**Contract No:**

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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SRNS-STI-2017-00098 Rev. 1

**Proceedings of the ASME 2017 Pressure Vessels & Piping Division Conference**
**PVP2017**
**July 16-20, 2017, Waikoloa, Hawaii**

**PVP2017-65054**

# LIFE EXTENSION OF THE 9975 PACKAGE AS A STORAGE CONTAINER: THERMAL ANALYSIS

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## ABSTRACT

The 9975 shipping package is used to store plutonium bearing material with a heat release of up to 19 Watts at the Department of Energy (DOE) Savannah River Site (SRS). Individual 9975 packages have been used to store these materials for nearly 15 years. The 9975 package contains non-metallic components such as the elastomeric dual O-ring seals, used to ensure containment at the vessel closures, and a fiberboard over pack which provides impact and fire resistance to the containment vessels. These non-metallic components degrade during long term storage, particularly when higher heat generating contents are packaged. Degraded fiberboard properties result in higher peak internal 9975 package material temperatures during a fire accident event.

The thermal performance of the 9975 shipping package was evaluated for a sequential accident consisting of a fire and drop which locally ruptures the outer drum. The package is exposed to an off-normal 58.3°C (137°F) ambient temperature prior to being fully engulfed in a fire for 1.5 hours at 815.6°C (1500°F). Subsequently the fiberboard smolders for 1.0 hour at 760°C (1400°F) at the location of drum rupture, followed by cool down to the ambient temperature. The thermal evaluation considered both the beginning-of-life (as-designed) condition and after 20 years of service as a plutonium material storage container. The results of the evaluation demonstrate that the 9975 shipping package maintains containment during initial and after 20 years of service. The maximum Primary Containment Vessel dual O-ring temperatures during the facility fire-drop-smoldering accident are 163.9°C (327°F) and 186.1°C (367°F) for beginning of life and after 20 years of service, respectively, which are within the allowable accident temperature limit of 204.4°C (400°F). Thus, the 9975 shipping package meets its intended function to provide containment.

## INTRODUCTION

The 9975 shipping package certified as a Type B package for up to 19 Watts of plutonium (oxide or metal) contents. As such, the package was not specifically designed for long term material storage. A surveillance program was initiated to evaluate and ensure the safety of the 9975 shipping package as a long term storage container at SRS. The package has been used to store plutonium bearing materials for up to 15 years, and an evaluation for up to 20 years storage is presented.

As illustrated in Figure 1, the 9975 Model is composed of a 35 gallon (132.5 liters) stainless steel drum with a bolted flange closure. The over pack material is Celotex® fiberboard made from sugarcane bagasse or softwood in accordance with American Society for Testing and Materials Specification C208 [1]. The fiberboard surrounds a nominal ½” (12.7 mm) thick lead shielding cylinder. Inside the shielding are two-nested primary and secondary stainless steel containment vessels (PCV and SCV) with two Viton GLT (newer packages utilize Viton GLT-S) O-ring seals designed in accordance with Section III, Subsection NB of the American Society of Mechanical Engineers (ASME) Code [2,3].

Other design features of the package include an air shield and a thermal blanket. The air shield surrounds and shrouds the uppermost portion of the top Celotex subassembly. It has two thermal features. First it minimizes the potential for Celotex combustion in the fire event by preventing direct flame impingement, and second, it eliminates the post fire smoldering of Celotex that could possibly occur from a horizontal drum orientation (chimney effect). The chimney effect could be produced by air passing through opposite vent holes or openings in the drum closure. Each 9975 shipping package has an inner thermal blanket that sits atop the air shield primarily to remove axial clearance resulting from the stack-up of nominal Celotex sheet thicknesses (12.7 mm / 0.5 inch), but it also reduces the thermal flux through the top of the drum.

The 9975 shipping packages are stored using metal pallets containing four drums each stacked up to three pallets high. The fire accident scenario consists of a 815.6°C (1500°F) fire for 1.5 hours duration that presumably weakens the pallets and causes collapse of a stacked array resulting in a postulated free drop of the drum. The drum is conservatively assumed to breach due to the drop and expose the fiberboard inside to the ambient air which causes the fiberboard to smolder. The potential breach is assumed to occur on the drum side, bottom or top depending on orientation of the drum during the pallet collapse.

There have been several papers published which specifically address the 9975 shipping package and its response to fire accidents [4, 5, 6]. Reference 6 addresses the drop and breach scenario discussed here. This work differs in that it considers degradation of fiberboard due to aging from long term radioactive material storage and the influence of this degradation on the temperature response of the 9975 package. Degraded fiberboard material properties are provided as inputs, based on work internal to SRS, and the focus of this paper is on the fire analysis and resulting temperatures. The criteria for maintaining containment is peak O-ring temperature and comparison with an O-ring temperature limit.

## LONG TERM STORAGE AND AGING

A surveillance program was developed to evaluate the condition and degradation of the 9975 package with examination of field specimens. Table 1 provides summary of the results from the field surveillance. Additionally, degradation of the fiberboard and O-rings are studied via laboratory aging studies.

Table 1: 9975 Shipping Package Field Surveillance

| Shipping Package Components | As-found Degradation |
|---|---|
| Drum | Corrosion[1] |
| PCV/SCV O-rings | Decreased elasticity[2] |
| Fiberboard | Mold (bottom fiberboard) |
| | Axial air-gap > 25.4 mm (1 in.)[3] |
| | Elevated Moisture Levels |
| Lead Shielding | Outer Surface Lead Corrosion[4] |

1. Small amount of localized corrosion identified on the drum bottom and on the drum lid closure bolts.
2. O-rings performed acceptably with some degradation.
3. The highest axial air-gap measured to date is 44.2 mm (1.74 inches).
4. Lead carbonate is formed at a rate < 0.05 mm (2 mils) per year on those packages without aluminum clad lead shielding

Laboratory studies have shown that fiberboard and O-ring degradation rates are highly temperature and humidity dependent. Although the 9975 packages stored at SRS are exposed to varying air temperature (e.g. cooler during winter and warmer during summer) the storage conditions are much less severe than those defined in 10 CFR 71 for Normal Conditions of Transport (NCT). In general, degradation appears to be minimal for packages with low heat generation; however, fiberboard degradation is noticeable and measurable in packages with heat generation approaching 19 Watts. The surveillance program has shown that the fiberboard density and thermal conductivity decrease with age. Additionally, moisture within the fiberboard tends to migrate to the drum bottom for higher heat packages causing the fiberboard to compress resulting in a gap at the package top between the drum top and upper fiberboard.

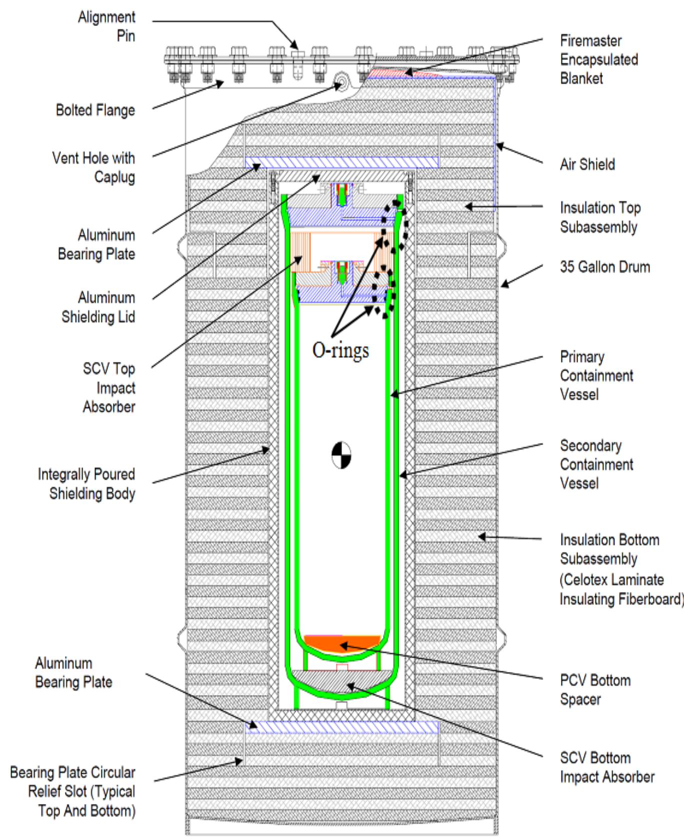


Figure 1: 9975 Shipping Package Two-Dimensional view

## THERMAL LIMITS

The 9975 shipping package design ensures that package containment components (especially the limiting package components such as the dual O-rings) operate below their thermal design limits in order to maintain containment. The design temperature accident limit for the dual O-rings is chosen as the acceptance criterion. This value of 204.4°C (400°F) is documented in the 9975 Safety Analysis Report for Packaging

(SARP). This value is provided by the manufacturer and is generally based on 1,000 hours exposure at this temperature. Thus the SARP very conservatively applies this criterion to short-term exposure due to the fire transient. Actual failure temperature for the O-rings is expected to be considerably higher (perhaps 300°C or more). Therefore, for the purpose of this work the 204.4°C (400°F) criterion is appropriately applied to aged O-rings.

## MODELING APPROACH

An axisymmetric finite element model of a single 9975 package is created using COMSOL Multiphysics Computer Code to evaluate the fire accident scenario. A 12 inch drum breach vertically along the side, at mid-height, is assumed to occur immediately after the fire accident. The breach is conservatively assumed to circumferentially encompass the entire drum. The fire is an instantaneous 815.6°C (1500°F) temperature that fully engulfs the package. The axisymmetric finite element model consists of approximately 120,000 nodes. The model uses non-homogenous mesh with various element sizes. The maximum and minimum mesh sizes are 1.5in and 0.048in, respectively. The meshes are refined around the limiting regions. Mesh sensitivities are also performed which showed no difference to the final results of interest. Figure 2 presents the mesh of the axisymmetric finite element model.

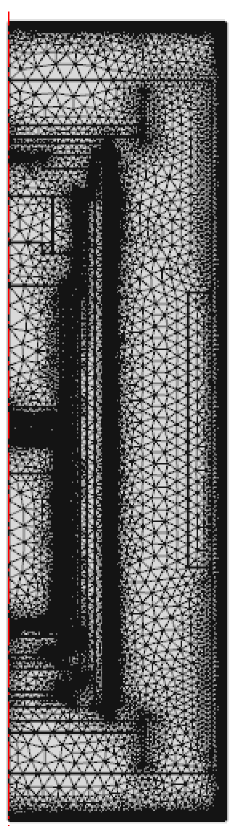

Figure 2: Mesh topology of the axisymmetric finite element model.

### Governing Equation

The steady-state equation governing the conduction-radiation heat transfer problem of the 9975 package under the axisymmetric 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

There are three inputs needed to describe the boundary conditions, namely, ambient temperature of the storage room, convection heat transfer coefficient, and emissivity for radiation heat transfer.

The ambient room temperature prior to and after the fire event is conservatively considered to be the maximum theoretical ambient of 58.3°C (137°F) which could occur during a facility loss of ventilation occurrence. Furthermore, this ambient provides the initial steady-state condition for the 9975 prior to the fire event.

The convective heat transfer boundary conditions at the outer drum surface are natural convection prior to the fire event and after the fire event, and they are forced convection during the fire event. The natural convection heat transfer coefficients for initial accident storage conditions are obtained from COMSOL. The different surface orientations and corresponding correlations are also provided in Table 2. For the fire accident scenario, the convection heat transfer coefficients are based on a forced flow at a velocity of 20 m/s on the entire outer surfaces of the drum. The typical air velocity in the storage area is significantly lower than 20 m/s which is used in existing internal analyses of records and maintained in this analysis to add conservatism.

It should be noted that the convection heat transfer inside all the shipping package gaps including, the PCV and SCV, are excluded. These gaps are relatively small so this simplification is expected to have minimal effects on the computed temperatures. Conduction and radiation heat transfer is considered across these gaps. The gas within these gaps in the 9975 package is considered to be a non-participating media for the purpose of radiative heat transfer. Radiation heat transfer between the drum outer surface and ambient is included prior, during, and after the fire.

Radiation view factors were calculated for each gap based on the surface emissivity values of the package components as provided in Table 3. These values are consistent with those used in the 9975 SARP.

Table 3 - Surface Emissivity Values of Package Components

| Component | Emissivity |
|---|---|
| Aluminum plates, honeycomb | 0.20 |
| Fiberboard (Celotex) | 0.50 |
| Lead/ Lead shield stainless steel liner | 0.28 |
| Drum Inner Surface | 0.80 |
| Drum Outer Surface (Initial pre-heatup) | 0.21 |
| Drum Outer Surface (Heat-up) | 0.90 |
| Drum Outer Surface (Cool-down) | 0.80 |
| 3013 Outer Can | 0.30 |
| Rocky Flats Inner Can | 0.30 |
| Rocky Flats Convenience Can | 0.20 |
| Pu Oxide | 0.90 |
| PCV/SCV | 0.30 |

### Material Properties

The fiberboard surveillance results indicate that the material properties density and thermal conductivity decrease with age. In order to determine the rate of aging, the fiberboard temperature during storage needs to be determined. For simplicity and conservatism, the maximum fiberboard temperature is determined for a 19-Watt content and it is applied to the entire fiberboard in the 9975 package. During normal storage conditions the ambient temperature ranges from approximately 35°C (60°F) to 40°C (104°F). A bounding average ambient temperature of 35°C (95°F) is used to determine this maximum fiberboard temperature at the beginning-of-life. This temperature, 63.3°C (146°F), is used to determine the degradation rate which is about 0.3% per year based on work internally published at the Savannah River National Lab at SRS. Therefore, after 20 years the thermal property degradation is 6%. For comparison, the degradation at 51.7°C (125°F) is essentially negligible while at 85°C (185°F) it is approximately 0.5% per year. Note that given the relatively low amount of total degradation, a linear application of the degradation rate is reasonable over the 20 year duration.

## COMPUTATIONAL ANALYSES

The analytical conditions for pre-fire, fire, smoldering, and post-fire analyses are described below. These steps were performed for both beginning of life and degraded fiberboard properties.

The pre-fire analysis consists of a steady-state conservative off-normal storage scenario where facility ventilation has been lost.

- The drum is in its normal storage upright position.
- The bottom surface is adiabatic.
- There is radiation heat transfer from the sides and top of the drum to the ambient.
- There is natural convection heat transfer from the drum sides and top to the ambient.
- The ambient temperature is 58.3°C (137ºF) (no insolation).
- The heat generation is 19 watts.

The fire duration is 1.5 hours.

- The fire is 815.6°C (1500°F) and lasts 1.5 hours.
- The drum is in an upright position.
- There is thermal radiation heat transfer from all the sides of the drum to the ambient (fully engulfed).
- There is forced convection heat transfer from all sides of the drum. The convection coefficients based on 20m/s air velocity are:
    a. 33.5 W/m$^2$-K (5.9 Btu/hr-ft$^2$- ºF) for the top and bottom of the drum.
    b. 17.0 W/m$^2$-K (3.0 Btu/hr-ft$^2$- ºF) for the side of the drum.
- Char properties used when fiberboard reaches 403.9°C (759ºF).
- The content total decay power is 19 watts.

The smoldering phase starts at the end of the 1.5-hour long fire.

Postulated Side Drop (worst orientation)

- The drum is in a horizontal position (suspended).
- There is radiation heat transfer from all sides of the drum to the ambient.
- There is natural convection heat transfer from the bottom and sides of the drum to the ambient.
- The ambient temperature is 58.3°C (137ºF).
- Char properties used when fiberboard reaches 403.9°C (759ºF).
- The content total decay power is 19 watts.
- The smoldering duration is 1 hr. which is reasonable based on literature for wood material [7].
- A postulated 12 inch tear is assumed in the middle of the drum surface. One inch of charred fiberboard at the tear site is considered to smolder.
- The char at the 12-inch tear side is set to 760°C (1400ºF). This is conservative since the smoldering temperature for wood material is about 648.9°C (1200°F) as reported in literature [7].

The cooldown begins after the 1 hr. smoldering phase. The cooldown period corresponds to the time at which the temperatures of the PCV/SCV and their O-rings peak.

- The drum is in a horizontal position.
- There is radiation heat transfer from all sides of the drum to the ambient.

- There is natural convection heat transfer from all sides of the drum to the ambient.
- The ambient temperature is 58.3°C (137ºF).
- Char properties used when fiberboard reaches 403.9°C (759°F).
- The contents total decay power is 19 watts.
- Char properties are replaced with fiberboard properties at temperatures < 403.9°C (759°F) are set to fiberboard properties during cooldown. This is conservative since it increases the cooldown time.
- The cooldown phase is analyzed until peak char-depth and temperature are reached. The typical cool down duration

## RESULTS

Both beginning of life and a 20 year degraded conditioned were analyzed for the fire accident. Temperature plots of the entire package assembly and also at the limiting PCV dual O-rings are presented in Figures 3 thru 5. Figures 3 and 4 are the representative contour plots during fire and smoldering fire events. As expected, the results show the transfer of heat from high temperature regions to the low temperature regions. Most of the charring of the fiberboard occurs during the heat-up phase. Additional charring of the fiberboard continued during the cool down phase as well. Figure 5 illustrates the temperatures as a function of time at the PCV dual O-rings for the beginning of life and after 20 years of storage conditions during the entire facility accident scenario. The results clearly show the increase in temperature as the fiberboard degrades overtime. The results also show that the highest temperature increase occurs during the cool down phase.

A summary of the results of the shipping package limiting components for the beginning of life and degraded condition are as follows:

1. For the beginning-of-life shipping packages, the maximum PCV/SCV O-ring temperature is 163.9°C (327°F) which is well below the maximum O-ring temperature limit of 204.4°C (400°F). The final maximum side-char depth is 79.8 mm (3.14 inches). The remaining 43.4 mm (1.71 inches) (~35%) is uncharred fiberboard.

2. After 20 years of storage service, the maximum PCV/SCV O-ring temperature is 186.1°C (367°F) which is below the maximum O-ring temperature limit of 204.4°C (400°F). The final maximum side-char depth is 100.33 mm (3.99 inches). The remaining 21.8 mm (0.86 inches) (~18%) is uncharred fiberboard.

## CONCLUSIONS

Based on the fire evaluation results, the peak temperature of the limiting package component (containment dual O-rings) is well below the O-ring material accident limit of 204.4°C (400°F). Therefore, the 9975 shipping package meets its intended function to provide confinement during initial storage time as well as after 20-year storage service.

## REFERENCES


1. Standard Specification for Insulating Board (Cellulosic Fiber), Structural and Decorative, ASTM C208-95, American Society for Testing and Materials (Reapproved 2001).

2. Parker O-ring Handbook, ORD-5700A, The Parker Seal Group, Parker Hannifin Corporation, Cleveland, OH, (2001), http://www.parker.com/o-ring.

3. *Rules for Construction of Nuclear Power Plant Components*, ASME Boiler and Pressure Code, Section III, Subsection NB, American Society of Mechanical Engineers, New York, NY (2004).

4. Hensel, S. J., et. al., "Hypothetical Accident Condition Thermal Analysis and Testing of A Type B Drum Package", PVP-Vol. 307, ASME 1995 Pressure Vessels and Piping Division Conference, Honolulu, HI.

5. Gupta, N. K., "Thermal Analysis of A 9975 Package In A Facility Fire Accident", PVP2011-57005, ASME 2011 Pressure Vessels and Piping Division Conference, Baltimore, MD.

6.Gupta, N. K., "Pu/SS Eutectic Assessment In 9975 Packaging In A Storage Facility During Extended Fire", PVP2012-78826, ASME 2012 Pressure Vessels and Piping Division Conference, Toronto, Ontario, Canada.

7. Fernando de Souza Costa and David Sandberg, Mathematical Modeling of a Smoldering Log, Combustion and Flame, 139, 227-238, 2004.

Table 2. Natural convective heat transfer coefficient used for the calculations [COMSOL 4.3a]

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

Table 3: Thermal Conductivity

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

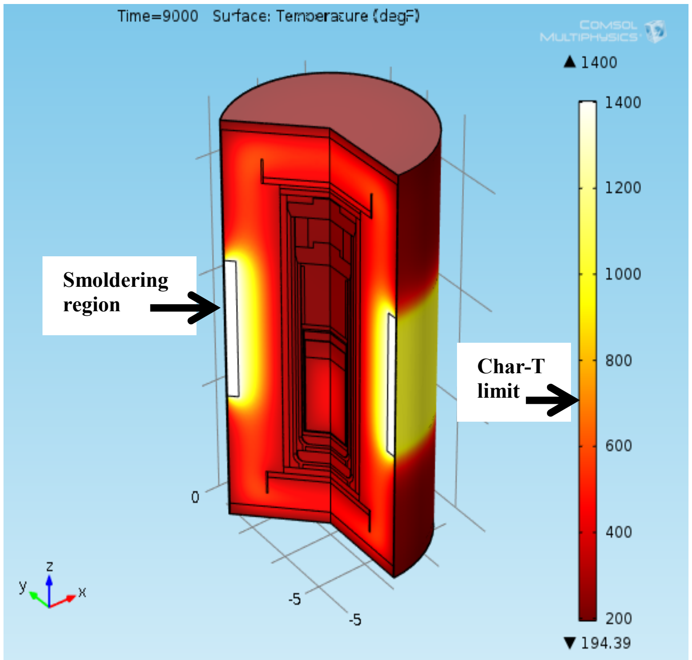


Figure 4: 9975 Shipping Package thermal contour plots at the end of the side smoldering fire accident.

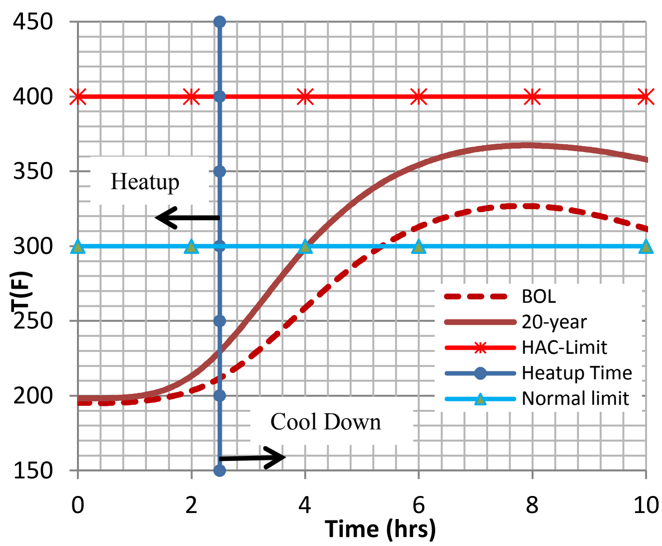


Figure 5: Temperature profiles of the PCV dual O-rings for Beginning-of-Life (BOL) and after 20 years storage service. HAC limit is the hypothetical accident condition limit for the dual O-rings.

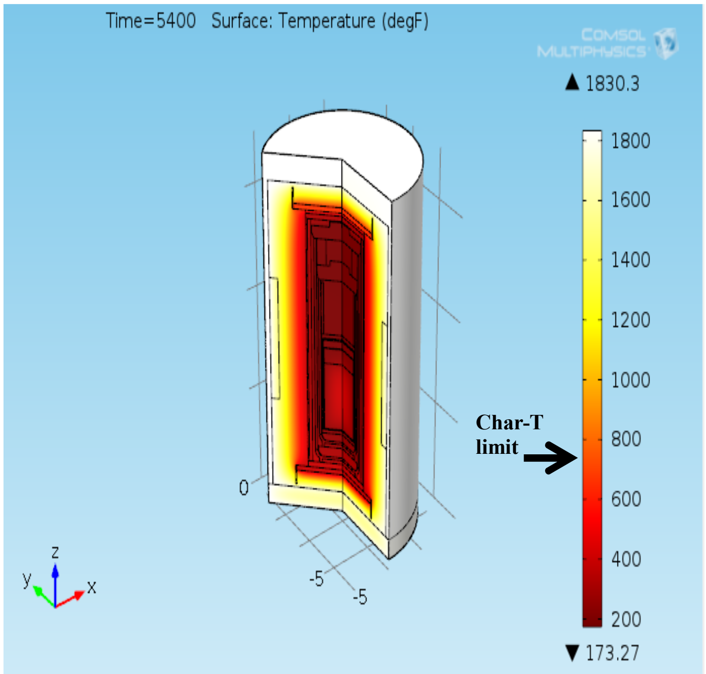


Figure 3: 9975 Shipping Package thermal contour plots at the end of the engulfing fire accident.