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Computer Simulations to Address Pu-Fe Eutectic Issue in 3013 Storage Vessel

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ABSTRACT

On November 22, 2005, the Manager of the Plutonium Finishing Plant (PFP) in Richland, WA issued an Occurrence Report involving a potential Pu-Fe eutectic failure mechanism for the stainless steel (SS) 3013 cans containing plutonium (Pu) metal. Four additional reports addressed nuclear safety concerns about the integrity of stainless steel containers holding plutonium during fire scenarios. The reports expressed a belief that the probability and consequences of container failure due to the formation of a plutonium-iron eutectic alloy had been overlooked. Simplified thermal model to address the Pu-Fe eutectic concerns using axisymmetric model similar to the models used in the 9975 SARP were performed. The model uses Rocky Flats configuration with 2 stacked Pu buttons inside a 3013 assembly. The assembly has an outer can, an inner can, and a convenience can, all stainless steel. The boundary conditions are similar to the regulatory 30 minutes HAC fire analyses. Computer simulations of the HAC fire transients lasting 4 hours of burn time show that the interface between the primary containment vessel and the Pu metal in the 9975 package will not reach Pu-Fe eutectic temperature of 400°C.

INTRODUCTION

Four recent ORPS reports (Ref. 1-4) addressed nuclear safety concerns about the integrity of stainless steel containers holding plutonium during fire scenarios. The reports expressed a belief that the probability and consequences of container failure due to the formation of a plutonium-iron eutectic alloy had been overlooked. As expressed in the two LANL ORPS reports (Ref. 3 and 4):

> " Historic metallurgy work has identified a eutectic effect (an alloy whose melting point is lower than that of any other alloy composed of the same constituents in different proportions) created when plutonium metal is heated in the presence of various other metals including iron. If plutonium metal and steel are heated to a sufficient temperature, but lower than the melting point of plutonium metal or steel, the plutonium reacts with the iron in the steel creating an alloy with a lower melting point than plutonium metal or steel. If enough alloy is generated it can cause the failure of the steel. Acceptable storage containers for plutonium metal include containers made of stainless steel. If the stainless steel containers were stored in areas whose maximum fire temperature exceeded the eutectic temperature of plutonium metal and steel, the containers might fail."

The statement above assumes that during fire scenarios, a plutonium-iron (Pu-Fe) eutectic alloy will form and then fail a stainless steel container by melting or other mechanisms. The phase diagram in Figure 1 shows eutectic melting points of plutonium-iron alloys. Of chief concern is the eutectic melting point for 90% plutonium and 10% iron, at 410°C. Plutonium with 1% gallium forms a eutectic with iron that melts at 400°C. Pure plutonium melts at approximately 640°C.

The temperatures from hypothetical fire scenarios can reach 800°C in a half hour (e.g., following the ASTM E-119 time-temperature fire curve), far above both the eutectic melting point and the melting point of pure plutonium. However, plutonium containers are usually stored within transportation packages (e.g., 9975 packages or 6M drums) with some thermal protection, so the main concerns of the ORPS reports were cases in which containers were out of transportation packages (e.g. for inspection) or accident scenarios where the transportation packages become damaged.

A key question is: Under what conditions will plutonium and the iron in stainless steel form a eutectic? Metallurgical interactions were considered in DOE-STD-3013-2004, but mainly for storage conditions. That standard concluded that plutonium can be stored in stainless steel up to 250°C for 10 years or more without significant eutectic formation. However, currently there seems to be no clear consensus on whether the plutonium-iron eutectic will form significantly at the eutectic temperature (400-410°C), near the melting point of pure plutonium (above 600°C), or at some higher temperature.

Pu metal and Pu bearing materials are stored at Savannah River Site (SRS). The main configurations are 9975 packages and 3013 vessels stored on metal racks. Since SRS has performed thermal analyses for the 9975 package containing 3013 assemblies, the Argonne National Laboratory (ANL), managing the Pu-Eutectic issue, requested the SRS to perform thermal analyses involving facility fires up to 4 hours long and their impact on the Pu container temperatures. 3013 cans stored on metal racks are directly impacted by any room fire. However, the 9975 package contain substantial fiberboard insulating material (Celotex) to protect the 3013 Pu containers from fires.

COMPUTER SIMULATIONS

Mathematical Model

The heat transfer governing equations for unsteady state system in axisymmetric cylindrical coordinates (r,z) are:

$$k_1 \frac{\partial^2 T}{\partial r^2} + \frac{k_1}{r}\frac{\partial T}{\partial r} + k_2 \frac{\partial^2 T}{\partial z^2} + q''' = \rho C_p \frac{\partial T}{\partial t} \qquad (1)$$

Where q''' is the volumetric heat generation by the fissile material per unit time, T is the temperature and k_1 and k_2 are the thermal conductivities of different materials in the radial (r) and axial (z) directions respectively. In general, k_1 and k_2 are functions of r, z, and T. ρ is the density and Cp is the specific heat of various materials. ρ is constant and Cp could vary with temperature for certain materials.

The boundary conditions are:

- Heat transfer coefficient (h) at the drum outer surface,
- Emissivity (ε) at the drum outer and inner surfaces and at other radiating surfaces inside the package, and
- Ambient temperature T_a = 1475°F.

The partial differential equation (1) is non-linear and is solved by numerical methods using MSC PATRAN Thermal solver [6].

Geometry and Contents

The 3013 assembly consists of a site specific stainless steel can which is stored inside a welded 3013 inner can. The inner can is in turn stored inside a welded 3013 outer can. The site specific can contains the payload of Pu metal with a maximum of 19 watts. The Pu buttons are in direct contact with the can metal surfaces forming a potential Pu/Fe eutectic interface. Rocky Flats configuration contains two Pu buttons that could be stacked in one can or the buttons could be stored in separate cans. The configuration with the stacked buttons will yield higher local temperatures and, therefore, this is the configuration that is analyzed in this paper. This configuration was analyzed in the certification of 9975 package and the supporting thermal analyses are documented in Reference 6. The 3013 dimensions are taken from the Rocky Flats configuration in Reference 6 and are also used in the 3013 assembly model on the metal rack.

Material Properties

The thermal properties are identical to the 9975 package 30 minutes HAC fire analyses [6]. The fiberboard properties for the 30 minutes burn duration are derived from benchmarking performed for the 9975 package. Since charred fiberboard is removed for each successive 30 minutes burn (discussed below), charred properties that were used for the post fire analyses of the 9975, are not used here in this investigation. Different materials inside the 9975 package and inside the 3013 assembly are shown in Figures 2 and 3.

Thermal Models

Two thermal models are prepared to analyze the 3013 storage configurations. In the first model the 3013 assembly is stored on a metal rack and is exposed to the fire conditions directly. The second model is the 9975 configuration with Rocky Flats configuration. In the 9975 configuration, there is good amount of insulation between the fire and the 3013 assembly. As mentioned earlier, since the fire test data for the 9975 storage configuration were benchmarked for 30 minutes fires only, an HAC model for the 9975 package could not be used for 4 hour burn. In the absence of real test data for the fiberboard for such a long burn time, a reasonable simple thermal model was agreed upon with the Argonne National Laboratory (ANL) in which charred Celotex (≥500ºF) will be removed for each subsequent 30 minute transient run. Therefore, 7 thermal models with Rocky Flats configuration each containing 2 stacked Pu buttons were developed to study 60 minutes to 240 minutes long fires. Only one model was required to analyze the 3013 assembly on the metal rack.

During HAC testing of 9975, a 30 minutes fire test showed that up to 2 cm (0.8 inches) of fiberboard was lost at some locations around the drum periphery and an average of 1.4 inches of fiberboard was found to be charred. The fire tests showed that fiberboard did not ablate but left behind ash that acts as good insulator for the ensuing fire. For this evaluation, the fiberboard layer experiencing a temperature of about 500ºF is considered ash and is removed (conservative approximation) for each subsequent 30 minutes burn step. Therefore, the 30 minutes FE model from 30 minutes to 60 minutes step was modified where 1.4 inches of fiberboard was removed for the 30 minutes burn. Figure 3 shows the color representation of a model for a burn period from 30 minutes to 60 minutes. After each 30 minutes run, temperature profiles were determined. The fiberboard material that experienced temperatures from 450ºF to 500ºF was replaced with air for the next 30 minutes burn. Radiation heat transfer is maintained between the drum and the unburned fiberboard for each 30 minutes burn. The emissivities of the inner surface of the drum and the charred Celotex surface are set to 0.9 to maximize radiant heat exchange between the drum and the unburned Celotex. In addition, no credit is taken for the heat lost in the gases escaping through the vent plugs.

For the first 30 minutes of the fire transient, no air gap exists between the drum inner surface and the fiberboard and, therefore, radiation is ignored between the two surfaces. For the subsequent 30 minute steps, radiation is considered between the drum and the unburned fiberboard surface.

RESULTS

3013 on Metal Rack

Figure 6 shows the temperature contours for the Pu buttons in the 3013 assembly stored on a metal rack for the first 10 minutes. It is found that the Pu-Fe interface reaches well above Pu-Fe eutectic temperature of 752ºF (400ºC) in less than 10 minutes.

3013 in 9975 Packaging

The temperature contours for the whole model for multiple 30 minutes burn steps are shown in Figure 7. The temperature scale range in Figure 7 is from 150ºF to 1475ºF. The temperature contours for the Pu buttons are shown in Figure 8. The temperature scale range in Figure 8 is from 150ºF to 400ºF. These results are summarized in Table 1.

Table 1: 9975 Pu Can Interface Temperatures (ºF) During the Fire Transient

Burn Time (Minutes)	Pu/SS Interface Temperature (ºF)
30	323
60	325
90	329
120	333
150	339
180	347
210	357
240	368

The multiple 30 minutes fire burn simulations show that the Celotex burn depth decreases during successive burn steps. This can be explained by the fact that the additional gas space provides good insulation. The rate of radiant energy absorbed at the drum surface decreases in time with the reduction in Celotex radius and the heat up of the drum wall to the fire temperature.

The analyses showed that the maximum Pu stainless steel interface temperature after 4 hours of 1475ºF (800ºC) fire was only 187ºC, well below the Pu-Fe eutectic temperature of 400ºC. This is due to the fact that the additional gases occupying the removed Celotex are better insulators than the Celotex and help reduce the energy available for burning the intact Celotex. This process is also helped by the reduced radiant heat available to the reduced size Celotex.

Effect of Temperature on Eutectic Formation

For a vulnerable region to exist, the composition in the region must approach the eutectic composition. Intimate contact of metallic iron and plutonium, and sufficient time for diffusion to occur are necessary for this condition. The diffusion process is strongly temperature dependent, and would be accelerated in the event of a fire. For temperatures attained in containers stored in 9975 packagings, negligible diffusion would occur. For a 3013 container stored in rack, a significant region of the eutectic composition could form in a fire lasting over an hour. Times required for diffusion to a depth of 0.1 cm in a 3013 container sitting on a metal rack are given in Table 2 [7].

Table 2. Time to attain Eutectic to a depth of 0.1 cm

Temperature, ºC	Diffusivity (m^2/sec)	Time
150	10^{-18}	Indefinite
350	10^{-10}	1 year
1000	10^{-6}	0.8 hr

CONCLUSIONS

1. For the bare 3013 on the metal racks, the Pu/SS304L interface reaches well above Pu-Fe eutectic temperature of 752ºF (400ºC) in less than 10 minutes in an HAC fire.

2. For the 3013 in the 9975 package, the temperature of the Pu/SS304L interface will reach about 368°F (187°C) after 4 hours of burn in an HAC fire. This temperature is well below the Pu/FE eutectic temperature of 752°F (400°C).
3. Due to lack of Celotex test data for extended burn time (> 30 minutes), the temperature results are approximate at best. Therefore, it is recommended that additional burn tests should be performed to validate the results. However, since there is large safety margin in the simulation results, it is believed that Pu/SS304L interface temperature of 400ºC will not be reached.

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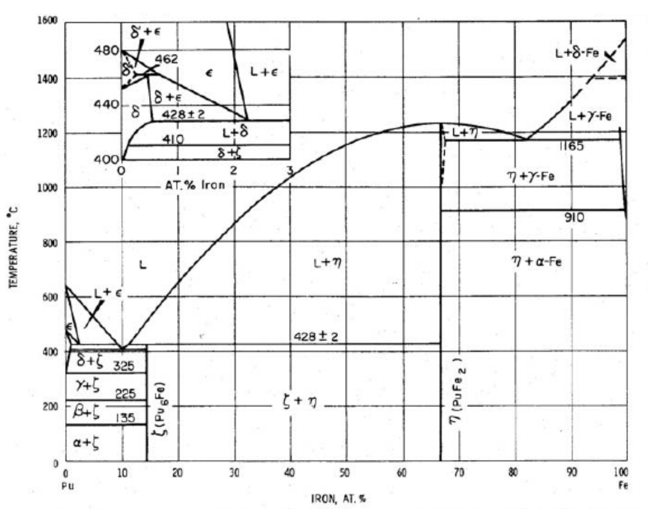

Figure 1 – Pu Phase Diagram

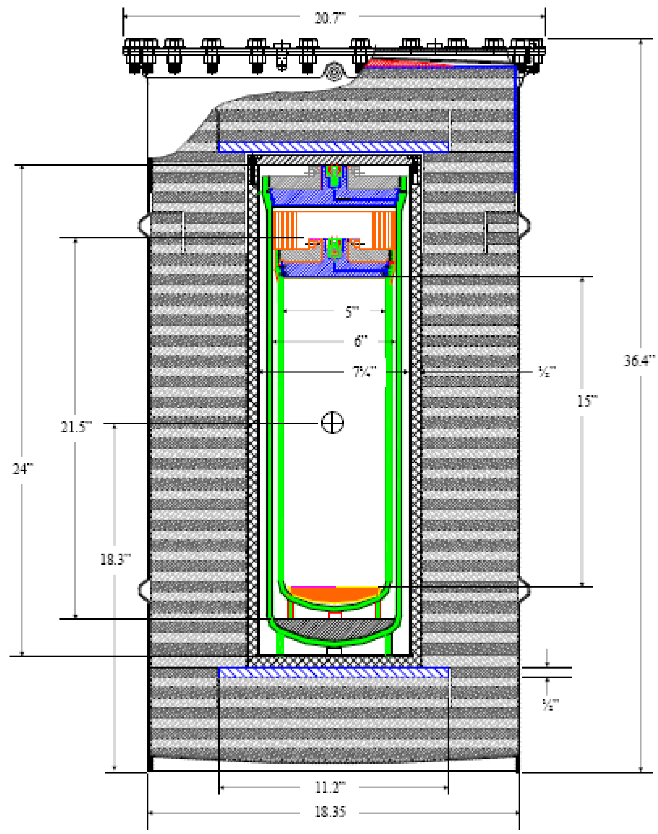

Figure 3 – 9975 Package Schematic

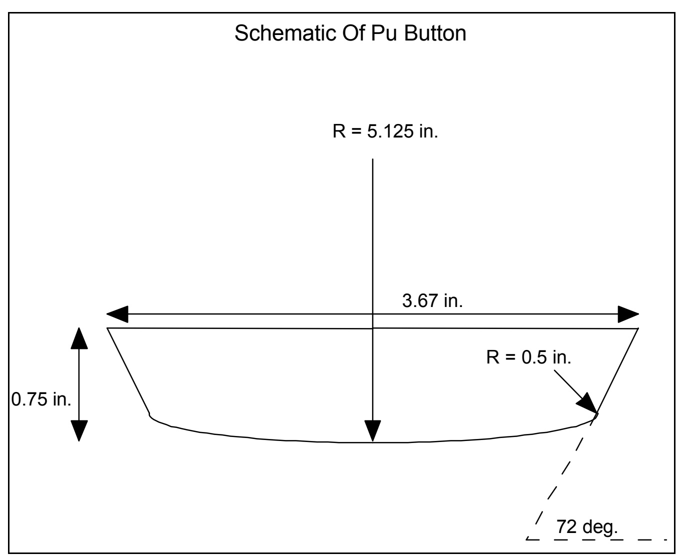

Figure 3 – Pu Button Geometry

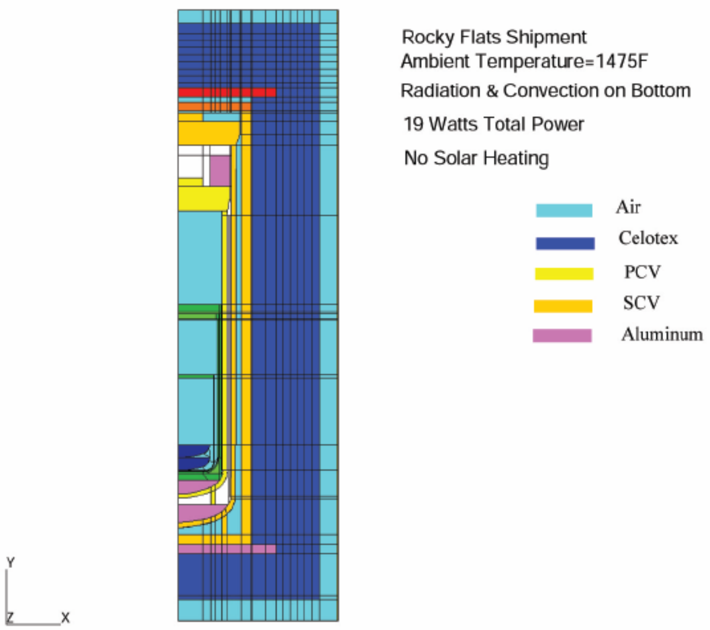

Figure 4 – 9975 Color Representation of Material Locations

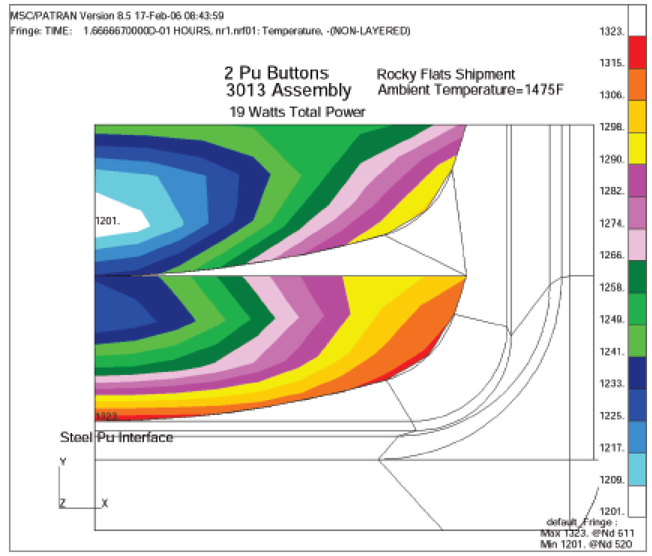

Figure 6 – Temperature Contours of 3013 Assembly on the Rack (10 minutes)

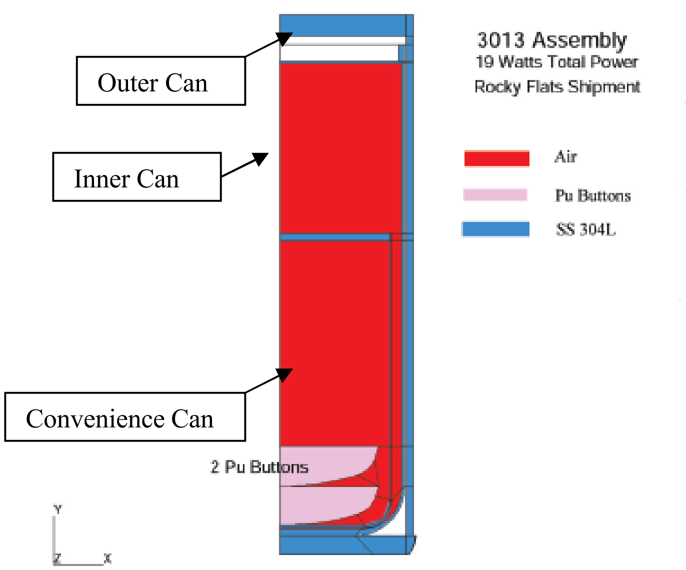

Figure 5 – Color Representation of 3013 on a Metal Rack

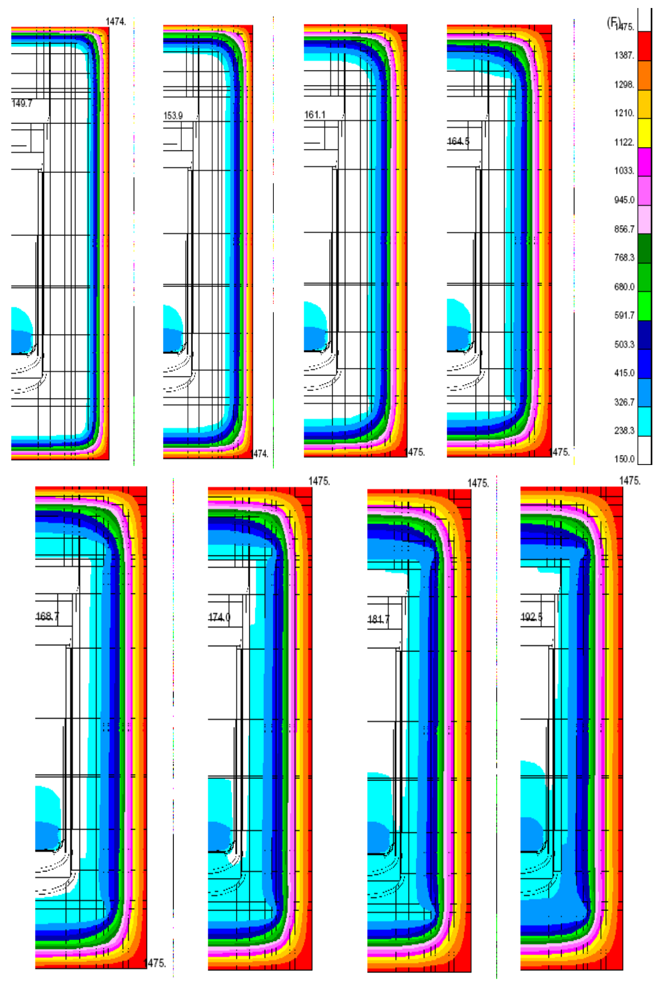

Figure 7 – 9975 Temperature Contours for Burn Time 4 Hours

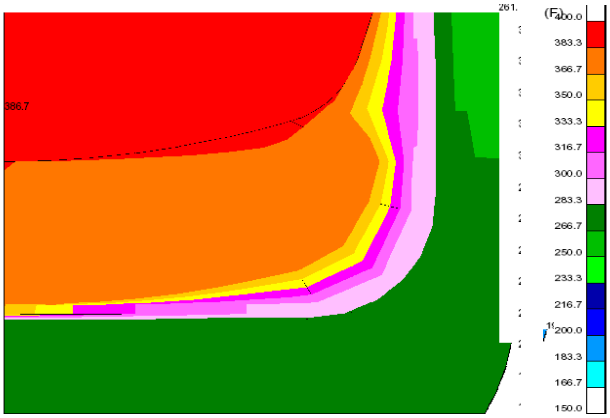

Figure 8 - Temperature Contours for Pu Buttons in 9975 at 240 Minutes (386.7ºF)