21	83	144	91	113
	0.10	83	197	92	134
Full fill	0.21	85	132	N/A	107
	0.10	85	175	N/A	124

Table 5: Correlation Coefficients of 3013 Locations of Interest

Cases	$K_{pu\ oxide}$ W/m-K	A_{weld} W/°C	A_{maxpu} W/°C	A_{HS} W/°C	A_{avePu} W/°C
Half fill	0.21	2.26	6.26	2.74	4.26
	0.10	2.26	9.74	2.79	5.63
Three-quarter fill	0.21	2.37	5.58	2.79	3.95
	0.10	2.37	8.37	2.84	5.05
Full fill	0.21	2.47	4.95	N/A	3.63
	0.10	2.47	7.47	N/A	4.53